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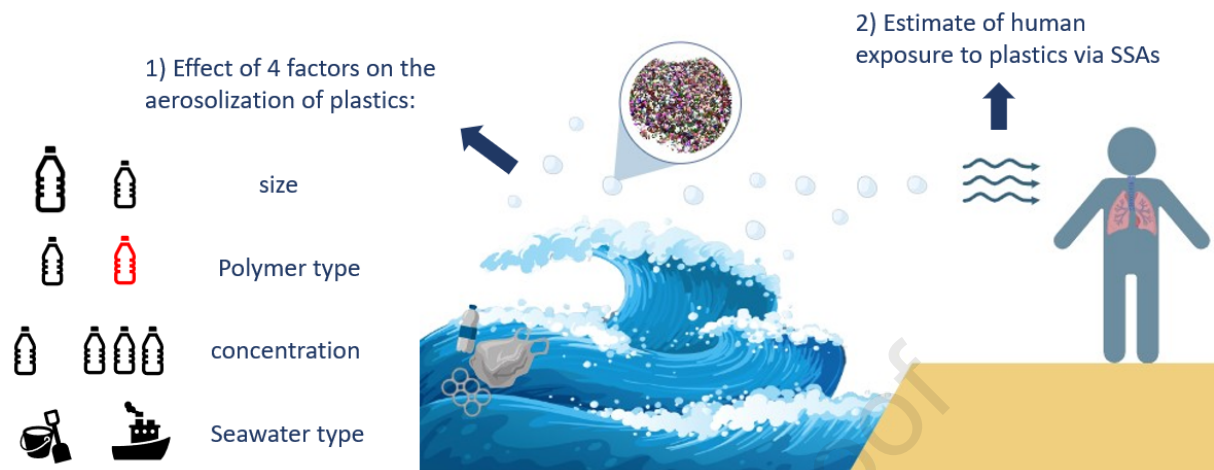
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Aerosolization of Micro- and Nanoplastics via Sea Spray: Investigating the Role of Polymer Type, Size, and Concentration, and Potential Implications for Human Exposure

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55 **Abstract**

56 Micro- and nanoplastics (MNPs) can enter the atmosphere via sea spray aerosols (SSAs), but the effects of plastic
57 characteristics on the aerosolization process are unclear. Furthermore, the importance of the transport of MNPs
58 via these SSAs as a possible new exposure route for human health remains unknown. The aim of this study was
59 two-fold: (1) to examine if a selection of factors affects aerosolization processes of MNPs, and (2) to estimate
60 human exposure to MNPs via aerosols inhalation.

61 A laboratory-based bubble bursting mechanism, simulating the aerosolization process at sea, was used to
62 investigate the influence of MNP as well as seawater characteristics. To determine the potential human exposure
63 to microplastics via inhalation of SSAs, the results of the laboratory experiments were extrapolated to the field
64 based on sea surface microplastic concentrations and the volume of inhaled aerosols.

65 Enrichment seemed to be influenced by MNP size, concentration and polymer type. With higher enrichment for
66 smaller particles and denser polymers. Experiments with different concentrations showed a larger range of
67 variability but nonetheless lower concentrations seemed to result in higher enrichment, presumably due to
68 lower aggregation. In addition to the MNP characteristics, the type of seawater used seemed to influence the
69 aerosolization process. Our human exposure estimate to microplastic via inhalation of sea spray aerosols shows
70 that in comparison with reported inhaled concentrations in urban and indoor environments, this exposure route
71 seems negligible for microplastics. Following the business-as-usual scenario on plastic production, the daily
72 plastic inhalation in coastal areas in 2100 is estimated to increase but remain far below 1 particle per day.

73 This study shows that aerosolization of MNPs is a new plastic transport pathway to be considered, but in terms
74 of human exposure it seems negligible compared to other more important sources of MNPs, based on current
75 reported environmental concentrations.

