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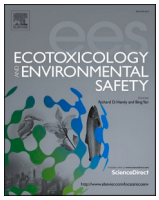

Microplastics in European sea salts – An example of exposure through consumer choice and of interstudy methodological discrepancies

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ABSTRACT

Microplastics are contaminants of emerging concern, not least due to their global presence in marine surface waters. Unsurprisingly, microplastics have been reported in salts harvested from numerous locations. We extracted microplastics from 13 European sea salts through 30% H_2O_2 digestion and filtration over 5-μm filters. Filters were visually inspected at magnifications to x100. A subsample of potential microplastics was subjected to Raman spectroscopy. Particle mass was estimated, and human dose exposure calculated. After blank corrections, median concentrations were 466 ± 152 microplastics kg^{-1} ranging from 74 to 1155 items kg^{-1}. Traditionally harvested salts contained fewer microplastics than most industrially harvested ones (t-test, $p < 0.01$). Approximately 14 μg of microplastics (< 12 particles) may be absorbed by the human body annually, of which a quarter may derive from a consumer choosing sea salt. We reviewed existing studies, showing that targeting different particle sizes and incomplete filtrations hinder interstudy comparison, indicating the importance of method harmonisation for future studies. Excess salt consumption is detrimental to human health; the hazardousness of ingesting microplastics on the other hand has yet to be shown. A portion of microplastics may enter sea salts through production processes rather than source materials.

1. Introduction

The oceans are known sinks for a range of contaminants, such as persistent organic pollutants and non-dissolvable anthropogenic materials such as plastic waste. Any plastic item that finds its way to the marine environment is destined to fragment into smaller particles through chemical and physical forces (Andrady, 2011; Barnes et al., 2009). Once these particles are smaller than 5 mm, they are classed as microplastics – a contaminant of emerging concern (Arthur et al., 2009; Thompson et al., 2004). The lower size limit is often debated, and usually stated as either 100 nm or 1 μm (EFSA, 2016; Hartmann et al., 2019; Nguyen et al., 2019). Microplastics can also be directly released into the environment, including from accidental spillages of pre-production pellets or wastewater releases containing microfibres from washing (Murphy et al., 2016; Napper and Thompson, 2016). Microplastics were first discovered in seawater over four decades ago (Buchanan, 1971; Carpenter and Smith, 1972) and research has accelerated since the publication of Thompson et al. (2004). It is now assumed that surface waters across the globe carry this contaminant (Eriksen et al., 2014; van Sebille et al., 2015).

Seawater is an important commodity. Salts dissolved in seawater are extracted mainly for the chemical industry and other non-food applications (e.g. agriculture and aquaculture) (Cnaani et al., n.d.; EUsalt, 2020; Kubitza, n.d.; Roy et al., 2007; Staurnes and Finstad, 2000). A fraction of global salt production is destined for human consumption; in Europe for example approximately 7% is harvested as food grade salt (EUsalt, 2020). Most of today's global salt demand is covered by mining subterraneous halite deposit: remnants of evaporated vast prehistoric water bodies (Brown et al., 2019). Some of the global salt demand is met through extraction from seawater and other natural brine solutions.

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Approximately 10% of salt produced in Europe is harvested via solar evaporation using waters from the Atlantic Ocean and Mediterranean Sea and their connected water bodies (EUsalt, 2020); France, Greece, Italy and Spain being the main producers (EUsalt, 2020).

In theory, any microscopic or molecular contamination present in marine waters can also be present in salts extracted from those waters. This is because marine waters are diverted from the sea into artificial evaporation ponds, natural lagoons (traditional method) or into other outdoor or indoor evaporation facilities (modern/industrial method). Here, through evaporation driven by (solar) heat and wind more concentrated brine waters remain, eventually leading to crystallisation—where a salt crust forms and any solids are left behind; water is drained and the salt crust harvested (EUsalt, 2020). Indeed, heavy metals, organic pollutants and microplastics have been reported in sea salt for human consumption (Cheraghali et al., 2010; Serrano et al., 2011; Yang et al., 2015). However, microplastics have also been found salts from other brine water bodies and in rock salts (Iñiguez et al., 2017; Schymanski et al., 2020; Yang et al., 2015).

The presence of microplastics in food-grade salts is likely to lead to human exposure. Concerns about possible impacts on human health have been raised (Peixoto et al., 2019). Potential exposure routes include passing of small particles (< 150 μm) through the human gastric tissue (Wright and Kelly, 2017). However, to date published research on microplastics in the human body is limited. The presence of microplastic in stool and colon samples (Ibrahim et al., 2020; Schwabl et al., 2019; Yan et al., 2020) shows that microplastics are ingested, but also that bodily mechanisms exist to remove at least a proportion of those particles again. Translocation into the human body seems possible; Ragusa et al. (2021) report spheroids and irregular microplastics 5–10 μm in size in human placental tissue. However, Braun et al. (2021) caution that high likelihood of microplastic contamination during the sampling in a labour & delivery setting warrants further work to confirm such findings. Leslie et al. (2022), using very stringent quality control measures to mitigate potential microplastic contamination of samples, recently identified microplastics in human blood samples. To date no evidence for health impacts exists, but the research area of quantifying the hazardousness of microplastics and therefore also risk determination is still in its infancy. Ingestion exposure to contaminants is generally calculated using ingestion exposure dose equations, which amongst other variables takes into account the exposure dose, contamination concentration, consumption or intake rate of the contaminated medium but also further variables such as the contaminant's bioavailability, duration and frequency of contact with the contaminant and is usually expressed as per body weight (ATSDR, 2005). However, such detailed knowledge about microplastics is lacking. To date, numerous studies establish exposure simply based on microplastic abundance in salt and daily recommended salt intake or annual salt consumption (Lee et al., 2019; Peixoto et al., 2019; Renzi et al., 2019), or of a range of different food products (Ageel et al., 2022; Jin et al., 2021; Rubio-Armendáriz et al., 2022).

A plethora of different methods to extract microplastics from various matrices of interest are used, often criticised as the root of preventing interstudy comparability (Hermsen et al., 2018; Hidalgo-Ruz et al., 2012). When it comes to microplastic abundance across regions, research focusing on individual countries is less powerful than individual studies investigating microplastics in salts across countries—even if analytical limitations exist as the degree of uncertainty around the results should be similar. However, method harmonisation is imperative for interstudy comparability. Many studies set out to investigate the most suitable method for a given matrix, sometimes employing dosing experiments to analyse the recovery potential of different extraction methods (e.g. Catarino et al., 2017; Karami et al., 2017a; Thiele et al., 2019 for marine biota). The most suitable extraction method for microplastics from salts has not been established yet. As pointed out by Kim et al. (2018) however, not only applying different extraction techniques but also identification methods could cause discrepancies in results hindering comparability. Despite the existence of numerous reviews of work surrounding microplastics in food-grade salts—including the assessment of different method steps used (Lee et al., 2019; Peixoto et al., 2019; Zhang et al., 2020), issues regarding method variations are seldomly addressed.

There were three aims to this work: Firstly to assess sea salt of an entire geographic region (Europe). When we began this work in 2016, a single study on microplastics in food-grade salts had been published by Yang et al. (2015), whose methods were closely followed to enable result comparison. As more studies have been published since, the second aim was to assess methodological issues impeding interstudy comparability by reviewing published work and standardising results using a pseudo-harmonisation attempt. The third aim was to calculate detailed microplastic exposure rates based on consumer choice and particle size and mass.

2. Methods

2.1. Origin of samples and production method

Sea salt was sourced from European supermarkets in the summer of 2016. Thirteen packages (180–1000 g) from different locations in seven European countries were used. Based on ethical and commercial considerations, neither the brand/supplier nor the exact location are provided here. Three products were from northern and southern areas of the North Atlantic and the Eastern Mediterranean, four products came from the Western Mediterranean area (supplementary information online, SI Fig. 1). A questionnaire survey was sent to each producer to gather information on production methods, but only one response was received so it is not included here. In addition, a desk study was performed at the time of sample procurement to establish the production method of each product, including equipment materials if possible. For this, information on the salt packages, web information by the producer, and Google Earth was used. Samples were categorised as industrial or traditional harvesting technique.

2.2. Particle extraction from samples

The method of Yang et al. (2015) was followed with minor adaptations to allow for interstudy comparability. Briefly, with two replicates of each sample, 20 g of salt were weighed onto aluminium foil and transferred into glass bottles using a paper funnel. Approximately 20 ml of pre-filtered H_2O_2 (30% in water, Fisher Bioreagents) was added before sealing the bottle with a glass stopper. Risk assessment compliance required this step to take place in a negative-pressure fume hood. Sample bottles were placed into an oven at 60 °C for 24 h for optimum digestion of organic materials. Then, 160 ml of pre-filtered H_2O was added, each bottle shaken manually until the digestate was fully dissolved. The bottles were transferred to an oscillation incubator (80 rpm, 50 °C) for 48 h and subsequently left to settle at room temperature. Lastly, sample supernatants were vacuum filtered using 5-μm cellulose nitrate (CN) filters (Whatman, 47 mm diameter) - after Yang et al. (2015); this size is around the lower limit that can be detected and characterised with the methods used. To enable filtration of the precipitate, which had not been processed by Yang et al. (2015), those remnants were resuspended with additional pre-filtered H_2O and poured over a second CN filter. This process was repeated until no visible traces of the precipitate remained in the sample bottle. Plastic Petri dishes and lids were lined with aluminium foil, filters were secured onto the bottom liner with small amounts of glue (Pritt Stick, mainly consisting of natural ingredients). Dishes were sealed with elastic bands and stored in darkness.