76 **Keywords:** microplastics, nanoplastics, sea spray aerosols, atmosphere, human exposure, inhalation

77 **Highlights**

- 78 - Aerosolization increased with decreasing Polyethylene particle size
- 79 - Plastic concentration and polymer type influence plastic enrichment in aerosols
- 80 - Human plastic exposure via sea spray seems negligible compared to urban and indoor

81 1. Introduction

82

83 An increasing number of studies show that the sea acts as a reservoir for atmospheric microplastics (MPs), either
84 through direct deposition of atmospheric MPs in the sea or through atmospheric fallout that enters the ocean
85 via runoff (Dris et al., 2017). Liu et al. (2019) revealed that suspended atmospheric MPs, especially textile
86 microfibers, are an important source of microplastic pollution in the ocean. Until recently, the sea has mostly
87 been seen as a sink for atmospheric plastics, but the sea could also be a source for atmospheric micro- and
88 nanoplastics. In addition to the long-range transport of particles from urban areas by wind, the ocean being a
89 source of atmospheric MPs itself could be an explanation for the high concentrations of microplastics in remote
90 uninhabited regions such as the Arctic (Bergmann et al., 2019). Brahney et al. (2021) suggest, based on
91 modelling, that this pathway from ocean to air is responsible for 0 to 17% of the atmospheric microplastics in
92 the USA. Among the hypothesized mechanisms, the transfer of plastic particles from the ocean to the air occurs
93 via sea spray aerosols (SSAs) (Allen et al. 2020).

94

95 SSAs are formed when breaking waves cause bubbles of trapped air to rise to the surface and burst. This first
96 step results in several hundreds of fine SSAs, which are called film drops. The burst bubble leaves a void behind
97 that is filled again with water. This second step creates a water jet that produces larger SSAs, namely jet drops
98 (Day, 1964; Blanchard, 1963). SSAs play a vital role in the Earth system, particularly in the interactions between
99 atmosphere, biosphere, climate, and public health (Fröhlich-Nowoisky et al., 2016). Together with the SSAs
100 particulate matter, microorganisms, fatty acids, carbohydrates, sterols, and proteins are aerosolized (Blanchard,
101 1963; Schiffer et al., 2018). For example, the study of Rastelli et al. (2017) indicated that SSAs were highly
102 enriched in organic matter compared to the seawater samples. Also, DNA, viruses and prokaryotes were
103 significantly enriched in SSAs (Pósfai et al., 2003). Furthermore, recent studies have shown that MNPs enter the
104 atmosphere via SSAs (Catarino et al., 2023; Shiu et al., 2022; Yang et al., 2022). So, this could be an important
105 transport pathway and an additional route for human exposure to MNPs. Exposure studies focused on inhalation
106 have, until now, mainly focused on urban exposure, but the exposure via SSAs on the coastal inhabitants and
107 tourists remains unclear.

108

109 Prior experimental evidence (Masry et al., 2021) has already indicated MNP particle transfer at the water-air
110 interface through a bubble bursting method in controlled laboratory conditions. Aerosolization is a complex
111 process dependent on characteristics of the water and substance. MNP size seemed to influence the
112 aerosolization capacity as it was reported that smaller particles aerosolize better (Catarino et al., 2023). Next to
113 size, the importance of factors such as MNP characteristics (e.g., polymer type) and concentration, that could
114 influence the aerosolization, remain to be elucidated. Insight into these processes is essential to estimate the
115 risk for human health and improve our understanding of this transport route. The aim of this study was,
116 therefore, two-fold: (1) to examine if attributes such as polymer types, sizes and concentrations of MNPs and
117 seawater affect the aerosolization process in an experimental setup, and (2) to estimate human exposure to
118 microplastics via aerosols inhalation based on the results of the aforementioned experiments.

119 2. Materials and methods

120 2.1 Experimental setup

121 The experimental setup we used for mimicking the SSA formation was performed as per Masry et al. (2021). A
122 schematic overview and detailed description can be found in SI A.1. To mimic the SSA formation, seawater was
123 collected at sea (specified locations, section 2.4), transported and stored in a 10L plastic barrel, kept at $15 \pm 1^\circ\text{C}$.
124 All experiments were performed in an exposure room at a constant temperature of $15 \pm 1^\circ\text{C}$. The seawater was
125 added in a glass container and air was pumped through a sintered air stone at the bottom. The aerosols formed
126 were caught on two types of filters placed in filter holders to capture both the aerosolized MNPs as well as
127 aerosolized sodium (Na^+). Na^+ is used as a proxy to quantify SSA densities as it correlates directly to the amount
128 of SSA (Lewis and Schwartz, 2004). MNPs were quantified using the cellulose nitrate filter (Whatman, diameter
129 47 mm, pore size of 0.8 μm or 0.2 μm ; Table A.1) (Semmoury et al., 2023). Aerosolized Na^+ was quantified using
130 the quartz filter (Whatman QM-A, diameter 47 mm, 2.2 μm pore size) as described in Van Acker et al. (2021b).
131 Van Acker et al. (2021) showed that the quartz filter is suitable to collect aerosols and optimized the full SSA
132 sampling method with quartz filters. The air supply through the air stone was 10 L/min or 5 L/min (depending
133 on the series, see Table A.1) and was calibrated using a rotameter (SKC, Inc). Aerosols were collected on the
134 filters using the Leland Legacy Sample Pumps (Cat. No. 100-3002; at 10 L/min) or 901-4011 SKC Flite4 sample

135 pumps (No. 22552498; at 5 L/min), depending on the series (Table A.1). Aerosols were collected for 24 hours
136 during each experiment. At the end of the experiment, the total runtime (24 h) and the total volume of air
137 pumped by each pump was verified to exclude any technical failures. The collected filters were stored in closed
138 glass petri dishes and stored at 4 °C before further analysis. A thorough validation confirmed the setup to be
139 suitable to study the aerosolization of MNPs in a controlled way. The filters' suitability to collect the aerosols
140 was validated and the bubble sizes created in the setup were measured and compared with the bubble
141 formation in the ocean (SI A.2).

142

143 Water samples were collected one hour after the experiment to compare the numbers of MNPs in the aerosols
144 with the MNPs in the surface and bulk water. Samples of the surface layer and the water column were collected.
145 For the samples of the surface layer (5 mL), a watch glass was placed just under the surface of the water and
146 lifted vertically as described by Harvey and Burzell (1972). The water column samples (10 mL) were collected
147 with a glass pipette. Three technical replicates of each water sample were taken: one replicate was used to
148 analyze the sodium concentration, one replicate was used for the quantification of the plastics, and one replicate
149 served as a spare sample. The water samples were stored at 4°C before further analysis.

150

151 To reduce contamination, the entire setup was cleaned thoroughly before the experiments using deionized
152 water (Brander et al., 2020). Additionally, where possible, commercial colored MNPs were purchased (color
153 range was selected based on the analysis technique) to be able to discriminate between contamination and
154 added MNPs. The filter holders were dried, and the filters were placed inside the filter holders in a clean laminar
155 flow cabinet, while a cotton lab coat was worn when preparing the filter holders. For every experiment, the
156 setup was filled with fresh seawater and a new air stone as well as new plastic tubes were used. Airborne
157 contamination was reduced as much as possible by storing all lab materials in a dust-free environment and by
158 covering all cups, beakers and bottles with aluminum foil or with pre-rinsed watch glasses. Glass, metal or
159 stainless-steel laboratory equipment was used when possible. Plastic tubing is used during the experiments, but
160 blank experiments were performed to verify that these were not causing additional MNPs to be present.

161

162 2.2 Series of experiments

163 Four different series of experiments were set up, to study the influence of (1) MNP size, (2) polymer type, (3)
164 MNP concentration and (4) seawater characteristics on the enrichment of plastics in sea spray aerosols.
165 Experiments were repeated between 1 to 3 times. The general protocol of each experiment was similar. MNPs
166 were added 24 h before the start of the aerosolization experiment to let the plastic particles distribute naturally
167 in the seawater. As there were some minor differences between the experiments, mainly to enable analysis of
168 different size classes of MNPs, an overview of all details of the experimental setup per experiment can be found
169 in Table A.1 in SI. Blank experiments (negative controls) were performed in between the experiments. These
170 blank experiments followed the exact same protocol without the addition of MNPs.

171

172 2.3 Microplastics and nanoplastics

173 Two polymer types were used: polyethylene (PE) and thermoset amino formaldehyde (TAF). TAF (1.3 g/cm²) has
174 a higher density than seawater and is used as a model for denser polymer types such as PET (1.38 g/cm³) and
175 PVC (1.38 g/cm³). To resemble the variety in plastic pollution in the environment and understand how density
176 may contribute to aerosolization, both floating and sinking polymers were tested. PE has a density lower than
177 seawater (0.995 g/cm³) and is used as a model for all buoyant polymer types such as PE and PP. Different sizes
178 of MNPs were used ranging from nanoplastics to 27 µm plastic particles (Table A.1). In this study microplastics
179 are defined as plastics between 1 and 5000 µm, and nanoplastics are all plastic particles smaller than 1 µm
180 (Gigault et al., 2018). All MNPs solutions used are polydisperse, to maximize the environmental relevance of the
181 results (Zimmermann et al., 2020). All used MNPs were characterized (Fourier-transform infrared spectroscopy
182 (FTIR) spectra, scanning electron microscope (SEM) images and microscope measurements) of which the results
183 can be found in SI A.