2.3. Contamination mitigation and control

Steps were undertaken to avoid contamination of samples with microplastics. Salt packages were stored in a sealed, air-tight container.

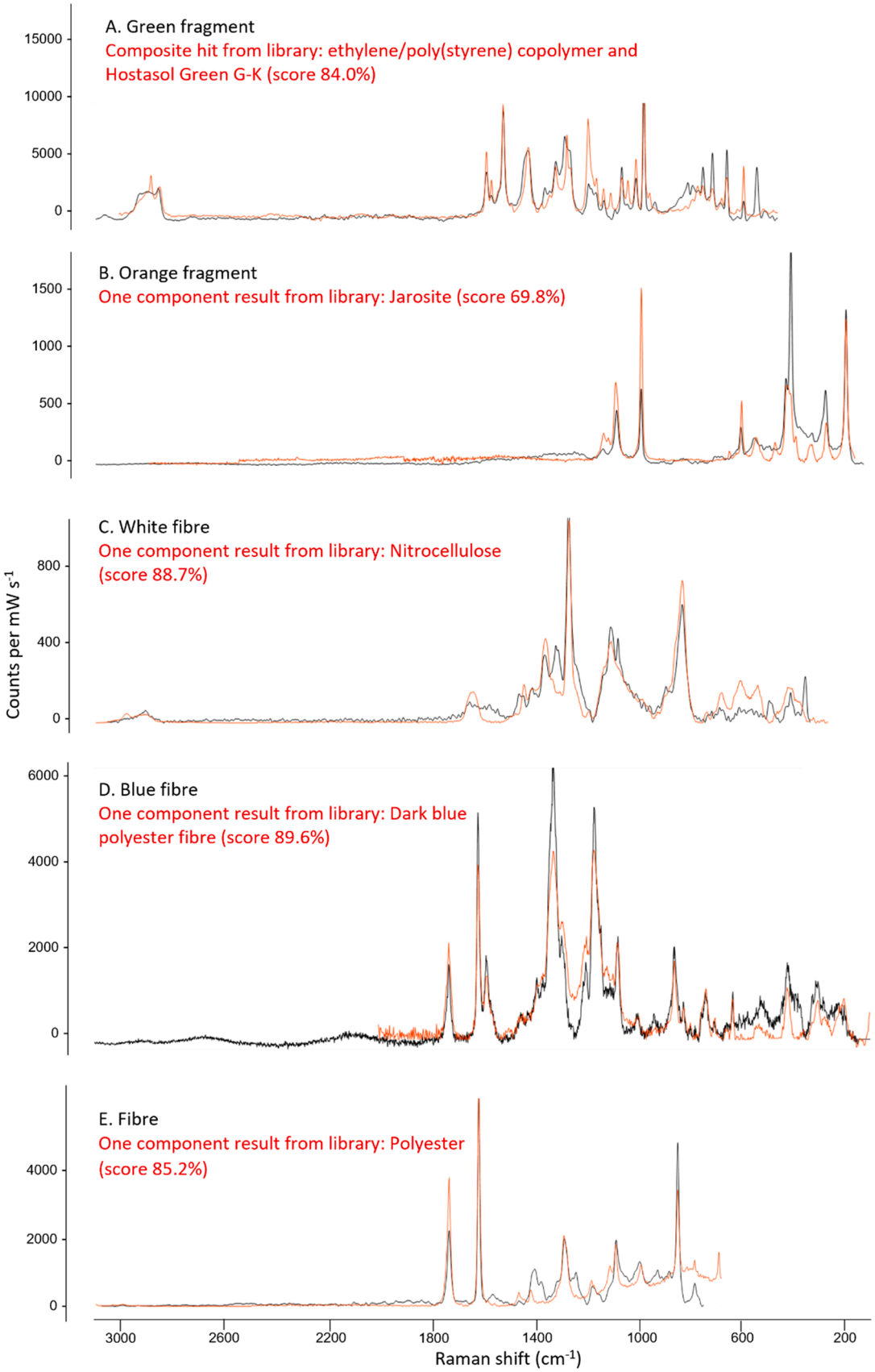

Fig. 1. Raman spectra of five plastic and mineral particles (A-E) found in sea salt samples (black) and their respective selected library hits (red), including match scores.

Extractions and enumeration were performed in a clean, low plastic laboratory environment with low footfall. Based on Browne et al. (2011), 100% cotton laboratory clothing was worn to mitigate potential plastic microfibre contamination. All reagents, including deionised water for equipment rinsing, were filtered through 5-μm CN filters. Prior to use, all glassware was placed into a 10% hydrochloric acid bath for 24 h. In between sample extractions, glassware was rinsed three times with pre-filtered water. All cleaned and in-use glassware was covered with aluminium foil when not handled as suggested by Dris et al. (2016). A procedural filter blank was created during each sample batch and analysed alongside the samples, to enumerate potential contamination that could have been introduced during the extraction process.

2.4. Quantification of potential microplastics

Filters were inspected under an optical light microscope (Olympus BH2 with attached Nikon D5000 digital camera) at magnification x4 and x10 with x10 eye pieces. Any greater magnification would have led to contact between particles and lens potentially disturbing the former. Particles were suspected to be microplastic if they lacked visible cellular or organic structure, were either transparent of homogeneous colour, but potentially patterned or striped, and in case of fibres if their diameter appeared constant throughout, but potentially with frayed or split ends (Hidalgo-Ruz et al., 2012; MERI, 2017). Approximately 1/3 of each filter was systematically analysed (Supplementary information online, Section 2) due to high particle loads as suggested by (Schymanski et al., 2020).

Counts from supernatant and precipitate filters per sample were combined. Blank corrections were performed. Limits of detection (LOD) were calculated for each type and colour of potential microplastic to account for different sources of airborne contamination (Equation 1) and subtracted from counts (Macdougall et al., 1980).

$$LOD_i = 6SD_{Blanks_i}$$

Equation 1 – Blank correction of sea salt samples using the limit of detection based on 3x standard deviation of potential microplastic counts in procedural blanks per category. Category here refers to each individual particle type of each colour and is expressed as *i*. Values are further multiplied by two since salt samples consist of two filters, i.e. one each for the supernatant and one for the precipitate filtrate.

2.5. Raman spectroscopy

Polymer identification was performed with a Raman laser spectrometer (785 nm Renishaw inVia, WiRE 4.1 software). While a 613 nm Raman spectrometer was also available, 785 nm frequency was chosen to limit fluorescence (Karami et al., 2017a). Potential microplastics were manually located again with an integrated Leica DM 2500 M microscope. Spectra were obtained with a 50x magnification lens, power setting 0.1 – 5%, one acquisition and exposure time of 10 s over the entire spectral range. For each particle, laser power was initially set to 1% to avoid burning or melting of the particle. From salt samples, 6% of potential microplastics and 10% from blanks were assessed representing the most common visually counted particles (for details see Supplementary information online, Section 3 incl. SI Table 1). Spectral baselines were adjusted, cosmic rays removed if present and all spectra were smoothened using the WiRE software's default setting (Savitsky-Golay filter, smooth window 9, polynomial order 3). Artificial peaks from partial fluorescence or oversaturation were removed by truncating spectra. Further, the library software automatically applied optimised corrections during spectral match searches.

Spectral library searching was undertaken with BioRad KnowItAll® Informatics System - Raman ID Expert (2016) software using the software's integrated library but also a purpose-built one by the author and a freely available Raman library (Munno et al., 2020). After Yang et al. (2015), a match score of $\geq$ 60% was accepted. However, instead of automatically accepting a score $\geq$ 70% and manually assessing the result for lower hits, all results were visually assessed as a matter of good practice in spectroscopy (Horton et al., 2017; Smith, 2011).

Table 1
Manufacturing method and equipment used for harvest/manufacture of sea salt products based on packaging information, web information by the producer and Google Earth.