184 For experiments using PE (22-27 µm and 0.74-4.99 µm) and TAF (1-5 µm), plastic powders were weighted and
185 added to the seawater. For the experiments using the nanoplastics (PE mostly <1 µm), a solution of cryo-milled
186 PE particles was provided by the European Commission's Joint Research Center (JRC). The nanoplastics were
187 dissolved in a water:methanol (1.33:1) solution, with 0.1% (v/v) TritonX. To remove the solvent and dispersant,
188 the solution was centrifuged (5 min, 3500 RPM, 20 °C), the supernatant was removed and the nanoplastics were

189 dissolved in natural seawater. Nanosight characterization of the solution indicated that all particles are smaller
190 than 5 μm and that 80% of them are smaller than 1 μm (SI A). The lower size limit of the experiment is 40 nm,
191 based on the detection limit of the Nanosight, used to analyze the nanoplastics.

192 2.4 Seawater

193 Series 1 (except the nanoplastic experiments), 2 and 3 were performed using offshore seawater collected with
194 the RV Simon Stevin, in collaboration with the Flanders Marine Institute (Belgium). The seawater was sampled
195 using Niskin Bottles at station 330 (latitude 51° 25' 59.98", longitude 02° 48' 29.99") in the Belgian part of the
196 North Sea, at a depth of 3 m on the 6th of December 2021. It is assumed that this type of seawater did not
197 include the original sea surface microlayer (SSML) given the depth of sampling, as the SSML is defined as the
198 uppermost 1-1000 μm layer (Wurl and Holmes, 2008). For the series 1 nanoplastic experiments, coastal seawater
199 collected at a pier located in Ostend was used. This seawater was diluted using filtered near shore seawater
200 from the Flanders Marine Institute (30 % coastal, 70 % filtered) to reach similar turbidity as the offshore seawater
201 samples. In the experiment of series 4, coastal seawater collected at the beach in Ostend was used. It is assumed
202 that this type of seawater did include the original SSML. Which type of seawater was used in which experiment
203 is clearly stated in Table A.1.
204

205 2.5 Sample extraction and analysis

206 *Plastic concentration*

207 MNP concentrations were analyzed with different quantification techniques appropriate for their size, as no
208 single technique was able to measure across this large range of particle sizes (SI A). All blanks underwent the
209 exact same extraction and analytical procedure. To avoid contamination during the process, we adhered to the
210 same guidelines as specified under the experimental setup. The largest particles, defined as PE 22-27 μm , were
211 detected and quantified with the BX41 Olympus microscope directly from the cellulose nitrate filter. The water
212 samples (surface and bulk) were filtered over cellulose nitrate filters and the filters were placed directly under
213 the microscope. Images of a control sample (using the same type of filter and the same settings) were taken and
214 based on these, a color threshold was set in Image J to differentiate the plastic particles from the filter
215 background and other components. The color threshold, together with an additional visual inspection of every
216 sample, was used to determine if microplastics were found on the filter.
217

218 For concentration measurements of particles between 0.74 and 5 μm in size, flow cytometry (Attune NxT Flow
219 Cytometer, Thermo Fisher Scientific) was used. The cellulose nitrate filters were digested before analysis with
220 10 % KOH at 60 °C. The pH was corrected for the flow cytometer with HCl. As the flow cytometer can only analyze
221 small volumes and high concentrations of particles, the samples were evaporated in the oven at 60 °C and
222 dissolved in deionized water to increase the concentration. Finally, the samples were filtered over 20 μm filters
223 to remove all big salt crystals that could lead to obstruction of the flow cytometer. The water samples (surface
224 and bulk samples) were also evaporated and dissolved again in deionized water to increase the concentrations.
225 For the flow cytometry analysis, the wells of a 96 well plate with a flat bottom were each filled with 200 μL of
226 sample. For each sample replicate measurements were performed.
227

228 The already red fluorescent plastic particles did not need staining and were analyzed directly (at BL1 (530 nm)
229 and RL1 (670 nm)). The non-fluorescent particles were stained first with Bodipy. The Bodipy 496/503 came from
230 Invitrogen by Thermo Fischer Scientific (catalog code: D3922). A solution of 12.5 $\mu\text{g}/\text{mL}$ of Bodipy dissolved in
231 DMSO was created and 2 μL of this solution was added to each well and analyzed (at BL1 (530 nm) and BL3 (695
232 nm)). Positive control and negative control samples were measured together with the other samples. Control
233 samples were also filtered over a 20 μm filter to make sure that this filter step, used in the preparation of the
234 cellulose nitrate filter samples, does not remove any microplastics.
235

236 The data of the flow cytometer was analyzed using the flowCore package in R (Hahne et al., 2009). As there was
237 no clear pattern observable in the data of the flow cytometer, defining a gating set was challenging. Hence, the
238 mean value from the blank samples was subtracted from the results. This way other particles present in the
239 seawater are subtracted from the results and also the noise created by the cellulose nitrate filters themselves is
240 subtracted. This gave an indication of the number of microplastics present in the samples.
241

242 The smallest particles (nanoplastics) were measured using nanoparticle tracking analysis (Nanosight, Malvern
243 type LM10). The cellulose nitrate filters were digested with 10 % KOH at 60 °C. For the water samples (surface

244 and bulk), no pretreatment was needed. For each measurement, one mL of the sample was sucked up. For each
 245 sample five replicate measurements were performed, each consisting of a video of 60 seconds. Based on these
 246 videos, five size distributions were generated that were combined into one general size distribution of each
 247 sample based on the mean and standard error of the five measurements.

248
 249 To validate the reliability of the results, a second measurement was done of the samples of one replicate
 250 experiment. Different options were studied to determine the optimal settings for the analysis of these
 251 heterogeneous samples. The samples themselves were measured in fluorescence mode instead of scattering
 252 mode to try to only count the fluorescent plastic particles.

253
 254 *Sodium concentration*

255 Na⁺ was extracted from the quartz fiber filters and quantified, together with water samples, using inductively
 256 coupled plasma optical emission spectroscopy (ICP-OES) following the procedures in Van Acker et al. (2021b)
 257 (Supporting information A). Image analysis (Image J) was used to calculate the surface area used for Na⁺ analysis
 258 and to recalculate the measured Na⁺ mass for the full filter, assuming Na⁺ is spread concentrically symmetrical.
 259 The total Na⁺ mass is the sum of the Na⁺ collected on the complete surface area of the filter, the Na⁺ found in
 260 the first rinse of the filter holder and in the second rinse of the filter holder. The Na⁺ mass from the blank filter
 261 and the blank reagent samples were deducted from the results to account for background Na⁺ concentrations.
 262

263 2.6 Calculation of enrichment factors

264 Data from the MNP and Na⁺ analysis is used for the calculation of enrichment factors (EFs) using formulae by
 265 Van Acker et al. (2021b). The enrichment of MNPs in the SSAs and SSML is expressed as enrichment factors and
 266 is determined relative to Na⁺. Na⁺ is a common proxy to quantify SSA densities and is considered to have no
 267 enrichment (Lewis and Schwartz, 2004). That's why in the calculation of the enrichment factors, the sodium
 268 concentration is taken into account. Two types of EFs of MNPs were calculated namely the $[EF]_{SSA}$ and the
 269 $[EF]_{SSML}$, following respectively Equations 1 and 2. The $[EF]_{SSML}$ shows the enrichment of MNPs in the surface
 270 layer compared to the bulk water, while the $[EF]_{SSA}$ shows the enrichment of MNPs in SSAs compared to the bulk
 271 water. Enrichment factors were compared in the different series of experiments in a descriptive manner.
 272

$$273 \quad EF(SSA) = \frac{\frac{[MNP]_{SSA}}{[Na^+]_{SSA}}}{\frac{[MNP]_{bulk}}{[Na^+]_{bulk}}} \quad (\text{Eq. 1})$$

$$274 \quad EF(SSML) = \frac{\frac{[MNP]_{SSML}}{[Na^+]_{SSML}}}{\frac{[MNP]_{bulk}}{[Na^+]_{bulk}}} \quad (\text{Eq. 2})$$