Sample ID	Salt making	Further equipment information if available
North Atlantic connected products		
N1	industrial	plastic equipment
N2	unknown (but could be industrial due to climate)	
N3	industrial	water used is mix of fresh but also effluent from washing seaweed, some plastic equipment
S1	industrial	equipment looks metallic in appearance
S2	industrial	metallic trays, very clinical process
S3 *	industrial	filtered through mussel bed and then charcoal, rinsed in brine and stored in plastic trays
Mediterranean Sea products		
W1	traditional	appear to use natural materials
W2	unknown (but could be traditional due to long history of salt flats in the area where the product is from)	
W3	traditional	
W4	traditional	not much known: untreated, unrefined and unwashed
E1	traditional	
E2	unknown (but could be traditional due to presence of large salt flats at that location on Google Earth)	
E3	traditional	

* Microplastic concentration results excluded as an outlier

2.6. Data presentation and statistical analysis

The final number of microplastics was calculated based on the proportions of particle categories verified to be plastic; if a particle category was not assessed those particles were excluded (Horton et al., 2017). For example, one 'green angular' was tested and found to be plastic, hence all 'green angular' were assumed to be plastic; none of the 'black other' were plastic, hence all 'black other' were excluded (supplementary information online, SI Table 1). To extrapolate to particles per kg of sea salt a conversion factor was applied (supplementary information online, Section 2). Mean values of duplicates were reported. However, for regional and production-specific statistics, the median was reported using all individual sample values (supernatant and precipitate counts always already combined) since it is more robust to outliers, which is preferred for reporting contamination. The variation between duplicates and geographic/production-specific statistics was measured with one standard deviation. As an estimate for the true mean of the sample population, confidence intervals at 95% were calculated. Relative standard deviations were calculated to provide a measure of uncertainty for comparative purposes. Samples were compared with an unpaired t-test. The variance of the two groups was assessed with an f-test. In case of unequal variance, a type 3 t-test in Excel was employed (Dytham, 2009).

Particle sizes were established from photographs taken of potential microplastics after visual counting by measuring their largest dimensions and the longest dimension perpendicular to the former using 'ImageJ' (Schneider et al., 2012). Particles were described by size class ($<$ 150, 150–499, 500 – 1000 and $>$ 1000 μm). The smallest size class ($<$ 150 μm) was divided further using 10-μm bins starting at 5 μm. Fibres were categorised by their diameter. Kernel density estimation for

histogram data was performed using an Excel Add-In (Thompson, 2006) to assess the size distribution.

2.7. Review of existing studies and comparison with current study

The following databases were searched on 08/07/2022 using the terms "micro$plastic* AND salt": Web of Science, PubMed, Directory of Open Access Journals, Bielefeld Academic Search Engine, PLOS ONE, Wiley Online Library, Springer Link and JSTOR. Reference lists of extracted studies were examined for additional studies. No exclusion criteria were applied to obtain an overview of the following information to uncover possible methodological discrepancies:

- Region/country
- Extraction technique information: complete sample or density separation,
  - smallest particle assessed/found,
- Spectroscopy analysis information:
  - % potential microplastics subjected to spectroscopy,
  - % plastic confirmation rate,
  - library search minimum match score,
- Contamination control information:
  - filtering of reagents,
  - clean environment,
  - airborne controls,
  - procedural blanks,
  - results adjusted for findings in blank samples,
- Results:
  - microplastic concentration,
  - statistic used (i.e. median, mean etc.),
  - particle categories found.

Results:

To evaluate if differences in analytical methods could explain differences in results between this study and other European sea salt samples a rudimentary approach of pseudo-harmonisation was performed. Only studies investigating European sea salts were included to avoid wider geography and type of salt as confounding factors. Many of the methodological differences may be difficult to quantify, e.g. clean vs normal laboratory conditions, use of air controls, thresholds of match scores. Therefore, easily adjustable parameters for interstudy comparability were chosen, namely target particle size, application of density separation (assessment of entire sample or supernatant only), focus on single particle type (i.e. fibres) and lack of polymer confirmation through spectroscopy. Values were adjusted proportionally as follows based on the findings from this study (Table 5) as an assumed common denominator.

- Minimum target particle size: when particle assessment was based on larger microplastics (e.g. $\geq$ 100 or 150 μm), the study's microplastic concentrations were adjusted proportionally compared to our counts between said target particle size and our minimum size. In our study 36.2% of particles were $<$ 30 μm, 55.7% $<$ 50 μm, 61.4% $<$ 60 μm, 75.7% of particles $<$ 100 μm, 86.2% $<$ 150 μm and 13.8% $\geq$ 150 μm.
- Entire sample or particle extractions from supernatant only: when only supernatants were assessed, it was assumed that only 27.3% of particles were extracted from samples.
- Particle class: When only microfibres were reported, it was assumed that other particles (24.4%) were ignored during the identification process.
- Anthropogenic particle adjustment: when polymer composition was not assessed, a polymer confirmation rate of 29.6% was applied.

2.8. Calculation of human dose exposure

Our calculations of human exposure to microplastics from sea salt ingestion are based on consumer choice, microplastic particle size, intestinal absorption rates and particle mass. Since European salts are most likely being consumed in Europe, European salt consumption values were used for exposure calculations. Further, we assumed that sea salt is only consumed when added by the end consumer and salt outside the consumer's choice would be rock salt, mainly because it is generally cheaper than sea salt. While 70–75% of salt intake in a Western diet comes from processed foods, the end consumer adds approximately 10–15% of the overall salt intake (Sanchez-Castillo et al., 1987). These proportions likely differ in other regions. In Japan and China, for example, most salt is added at home; but there, soy sauce is an important source of sodium (Brown et al., 2009), which in turn may be high in added salt. The average daily dietary salt intake of European adults is 8–11 g day^{-1} (EFSA, 2006), suggesting that only $<$ 2 g come from store bought salt of their choice (ingestion rates). Secondly, for the contaminant concentration only microplastics $\leq$ 150 μm were considered based on their potential hazardousness of being able to pass the gut tissue barriers (Welle and Franz, 2018; Wright and Kelly, 2017). The particle dosage was calculated by using the concentrations of microplastics found in sea salt in this study. Data from Yang et al. (2015) was used to estimate particle dosage from rock salt (for workings see appendix). Lastly, the lack of knowledge about microplastic toxicity, bioavailability, translocation and even egestion rates in humans complicates the assessment of human dose exposure to microplastics. Therefore, two scenarios were assumed: An unlikely maximum upper exposure scenario of 100% of small particles to pass the gut tissue barriers and an intestinal absorption rate of 1% as a more realistic exposure scenario (Welle and Franz, 2018).

In addition, since toxicological studies are based on mass by mass rather than items per mass values, the mass of microplastics was estimated. As before, focus was on particles $<$ 150 μm. First, the volume of potential microplastics previously photographed was established: fibre volumes were established using Equation 2 (Hermabessiere et al., 2018), where D is the fibre diameter, L the length of a fibre. For fragment volumes the approach by Hermabessiere et al. (2018) was adapted according to Simon et al. (2018) by assuming the thickness of the particle to be 67% rather than 100% of the minor dimension (Equation 3), where S is the longest axis perpendicular to the largest dimension (L).

$$V_{fib} = (D/2)^2 * \pi * L$$

Equation 2 – Fibre volume (V_{fib})

$$V_{frag} = S * 0.67S * L$$

Equation 3 – Fragment volume (V_{frag}).

Then, the mean volume per particle type was established. This was done to obtain a general estimate per type due to the limited size information available for individual particles confirmed to be microplastics. To obtain the estimated particle mass, the volume was multiplied by the polymer density. The mean particle mass per particle type was calculated (see supplementary information online, Section 2) and applied to the number of microplastics $<$ 150 μm found.

3 Results

3.1. Assessment of European sea salt

3.1.1. General product information

Thirteen salt samples were acquired in European supermarkets; all came in plastic packaging (supplementary information online, SI Table 2). None of the packaging polymer types were identified in respective samples during Raman spectroscopy. Based on a desk study, sea salts coming from waters connected to the North Atlantic (north of

Table 2
Publications reviewing microplastics in food-grade salt, showing if salt was the only food item assessed, if microplastic ingestion rates were calculated and if extraction methods were assessed. For the latter, it was reviewed if particle/filtration sizes, contamination control measures and extraction methods of the original research were assessed. Literature search conducted 08/07/2022.

Review	Salt only?	Calculation of microplastic ingestion rate?	Method assessment?	Do they take into account		
				Particle/ filtration size?	Contamination control?	Extraction method?
Peixoto et al. (2019)	Yes	Yes	Some	Yes	No	No
Lee et al. (2019)*	Yes	Abundance in products only	Some	Yes	No	No
Danopoulos et al. (2020)	Yes	Yes	Some	Yes	No	No
Lee et al. (2021)	Yes	Yes	Yes	Yes	Partly (mainly quality control)	Partly
Kim and Song (2021)	Yes	Yes	Some	Yes	No	No
Cox et al. (2019)	No	Yes	No	No	No	No
Zhang et al. (2020)	No	Abundance in products only	Yes	Yes	No	Yes
Shopova et al. (2020)	No	Yes	No	No	No	No
Kwon et al. (2020)	No	Abundance in products only	Yes	Yes	No	Yes
Myszograj (2020)	No	Yes	No	No	No	No
Senathirajah et al. (2021)	No	Yes	No	No	No	No
Razeghi et al. (2021)	No	n/a (only one study had looked at salt in Iran with no microplastics found)	Some	No	No	Yes
Mortensen et al. (2021)	No	Yes	Yes	Yes	No	General for all matrices
Jin et al. (2021)	No	Yes	Yes	No	No	No
Ageel et al. (2022)	No	Yes	No	No	No	No
Rubio-Armendáriz et al. (2022)	No	Yes	No	No	No	No

* includes original research

approximately 47°N) were harvested using industrial methods and south of approximately 47°N (here, in or close to the Mediterranean Sea) harvesting using traditional methods (Table 1).