277
 278 $[MNP]_{SSA}$ is the number of aerosolized MNPs collected onto the cellulose nitrate filter per m³ of air sampled
 279 (count/m³). $[Na^+]_{SSA}$ is the amount of sodium collected onto the quartz filter per m³ of air sampled (mg/m³).
 280 $[MNP]_{SSML}$ is the number of MNPs found in the surface layer sample (count/mL). $[Na^+]_{SSML}$ is the concentration
 281 of sodium found in the surface layer sample (mg/mL). $[MNP]_{bulk}$ is the number of MNPs found in the water
 282 column sample (count/mL). $[Na^+]_{bulk}$ is the concentration of sodium found in the water column sample (mg/mL).
 283

284 2.7 Estimate of human exposure to microplastics via inhalation

285 To determine the potential human exposure to microplastics via inhalation of SSAs, the results of the
 286 experiments in the lab were extrapolated to the field (detailed explanation in SI B.2). First, our results were
 287 recalculated to obtain a number of plastics per µg of Na⁺, based on the number of microplastics and mass of Na⁺
 288 on the filters. For this, the data of the three replicate experiments with the TAF particles with a size of 1-5 µm
 289 are used. The focus is on particles between 1 and 5 µm as plastics smaller than 5 µm have the potential to reach
 290 the lungs (Jabbal et al., 2017; Lipworth et al., 2014). Nanoplastics (<1 µm) are excluded given the lack of reported
 291 environmental concentrations.

292 The environmental microplastic concentration data of Everaert et al. (2020) was used to obtain data on
 293 microplastic concentrations in the surface layer of the North Sea. An average was calculated from the
 294 concentrations in the North Sea area, expressed in plastic particles/m³. The data was rescaled to cover the

295 microplastic size range from our experimental setup (from 1 μm -5 mm to 1 μm - 5 μm), based on the formula as
 296 described in Koelmans et al. (2020) and the most recent alfa value for marine surface water (alfa = 2.2) from
 297 Kooi et al. (2021). As it is predicted that the plastic concentrations in the ocean will keep increasing, a prediction
 298 of the future is made using the estimated plastic concentrations in 2050 and 2100 by Everaert et al. (2020).

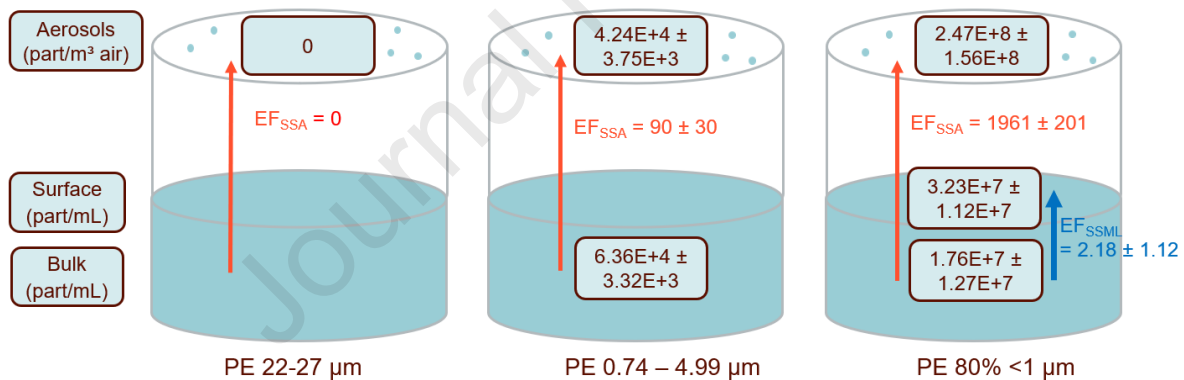
299 Based on these rescaled environmental microplastic concentrations, an extrapolation of the aerosolization of
 300 microplastics in our lab experiments to aerosolization of microplastics in the North Sea was performed. This
 301 results into the number of microplastics that can be found per μg of Na^+ at the coast (*plastic in SSA*). To
 302 determine the number of microplastics that are inhaled by coastal populations, information is needed about the
 303 volume of SSAs in coastal air, Na^+ is used as a proxy for the volume of SSA. Van Acker et al. (2021a) measured
 304 the Na^+ concentrations in the air at the Belgian coast in a 1-year SSA sampling campaign (March 19, 2018 - March
 305 19, 2019; 300 m from the waterline). The results of the SSA concentration range between 0.4 (minimum) and
 306 6.3 (maximum) $\mu\text{g}/\text{m}^3$. On average 1.8 $\mu\text{g}/\text{m}^3$ was collected. An average inhalation rate of 20 m^3 air/day was
 307 used based on Duarte-Davidson et al. (2001). This inhalation rate value is commonly used to determine the
 308 inhaled dose of a given air pollutant for adults. Walking at the coast thus gives an inhalation of 8 to 126 μg of
 309 SSAs per day, with an average of 36 μg of SSAs per day (*SSA rate*). Based on this data, the inhaled microplastics
 310 per day at the coast can be calculated using the following formula:

$$311 \quad \text{inhaled plastics } [\# \text{ plastics/day}] = \text{plastic in SSA} \left[\frac{\# \text{ plastics}}{\mu\text{g SSA}} \right] * \text{SSA rate} \left[\frac{\mu\text{g SSA}}{\text{day}} \right]$$

312 3. Results

313 3.1 Enrichment factors

314 Series 1: Aerosolization in function of size with PE particles



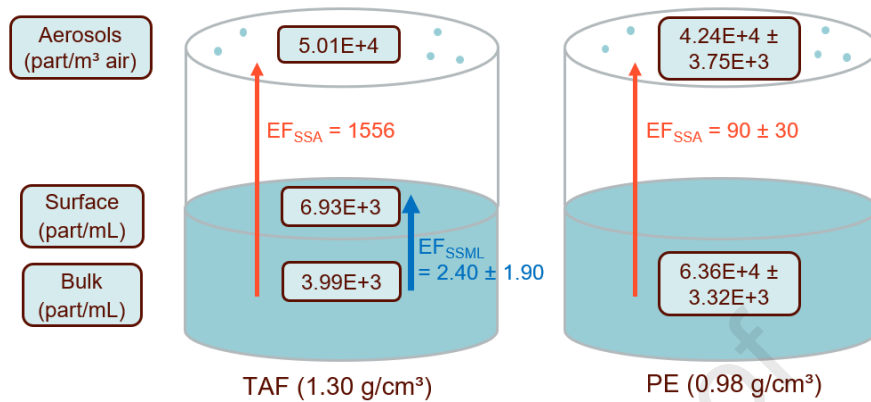
315
 316 *Figure 1: The mean (of the replicates) plastic concentrations in the bulk water samples, surface water samples and aerosol*
 317 *samples for three different sizes of PE particles. The red arrows show the mean enrichment factors of the sea spray aerosols*
 318 *(EF_{SSA}) and the blue arrow shows the mean enrichment factor of the SSML. The EF_{SSML} could not be calculated for every*
 319 *series. For the PE 0.74-4.99 μm particles, the mean [EF]_{SSA} value of 90 ± 30. For replicate experiment 3, the number of*
 320 *particles was lower than the limit of detection and could not be used for the [EF]_{SSA}. If a value of zero is taken for the [EF]_{SSA}*
 321 *of experiment 3, the mean value for the PE 0.74-4.99 μm particles is 59 ± 56. If the limit of detection value is used in the*
 322 *calculation, the mean [EF]_{SSA} becomes 69 ± 41. The mean value of the [EF]_{SSA} will thus be between 59 and 90.*

323 A size dependent effect was observed where the enrichment factors increased with decreasing size (Figure 1).
 324 No 22-27 μm plastics were found on the filters, meaning no aerosolization was observed for PE particles of 22-
 325 27 μm . 10⁶ plastic particles were added to the experimental setup, but none were found on the filter: this leads
 326 to [EF]_{SSA} values which are zero. The PE particles of 0.74-4.99 μm were enriched in the aerosols with a mean
 327 [EF]_{SSA} value of 90 ± 30. For replicate experiment 3, PE particles were found on the filter, but the number of
 328 particles was lower than the limit of detection and could not be used for the [EF]_{SSA}. If a value of zero is
 329 taken for the [EF]_{SSA} of experiment 3, the mean value for the PE 0.74-4.99 μm particles is 59 ± 56. If the limit of
 330 detection value is used in the calculation, the mean [EF]_{SSA} becomes 69 ± 41. The mean value of the [EF]_{SSA}
 331 will thus be between 59 and 90. The [EF]_{SSML} of these experiments could not be calculated as the concentration of
 332 PE particles in the surface water samples was under the detection limit and could thus not be trusted as results.
 333 For the experiments performed with the PE nanoparticles, the mean [EF]_{SSA} value is 1961 ± 201. The mean

334 $[EF]_{SSML}$ is 2.18 ± 1.12 , indicating that the surface layer is enriched in nanoplastics compared to the bulk water,
 335 but the enrichment in the aerosols is a factor 1000 higher. The data used for the calculation of the enrichment
 336 factors can be found in Supporting information B.1.