3.1.2. *Microplastics in sea salt samples*

Procedural blanks (n = 5) contained mean concentrations of 9.2 (± 3.2) potential microplastics filter^{-1}. Of those, 84.8% were fibres, 13.0% spheroids and 2.2% fragments. The colour of 87.0% was transparent/clear, 8.7% were blue and 4.3% black. Therefore, the LOD for black spheroids was 13, and three for blue and black fibres and particles of the category 'other'/'other'. Sample concentrations were adjusted accordingly. Transparent fibres were excluded entirely from the results.

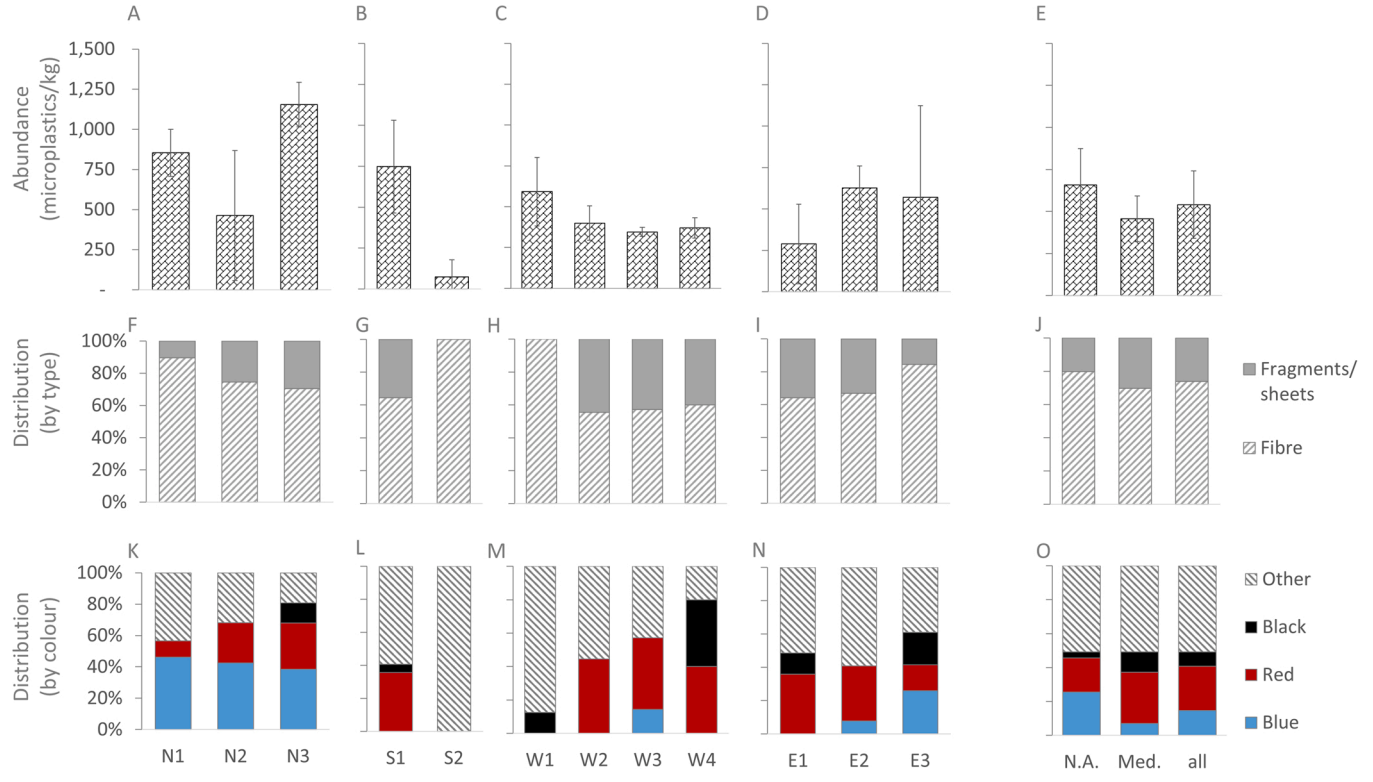

Fig. 2. Microplastics found in sea salt samples from 12 locations (see supplementary information online, SI Fig. 1 for geographic subregions). A-E show mean abundances of microplastics kg^{-1}, graphs F-J show proportions of types of microplastics (%) and K-O show colours of microplastics (%). Sample codes: N = northern area of North Atlantic, S = southern area of North Atlantic, W = western area of Mediterranean, E = Eastern Mediterranean, N. A. = North Atlantic means, Med. = Mediterranean means. Error bars = 1x standard deviation. Sample S3 was excluded as an outlier.

This was done due to the consistently high presence in blanks. Overall, 27.3% of potential microplastics were found in the supernatant and the remainder in the precipitates of the salt samples.

Raman spectroscopy confirmed microplastic abundance in sea salt ranging 74 (± 105) microplastics kg^{-1} (no microplastics detected in one duplicate) to 1155 (± 140) microplastics kg^{-1} (Fig. 2A-D) per package with a median of 466 (mean 540 ± 152) microplastics kg^{-1} (Fig. 2E). Mean microplastic mass estimate was 4.51 ± 6.74 μg kg^{-1}. Detailed mass calculations can be found in the supplementary information online, section 8. Sample S3 contained 10.3x as many microplastics than the mean of the remaining samples and was therefore excluded from the analysis as a potentially contaminated outlier. Microplastics were confirmed to be mainly rayon, polypropylene, polyester and polyethylene, but also nitrocellulose and copolymer (either ethylene/polystyrene or acrylonitrile butadiene) (Fig. 1).

Two particle types were found to be microplastics: fibres (75.6%) and fragments/sheets (24.4%), but not spheroids. Samples S2 and W1 contained only microfibres. Fragment proportions were highest in W2 (44.4%) and W3 (42.9%) (Fig. 2F-J). In its greatest proportion, the colour of confirmed microplastics was described as 'other' (54.2%) i.e. not fitting any of the colour categories (Fig. 2E-O), followed by red (24.4%), blue (13.5%) and black (7.9%). In northern North Atlantic samples the second most prevailing colour was blue (Fig. 2 K). There was no difference in microplastic concentration between Mediterranean and Atlantic-connected samples (t-test $t = 1.3705$, $p > 0.05$)—429 vs 750 microplastics kg^{-1}. However, when excluding S2 due to the producer's clinical setup in salt harvesting, traditionally harvested microplastics (Mediterranean region) contain significantly less microplastics than industrially harvested sea salts (Atlantic-connected region) (t-test $t = 3.3778$, $p < 0.01$)—429 ± 227 vs 849 ± 332 microplastics kg^{-1}.

Particle sizes were established from potential microplastics (Fig. 3). The smallest particles were fragments of 12.6 μm and 13.6 μm (Atlantic and Mediterranean samples respectively) and fibres with a diameter of 6.4 μm (Atlantic) and 8.2 μm (Mediterranean). Median (mean ± SD) fibre diameter was 17.5 μm (17.3 ± 8.0) in Atlantic samples and 16.9 μm (16.8 ± 6.1) in Mediterranean salts. In Atlantic samples, six fragments were > 500 μm (ranging 577–2054 μm). In Mediterranean samples, two fragments were > 500 μm (513 and 566 μm) and the next largest was 291 μm. The median (mean ± SD) largest diameter of non-fibrous particles was 74.2 μm (171.8 ± 298.6) in the Atlantic and 64.8 μm (95.9 ± 95.7) in Mediterranean salts. Potential microplastics < 155 μm (supplementary information online, SI Fig. 3) were the most abundant (86.2%). By diameter, all fibres were < 155 μm and only three were longer than 155 μm. Of fragments/sheets, 80.1% were < 155 μm and 65% < 100 μm. By geographic region, 84.1% of particles from the North Atlantic and 95.0% from the Mediterranean were < 155 μm; their overall size distribution was similar (Fig. 4).

3.1.3. Additional data quality

Raman spectroscopy for polymer confirmation of a representative sample (n = 56) of potential microplastics resulted in microplastic confirmation of 13 spectra (23.2%). Match scores for confirmed plastics were 76.3–94.2% (Fig. 1 A,D+E). 42.9% of spectra were identified as not plastic, which included amorphous carbon, potential remnants of biota and minerals such as rhodochrosite, jarosite, muscovite and quartz (Fig. 1B). Such natural materials were confirmed with match scores ranging 75.0–99.3%. A further 3.6% were nitrocellulose fibres which were assumed to be contamination of the CN filters (Fig. 1 C). In addition, 25% of spectra were not usable—mainly due to oversaturation, and 5.4% provided inconclusive results (one was dye-related which could have equally been of natural or synthetic nature and two did not provide any results with any of the libraries). These unusable and inconclusive results are a potential source of underestimation of microplastic concentrations. Further data quality assessments can be found in the supplementary information online, Section 6.

3.2. Review of previously published work on food-grade salts

Up until July 2022, 31 studies were published on microplastics in salt for human consumption covering Africa, Asia, New Zealand, and Europe. For two of these, only the abstract is available in English, with full-text in Korean (Cho et al., 2019) and Turkish (Yurtsever, 2018). One study analyses de-icing salt for road gritting, with one sample being food-grade which was therefore included (Rødland et al., 2020). Three further studies concentrate on anthropogenic particles, i.e. no polymer identification was performed; these cover salt from the United States and Europe. Schymanski et al. (2020) investigate microplastic generation by domestic salt grinders, this study is included in the review since they analyse rock salt as their reference material Despite this limited number of studies, the topic has been reviewed 16 times (Table 2). Of these, five concentrate on salt for human consumption while the remainder investigate numerous food items. Most of these studies calculate microplastic ingestion rates to assess human exposure to microplastics. Methods for microplastic extraction and identification are rarely scrutinised in depth; while half of the reviews assess particle and/or filtration sizes, none evaluate contamination control or mitigation (Table 2).

3.2.1. Review of reporting analytical methods and results of relevant studies

In general, microplastic findings are reported as mean values, but 15 studies do not state which basic statistic was used, one of those not reporting microplastic concentrations at all. Only study 18 (Table 4; Fischer et al., 2019) provide mean and median values. Microplastic concentrations vary greatly amongst studies (Table 3) from 0 to over 1 million microplastics kg^{-1} of salt. Similarly, when assessed, microplastic mass ranges 14 μg to 35,000 μg kg^{-1} of salt. Interstudy comparison is hindered by partial assessments of samples, minimum particle size and potentially by category focus (Table 4). Thirteen studies only analyse the supernatant of the samples; five do not report this information, four are unclear and one study reverts to supernatant when filters are too caked with particles. Furthermore, minimum particle size assessed ranges from

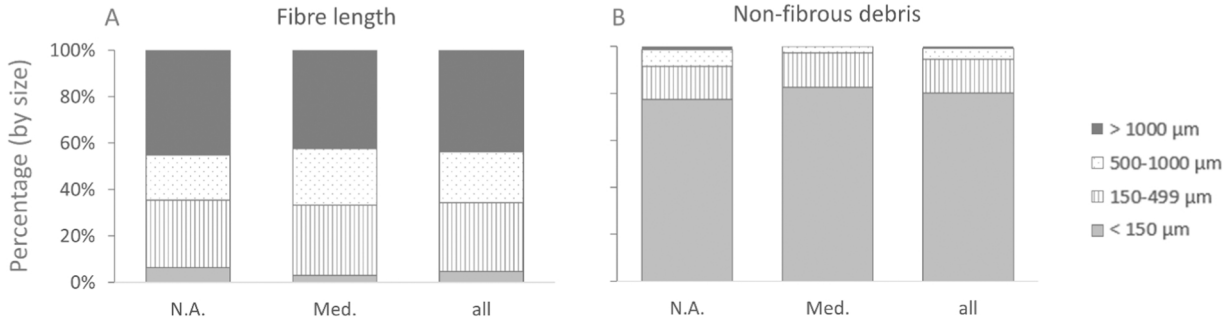

Fig. 3. Sizes of potential microplastics based on 210 images in four size classes (< 150, 150–499, 500–1000 and > 1000 μm) for fibre length (graph A) and largest dimension of non-fibrous debris (graph B) by means per geographic region (N. A. = North Atlantic, Med. = Mediterranean).

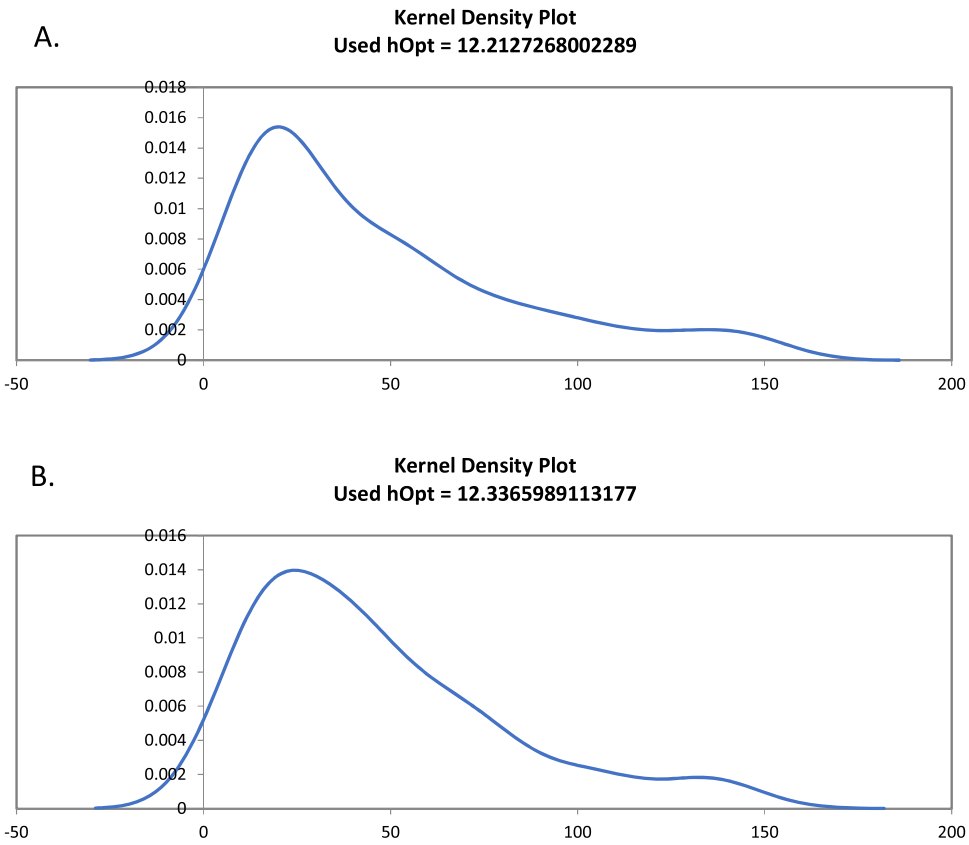

Fig. 4. Kernel density estimation of potential microplastic size distribution < 155 μm. Size refers to largest dimension for fragments/sheets but diameter for fibres. (A: 92 particles from the North Atlantic; B: 89 from the Mediterranean). For breakdown by size bins and particle types see supplementary information online, SI Fig. 3.

1 μm to almost 400 μm. Nine studies do not report this information. Particle category proportions, not mentioned by nine, range from 100% fibrous to 7% microfibres and 93% fragments (Table 3). Using destructive pyrolysis-gas chromatography-mass spectrometry, Fischer et al. (2019) do not report categories.

Certain analytical steps vary across studies (Table 4). To identify if particles are of polymeric origin, FTIR is the tool of choice (20: studies 1, 5, 8–12, 14, 15, 18–20, 24–27, 32–35, with study 11 using automated FTIR), followed by Raman (9: studies 2, 21–23, 28, 30, with studies 3, 16 and 31 using automated Raman). Study 13 combines FTIR and Raman. The following techniques are used once: pry-GC/MS (study 17), visual assessment only (study 7 – later repeated as study 11), Rose Bengal (study 6) and Nile Red staining (study 29). While in six studies all potential microplastics are analysed with spectroscopy, others analyse between < 2% per filter and 2.5–83% of potential microplastics. Fifteen studies do not report this information. Plastic confirmation rates vary greatly between 4% and 93% and are not reported by more than half of the studies. Similarly, most studies do not reveal the minimum library search match score they had employed. Contamination mitigation reporting is also limited. Not all studies use procedural blanks, less use airborne controls and less than half of the studies state performing blank adjustments to their results. Only studies 3 and 26 employ airborne contamination controls despite using a clean environment to process their samples—interestingly Gündoğdu (2018) (Study 3) find microplastics contamination. Only two studies report that all clothing, not only laboratory coats, were made of 100% cotton. Fourteen studies do not report on any contamination control; a further two were written in Turkish and Korean, hence contamination control might have been covered in the main text, but not in the abstract (Table 4).

3.3. Comparison of results from other studies covering sea salts from Europe

The above review on microplastics in food-grade salts uncovered analytical differences between published studies (Table 3 + 4). Using the present study as the common denominator for methods and microplastic concentrations in European sea salts, results of other studies were adjusted accordingly. Table 5 shows microplastic estimates using pseudo-harmonisation of methods for European sea salts. While the purchase date is unknown for eleven samples, six were purchased between September 2016 and September 2017 and four in August 2018. One sample of French sea salt did not contain microplastics, hence the adjusted value did not change. For the remaining samples, concentration changes ranged − 70% to + 2635%. Reported microplastic concentrations were 1–320 items kg^{-1} (anthropogenic particles up to 19,800 items kg^{-1}) and after adjustments 26–4933 microplastics kg^{-1}. Since unclear reporting warranted minimum/maximum adjustments for four sampling locations (Table 5), adjusted mean microplastics concentrations for European sea salts are between 811.8 ± 1174.3 and 1414.3 ± 1891.4 per kg sea salt (respective medians 276.4 and 783.9 adjusted microplastics kg^{-1}).

3.4. Human dose exposure through consumer choice

Based on customer choice salt ingestion of 2 g day^{-1}, 293 microplastics < 150 μm (189 non-fibrous) could be consumed annually (equalling the maximum upper dose through intestinal absorption rate) which is estimated to be 378 μg $year^{-1}$. At an intestinal absorption of 1%, < 3 microplastics $year^{-1}$ are entering the human body across the gut epithelium through sea salt consumption, approximately 4 μg $year^{-1}$. To this, an additional < 9 microplastics $year^{-1}$ might translocate from salts

Table 3
Geographical information, microplastic concentrations, sample completeness, minimum target size and proportions of particle categories provided by studies investigating microplastics and anthropogenic debris in food-grade salts that were reviewed. Literature search conducted 08/07/2022.

Region/ country	Microplastic concentrations reported (unless otherwise stated particle/kg)	Complete samples analysed?	Smallest particle assessed/found	Microplastic categories found			
				Fibres	Fragments	Sheet	Study ID
China	550–681	Supernatant only	45 μm	c.57.5%	c.40%	c.2.5%	1
Global	Totals per country 1–10 mps/kg	Supernatant only	160 μm	25.6%	63.8%	10.6%	2
Turkey	16–84	Supernatant only	20 μm, but only 3% were < 100 μm.	70%	18%	12%	3
Turkey	56	n/r	n/r	Not reported in abstract (article in Turkish)			4
Spain	50–280	Supernatant only	30 μm	100%			5
Global	47–806 *	Yes	Smallest assessed 0.1 mm	99%	1%		6
Italy	0–600 *	Yes	Smallest found 4 μm; < 100 μm most-found particle size	Mainly fragments			7
Croatia	13,500–19,800 *	Yes	"	80%	20%		"
India	56–103	Yes	n/r	37%	63%		8
Global	0 – 1674 * *	Supernatant only	100 μm	31%	63%	6%	9
Taiwan	9.77	Yes	90 μm	7%	93%		10
Italy and Croatia	70–320	Yes	Only particles 10–150 μm assessed.	Mainly fibres			11
Indonesia	6.7–53.3	Yes	390 μm	93%	7%		12
South Korea	1000	Yes	10 μm	Not reported in abstract (article in Korean)			13
India	n/r	Supernatant only	No info on smallest, but 60% smaller than 100 μm	42%	55%	3%	14
India	35–72	Supernatant only	55 μm	83%	17%		15
Germany	666,000–1060,000 (348,000–370,000)	Yes	≥ 1 μm (≥10 μm)	n/r			16
Europe	Mass 14–1993 μg/kg	Supernatant only	Filter 20 μm, but with pyr-GS/MS sizing not possible	ID method does not allow for categorisation			17
Spain	120	n/r	59 μm	11%	89%		18
India	500–1600	Unclear	47 μm	100%			19
India	115–575	Supernatant only	100 μm	Mainly fibres			20
India	1300–2248	If filter was caked, only supernatant.	38% 20–100 μm, 28% 100–1000 μm	39%	44%		21
China	212–413	Supernatant only	18 μm, c.20% particles < 200 μm	c.50%			22
Vietnam	64–115	n/r	Unclear	Mainly fibres and fragments			23
Bangladesh	78–137	Yes	250 μm, but mainly 500–1000 μm	24%	48%		24
Indonesia	55–403	Yes	n/r	n/r			25
Africa	24–80	Unclear	3.3 μm	94%			26
India	c.600–700	Yes	3.8 μm; 20% < 100 μm		50%		27
Indonesia	Mass 7700 μg/kg	n/r	n/r	n/r			28
France	33–486 *	Unclear	50 μm	n/r			29
Germany	688 *	Unclear	50 μm	n/r			"
Indonesia	Mass 35,000 μg/kg	n/r	n/r	n/r			30
Iran	151–1417	Supernatant only	39–43% 1–9 μm, 32–34% 10–50 μm, 16–17% 50–100 μm	Mainly fibres			31
Sri Lanka	17–122	Supernatant only	n/r	n/r			32
India	3–52	Supernatant only	16% < 500 μm	Mainly fragments			33
Spain	100–380	Yes	n/r	94%			34
New Zealand	120	Yes	n/r	Mainly fragments			35

* anthropogenic particles, * * study excluded an outlier of 13,629 microplastics kg^{-1}, n/r = not reported; Study ID: 1: Yang et al. (2015), 2: Karami et al. (2017b), 3: Gündoğdu (2018), 4: Yurtsever (2018), 5: Iñiguez et al. (2017), 6: Kosuth et al. (2018), 7: Renzi and Blašković (2018), 8: Seth and Shriwastav (2018), 9: Kim et al. (2018), 10: Lee et al. (2019), 11: Renzi et al. (2019), 12: Tahir et al. (2019), 13: Cho et al. (2019), 14: Selvam et al. (2020), 15: Sathish et al. (2020), 16: Schymanski et al. (2020), 17: Fischer et al. (2019), 18: Rødland et al. (2020), 19: Nithin et al. (2021), 20: Vidyasakar et al. (2021), 21: Yaranal et al. (2021), 22: Feng et al. (2021), 23: Khuyen et al. (2021), 24: Rakib et al. (2021), 25: Dwiyitno et al. (2021), 26: Fadare et al. (2021), 27: Sivagami et al. (2021), 28: Wibowo et al. (2021), 29: Sturm et al. (2021), 30: Luqman et al. (2021), 31: Sharifi and Attar (2021), 32: Kapukotuwa et al. (2022), 33: Manimozhi et al. (2022), 34: Masiá et al. (2022), 35: Mazlan et al. (2022).

outside consumer choices (based on Yang et al., 2015; for workings see supplementary information online, sections 7–8). Approximately 14 μg of microplastics are therefore estimated to pass the gut barrier into the human body through overall annual salt consumption at an intestinal absorption rate of 1% (supplementary information online, SI Table 9).

4. Discussion

Microplastic contamination has previously been found in salts for human consumption, but little is known about human exposure potential, and how microplastic concentrations in sea salts from different locations – and maybe even more importantly – between different studies differ. We therefore set out to calculate exposure rates based on particle characteristics and consumer choice. In addition, we assessed how individual studies using different method approaches differed and compared to microplastic concentrations in our salt samples. This novel multidisciplinary approach has not been taken by other authors to date.

This study analysed European sea salts; all but one replicate contained microplastics. Kim et al. (2018) suggest that microplastic contamination load in sea salts is correlated to contamination of surrounding sea water. However, despite the Mediterranean being considered one of the global hotspots for microplastic contamination

Table 4
Spectroscopy analysis and contamination control details of studies investigating microplastics and anthropogenic debris in food-grade salts that were reviewed. Literature search conducted 08/07/2022.

Spectroscopy analysis information			Contamination control information						
% potential microplastics subjected to spectroscopy	% plastic confirmation rate	Library search minimum match scores reported?	Reported on other than laboratory coat to be cotton?	Filtered reagents?	Clean environment?	Airborne controls	Procedural blanks	Blank adjustments?	Study ID
152 of unknown total	85%	Yes (70%)	No	Yes	No	No	Yes	n/r	1
All	42%	Yes (70%)	No	Yes	Yes	No	Yes	n/r	2
All	n/r	No	No	No	Yes	Yes (dry filter papers)	No	n/r (airborne controls contained microplastics)	3
n/r		No	Not reported in abstract (article in Turkish)						4
Some	93%	Indirectly*	No	No	No	No	Yes	n/r (blanks contained microplastics)	5
n/a	n/a	n/a	No	No	Yes	No	Yes	Yes (subtracted what was found in blanks)	6
n/a	n/a	n/a	No contamination control reported				n/a		7
Some* *	n/r * *	No	No	Yes	No	No	Yes	Yes (unknown how)	8
All	76%	Yes (70% accepted, 60% visually assessed)	No	Yes	No	Yes (dry filter papers)	Yes	Yes (set method detection limit to 0.72 particles/kg salt for PET fibres)	9
All	7%	No	No	Yes	No	Yes	Yes	No microplastics in blanks	10
All	4–45%	Yes (65%)	Yes	Yes	Yes	No	Yes	Yes (set LOQ to 2.8 despite blanks void of microplastics)	11
All	n/r	No	No contamination control reported						12
113 of unknown total	9%	No	Not reported in abstract (article in Korean)						13
Unknown	73%	No	No contamination control reported						14
75 of unknown total	88%	Yes (> 80%)	No	Yes	No	No	Yes	Yes (values from blanks subtracted)	15
All	n/r	No	No	Yes	Yes	No	Yes	n/r	16
n/a	n/a	n/a	Yes	Yes	No	No	Yes (also internal standards-PAHs and couple of acids)	Yes	17
47%	17%	Yes (60%, all manually assessed)	No	Yes	Yes	No	Yes	Yes	18
n/r	n/r	No	No	Some?	No	No	No	No	19
2.5%	90%	No	No	Some?	No	No	Yes	n/r	20
50 per sample of various 1000 s	60%	No	No	No	No	No	No	No	21
60%	90%	Yes (accepted >70%)	No	Some?	Some?	Yes	Yes	n/r	22
n/r	n/r	No	No	Some?	No	Yes	Yes	No fibres found	23
unknown	n/r	No	No	Yes	No	No	Yes	No microplastics in blanks	24
n/r	n/r	No	No	Some?	No	n/r	n/r	No microplastics in blanks	25
11% of 82 fragments, none of 1246 fibres	n/r	No	No	No	Yes	Yes	Yes	No microplastics in blanks	26
n/r	n/r	Yes (<60% rejected, >70% accepted)	No	No	No	No	Yes	n/r	27
n/r	n/r	No	No	No	No	No	No	No	28
n/a	n/a	n/a	No	No	Partly	No	Yes	Yes	29
n/r	n/r	No	No	No	No	No	No	No	30
< 2% per filter	n/r	No	No	No	Yes	n/r	n/r	Yes	31
50%	n/r	No	No	Yes	Yes	No	Yes	No microplastics in blanks	32
n/r	n/r	No	No	No	No	No	No	No	33
7%	n/r	No (assumed that highest score was the correct one)	No	Yes	No	No	Yes	n/r	34
10 of 12	10%	No	No	No	No	No	No	No	35

* Referred to Woodall et al. (2014), * * according to Lee et al. (2019) 1.5% were assessed with 80% plastic confirmation rate, n/r = not reported; Study ID: 1: Yang et al. (2015), 2: Karami et al. (2017b), 3: Gündoğdu (2018), 4: Yurtsever (2018), 5: Iñiguez et al. (2017), 6: Kosuth et al. (2018), 7: Renzi and Blašković (2018), 8: Seth

and Shriwastav (2018), 9: Kim et al. (2018), 10: Lee et al. (2019), 11: Renzi et al. (2019), 12: Tahir et al. (2019), 13: Cho et al. (2019), 14: Selvam et al. (2020), 15: Sathish et al. (2020), 16: Schymanski et al. (2020), 17: Fischer et al. (2019), 18: Rødland et al. (2020), 19: Nithin et al. (2021), 20: Vidyasakar et al. (2021), 21: Yaranal et al. (2021), 22: Feng et al. (2021), 23: Khuyen et al. (2021), 24: Rakib et al. (2021), 25: Dwiyitno et al. (2021), 26: Fadare et al. (2021), 27: Sivagami et al. (2021), 28: Wibowo et al. (2021), 29: Sturm et al. (2021), 30: Luqman et al. (2021), 31: Sharifi and Attar (2021), 32: Kapukotuwa et al. (2022), 33: Manimozhi et al. (2022), 34: Masiá et al. (2022), 35: Mazlan et al. (2022).

Table 5
Pseudo-harmonised microplastic concentrations in European sea salts from previously published studies based on mean values of this study. Literature search conducted 08/07/2022.

Region/ country	Study	Samples bought/ collected	n	Smallest particle assessed/ found.	Reported microplastics/kg (for min/max range, mean value was used)	Concentration after adjustment for size	Concentration after adjustment when study focus on			Adjusted microplastics/kg
							supernatant	microfibres	anthropogenic particles	
	This study	Summer 2016	12	5 µm (smallest fragment 13 µm)	74-1,155 (540)	36.2% < 30 µm; 55.7% < 50 µm; 61.4% < 60 µm; 75.7% < 100 µm; 86.1% < 150 µm; 13.9% ≥ 150 µm	27.3% in the supernatant	75.6% fibres	29.6% plastic	
France	9	Jan-Sept 2017	1	100 µm	0	0.0	0.0	→	→	0.0
France	2	unknown	6	160 µm *	1	7.2	26.4	→	→	26.4
North Sea	6	Aug 2018	1	100 µm **	66.6 ^	274.1	→	→	81.2	81.2
Portugal	2	unknown	3	160 µm	3.3	23.7	87.0	→	→	87.0
Croatia	11	unknown	5	Only 10-150 µm assessed.	70-200 (135)	156.8	→	→	→	156.8
Mediterranean	6	Aug 2018	2	100 µm **	133 ^	547.3	→	→	162.2	162.2
France	29	unknown	n/r	50 µm ***	33-486 (260) ^	586.9	586.9-2,149.8	586.9-2,843.7	173.7-2,843.7	173.7-2,843.7
Celtic Sea	6	Aug 2018	2	100 µm **	113-187 (150) ^	617.3	→	→	182.9	182.9
Spain [a]	34	unknown	10	n/r ****	100-380 (240)	240-1726.6	→	→	→	240-1,726.6
Italy	9	Jan-Sept 2017	2	100 µm	4-30 (17)	70.0	256.3	→	→	256.3
Sicily	6	Aug 2018	1	100 µm **	220 ^	905.3	→	→	268.3	268.3
Italy [b]	11	unknown	6	Only 10-150 µm assessed.	170-320 (245)	284.6	→	→	→	284.6
Spain	18 [d]	unknown	1 [e]	59 µm *****	120	310.9	310.9-1,138.8	→	→	310.9-1,138.8
Germany	29	unknown	n/r	50 µm ***	688 ^	1553.0	1,553.0-5,688.8	1,553.0-7,524.9	459.7-7,524.9	459.7-7,524.9
Turkey	3	Feb-March 2017	5	20 µm ******	46	189.3	693.4	→	→	693.4
Croatia	9	Jan-Sept 2017	1	100 µm	58	238.7	874.3	→	→	874.3
Spain	5	Sept 16-June 17	21	30 µm	50-280	258.6	947.3	1,253.1	→	1,253.1
Italy	7	unknown	5	4 µm	4988 ^	→	→	→	1477.9	1,477.9
UK	9	Jan-Sept 2017	1	100 µm	136	559.7	2,050.1	→	→	2,050.1
Italy [c]	7	unknown	1	11 µm	7640 ^	→	→	→	2,263.7	2,263.7
Spain	5	Sept 16-June 17	21	30 µm	50-280	258.6	947.3	4,248.1	→	4,248.1
Croatia	7	unknown	5	4 µm	13,500-19,800 (16,650) ^	→	→	→	4,933.3	4,933.3

a bought from Spanish supermarkets, locations not disclosed; b their samples HC1-3 & LC1 + 3; c LC2; d unknown if entire sample or only supernatant were analysed leading to a range value to express this uncertainty; e remaining samples were not food-grade;
* adjusted to 150 instead of 160 µm; * * smallest size given as 0.1 mm, hence 100 µm may be overestimate; * ** lower size limit set to this value, but size of smallest particle found unknown, so possibly an underestimate; * ** * since particle size is not reported, estimates are very likely an underestimate (with the assumption that they only reported >150 µm, using this as upper value; ***** adjusted to 60 instead of 59 µm; ****** lower size limit set to this value, but only 3% were < 100 µm;
^ anthropogenic particles (i.e. only potential microplastics)
Study ID: 2: Karami et al. (2017b), 3: Gündoğdu (2018), 5: Iñiguez et al. (2017), 6: Kosuth et al. (2018), 7: Renzi and Blašković (2018), 9: Kim et al. (2018), 11: Renzi et al. (2019), 18: Rødland et al. (2020), 29: Sturm et al. (2021), 34: Masiá et al. (2022).

(GESAMP, Kershaw, 2015; Llorca et al., 2020), less microplastics were found in Mediterranean sea salts compared to salts harvested from European shores outside the Mediterranean basin and north of 45°N. It was previously established that sea salt is more heavily contaminated with microplastics compared to lake, rock and well salts (Kim et al., 2018; Yang et al., 2015). Kim et al. (2018) further found that Asian sea salts are significantly more contaminated than sea salts from other regions, but that rock salts are similarly contaminated globally. Since rock salts are usually mined from underground halite deposits, i.e. salts evaporated from ancient seas, the raw material is unlikely to be contaminated by solid anthropogenic contaminants such as microplastics. Therefore, we suggest that harvesting and processing techniques are the source of contamination. All but one of the products coming from facilities with industrial harvesting had significantly higher concentrations of microplastics compared to traditional harvesting. While the least contaminated sample (S2) comes from an industrial facility, a desk study suggests that a very clinical extraction process is applied at that factory. This suggests that adjustments to the harvesting process such as manufacture in plastic-free conditions could reduce microplastic loads in industrial harvested sea salts.

Before any global conclusions about microplastic concentrations between the marine environment and marine edible resources can be drawn, the analytical process will need to be harmonised. Interstudy result variation is often blamed on differences in study methods (Hidalgo-Ruz et al., 2012; Kim et al., 2018; Lee et al., 2019). For this reason, a previously published method by Yang et al. (2015) was closely followed for our analysis of salt samples. Superficially, results differ. However, Yang et al. (2015) only analyse the supernatant, which in our case yielded only 27.3% of potential microplastics. They report a maximum concentration of 681 microplastics kg^{-1} in sea salt. The supernatant of our samples contained concentrations of the same magnitude, i.e. 315 microplastics kg^{-1}. Likewise, Iñiguez et al. (2017) suggest that their results are not comparable to Yang et al. (2015). However, Iñiguez et al. (2017) only report fibres, and, of the overall total microplastics found by Yang et al. (2015) in sea salt approximately 45% were fibres, making the upper particle limit very similar with 280 microfibres kg^{-1} in Spain and 306 fibres kg^{-1} in China. Reviewing existing studies uncovered a great variation in analytical procedures, in line with

previous reviews (Kim et al., 2018; Lee et al., 2019). We went further and used a pseudo-harmonisation process of applying simple adjustment factors to numerically indicate the effects of different analytical steps. Once differences such as use of density separation, target sizes and particle types are addressed, microplastic concentrations in other European sea salts are generally more concordant with our own results. While this approach only offers estimates and cannot be used to accurately compare results, it is a clear indication that interstudy comparability can only be achieved with harmonised methods. Specifically, difference in target particle sizes leads to the greatest variation in results, making filtration the most crucial procedure for method harmonisation. Numerous studies only assess particles to ≥ 100 μm, which in our study would have captured only $< 25\%$ of microplastics we found to be present. Complete extractions, rather than density separation and filtering solely the supernatant, seems a further crucial procedure to harmonise. Preferably, extraction techniques should be capable of isolating particles to a single-digit micrometre size in line with current spectrometric limitations (Käppler et al., 2016). Automation in particle identification would be very beneficial to eliminate observer bias. For example, Schymanski et al. (2020) report almost 17-fold more microplastics using automated Raman spectroscopy compared to our study using manual Raman spectroscopy. It is worth noting that certain variations in methods such as contamination control and mitigation, the automatic acceptance of polymer library suggestions or how final microplastics results are calculated from initial particle counts of potential microplastics may also increase differences in microplastic concentration between studies (Hermsen et al., 2018; Horton et al., 2017; Smith, 2011). For this reason, it is imperative to report information including, but not limited to contamination control conditions, how many particles were subjected to the method of polymer identification, comparison scores to libraries etc.

Edible marine resources have been suggested to be a significant source of microplastics in human diets (Karami et al., 2017b; Yang et al., 2015). However, initial bias amongst the research community—focussing on the marine environment—may be the reason for this and many of the 'early adopters' who have led the direction of microplastics research have come from a marine science background. More recent work indicates greater levels of microplastics contamination in terrestrial food sources and indoor environments (Catarino et al., 2018; Kedzierski et al., 2020; Schymanski et al., 2018). In addition, the exposure potential from sea salt in a Western diet is low. We estimated that humans consume < 1200 microplastics smaller than 150 μm (< 1.4 mg) with salt annually, < 300 (< 0.4 mg) of those may result from choosing to consume sea salt. Previous estimates of microplastic exposure through salt consumption lack considering particle size and intestinal absorption rates. Particle hazardousness is assumed to be related to particle size (Gray and Weinstein, 2017; Wright and Kelly, 2017), but while sea salts may be more contaminated with microplastics than other salts, Yang et al. (2015) suggest that microplastics < 100 μm are more prevalent in lake and rock/well salts than in sea salts. From a human perspective, microplastics < 150 μm are likely of exposure concern due to their potential to translocate from the digestive tract into the body (Volkheimer, 2001; Welle and Franz, 2018; Wright and Kelly, 2017). Despite this potential, it is unlikely that all microplastics, even small ones, are absorbed. Schwabl et al. (2019) show that the human body is capable of excreting microplastics. Therefore, intestinal absorption is an important mechanism to consider since, i.e. it seems highly unlikely that all ingested particles translocate from the digestive tracts through the gut epithelium into the body. Also, while particles < 150 μm may pass the intestinal epithelium, they might only be systemically bioavailable at much smaller sizes (Paul et al., 2020). Recently, only microplastics 5–10 μm (filtration pore size 1.6 μm) were found in human placentas (Ragusa et al., 2021). Rodent models suggest absorption/translocation rates of $< 1\%$ across the mammalian gut epithelium for particles > 5 μm (Delie, 1998; Norris et al., 1998). Particles < 5 μm, on the other hand, may exhibit a greater absorption rate into the body (Delie, 1998; Norris et al., 1998), but knowledge of microplastics in food-grade salt – or any other matrix - is scarce for such small sizes. Work by Schymanski et al. (2020) suggest that concentrations of microplastics $\geq 1 < 10$ μm in rock salt may be almost twice as high than concentrations of microplastics ≥ 10 μm. We calculated that annually < 3 microplastics may not be excreted but translocated across the gut epithelium annually by choosing to consume sea salt (plus six microplastics if processed foods are prepared with rock salts). If such low bioavailability and overall low absolute number of microplastics could lead or contribute to any human health effects remain to be seen. Further, sea salts are also contaminated with non-microplastic foreign particles. This begs the question if the ingestion of cotton and cellulose fibres or sediment and amorphous carbon grains could lead to potentially similar health effects through damage, sorption capacities of harmful chemicals, leaching of dyes etc. Furthermore, microplastic is an umbrella term for all plastic polymers and these types exert different levels of toxicity. For example, polystyrene exerts a toxic effect, but polypropylene or polyethylene terephthalate may not (Nelson et al., 2011). Finally - the extraction and characterisation methods used usually used for 'conventional' microplastics overlook tyre wear microparticles, although they are probably present in the environment in similar amounts. We identified the possible presence of acrylonitrile butadiene which may indicate salt can be contaminated by these materials—an area where further work may be necessary (Knight et al., 2020).

At present, evidence is lacking that microplastics in foods lead to hazardous exposure in humans; however, their presence in sea salt likely both increases that exposure, and points to other routes to increasing microplastics in the environment. The presence of microplastics in food-grade salts suggests contamination of salts harvested for other uses, too. Salt is used for salt licks for cattle, deer and pets, supplementation in freshwater fish aquaculture, but also as dishwasher salts and gridding material in winter months (Cnaani et al., n.d.; Kubitza, n.d.; Roy et al., 2007; Staurnes and Finstad, 2000). Rødland et al. (2020) recently reported microplastics in de-icing salts. Through these applications and digestive process of animals and humans alike, microplastics may be released into the environment again—initially into other compartments and potentially in greater numbers through contamination added during the production process—making them an environmental concern. For example, microplastics present in fishmeal have been shown to be consumed by fish (Hanachi et al., 2019), therefore the same fate could be expected for both fishmeal used for animal feed (a very widespread practice), and microplastic contamination in salts for animal supplementation. Similarly to microplastics in personal care products or released from laundering, microplastics in salts may end up in waterways where they could be removed during wastewater treatment, escape the removal process or be dumped elsewhere and end up in the marine environment (again) (Browne et al., 2011; Murphy et al., 2016; Napper et al., 2015; Napper and Thompson, 2016; Rødland et al., 2020).

4.1. Limitations and outlook

This study may be limited by subjecting a small number of particles to Raman spectroscopy and potential issues with laboratory-based contamination. Only 6% of particles were subjected to spectroscopy, mainly because particles < 100 μm had to be kept in situ on filters after initial enumeration to avoid losing them. A potential confounding factor in our calculations is that industrial harvesting is usually conducted at higher latitudes and traditional harvesting around the Mediterranean Basin, but also as far north as 49° in Southern Brittany, France. More work is needed to understand differences in microplastic contamination through harvesting and production processes. We applied a pseudo-harmonisation approach between our study and other research conducted on European sea salt based on differences in extraction and identification approaches. Firstly, this approach is hindered by the uncertainty surrounding our own findings and the general heterogeneity in environmental contamination research. Secondly, only four

methodological differences were adjusted for as many others may be difficult to quantify, e.g. details in spectroscopic assessments such as match score thresholds, use of clean room conditions vs normal laboratory setups. However, we feel that this rudimentary approach was suitable to emphasise how apparent differences in microplastic abundance in sea salt may be removed by applying method standardisation or harmonisation in microplastics research. Furthermore, it has highlighted the urgent need to conduct further method standardisation research and implement harmonised protocols for future studies.

5. Conclusion

Microplastics are found in sea salts across Europe. Harvesting technique may influence those concentrations, with generally lower microplastic load in traditionally harvested products from the Mediterranean Sea. Using realistic intestinal absorption rates, rather than assuming 100% absorption, consumers who choose European sea salts may absorb < 3 microplastics year^{-1} (approximately 4 μg year^{-1}). Microplastics in food-grade salt have been frequently studied in recent years but inter-study comparability is hindered by lack of harmonisation of analytical techniques. It is recommended to harmonise and optimise such techniques. Research should also move away from simple quantification of microplastics in edible sea salt to establishing driving factors in this contamination such as harvesting techniques, to determining the fate of microplastics in other salt applications and conduct the groundwork to be able to perform adequate risk assessments is recommended.

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CRediT authorship contribution statement

Christina J. Thiele: Conceptualisation (except original salt research), Investigation (extractions, particle measurements, Raman spectroscopy, literature review), Analysis, Writing (original draft) **Laura Grange:** Supervision (microscopy), Writing (review & editing) **Emily Haggett:** Conceptualisation (original salt research), Investigation (extractions, particle measurements, desk study of production methods), Writing (review & editing) **Malcolm D. Hudson:** Conceptualisation (original salt research and questionnaire), Investigation (questionnaire), Supervision, Writing (review & editing) **Philippa Hudson:** Conceptualisation (questionnaire), Investigation (questionnaire), Writing (review & editing) **Andrea E. Russell:** Supervision (Raman spectroscopy), Writing (review & editing) **Lina Zapata Restrepo:** Supervision (laboratory), Writing (review & editing).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data supporting this study are openly available from the University of Southampton repository at https://doi.org/10.5258/SOTON/D2345.

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Disclosure of previous publications

A small selection of results from this manuscript were presented at ECEC21, online, in April 2021 as a talk.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2023.114782.

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