337

338 Series 2: Aerosolization in function of polymer specific properties with PE and TAF particles



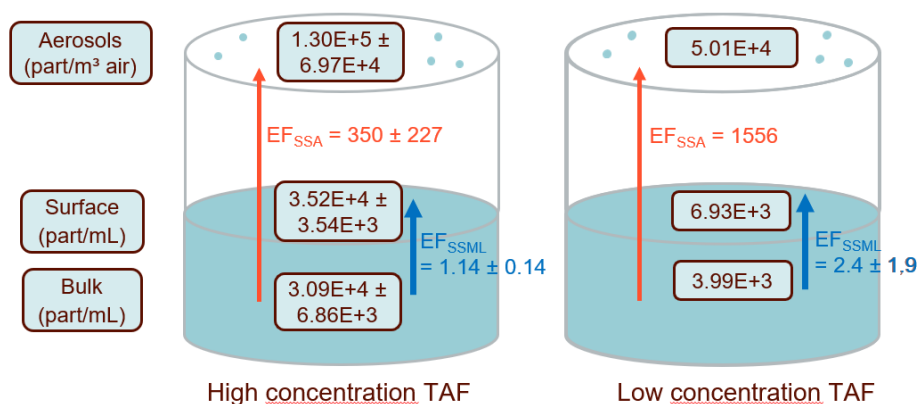
339

340 *Figure 2: The mean (of the replicates) plastic concentrations in the bulk water samples, surface water samples and aerosol*
 341 *samples for two polymer types (TAF and PE). The red arrows show the mean enrichment factors of the sea spray aerosols (EF*
 342 *SSA) and the blue arrow shows the mean enrichment factor of the SSML. The EF_{SSML} could not be calculated for every series.*
 343 *For the experiments performed with the TAF particles of 1-5 μm , the mean $[EF]_{SSA}$ value is 1556. For replicate experiments 1*
 344 *and 2, the number of TAF particles found on the filters was lower than the detection limit and the $[EF]_{SSA}$ could thus not be*
 345 *calculated. If for both experiments, an $[EF]_{SSA}$ value of zero is considered, the mean $[EF]_{SSA}$ is 519. If the limit of detection*
 346 *value is used for the number of particles on the filters, the mean $[EF]_{SSA}$ becomes 759 ± 720 . The mean $[EF]_{SSA}$ value will*
 347 *thus be between 519 and 1556.*

348 The type of polymer had an impact of the enrichment factors as aerosolization was higher for the sinking
 349 particles (TAF) than for the floating particles (PE) (Figure 2). For the experiments performed with the PE particles
 350 of 0.74-4.99 μm , the results are the same as in series 1. For the experiments performed with the TAF particles
 351 of 1-5 μm , the mean $[EF]_{SSA}$ value is 1556. For replicate experiments 1 and 2, particles were found, but the
 352 number of TAF particles found on the filters was lower than the detection limit and the $[EF]_{SSA}$ could thus not be
 353 calculated. If for both experiments, an $[EF]_{SSA}$ value of zero is considered, the mean $[EF]_{SSA}$ is 519. If the limit of
 354 detection value is used for the number of particles on the filters, the mean $[EF]_{SSA}$ becomes 759 ± 720 . The mean
 355 $[EF]_{SSA}$ value will thus be between 519 and 1556. The mean $[EF]_{SSML}$ is 2.4 ± 1.9 . This standard deviation is
 356 influenced by the outlier value of experiment 1. If this value is excluded a mean value of 1.3 ± 0.53 is found. The
 357 $[EF]_{SSML}$ indicates that the surface layer contains twice as much microplastics compared to the bulk water, but
 358 the enrichment in the aerosols is more than a factor 500 higher. The data used for the calculation of the
 359 enrichment factors can be found in Supporting information B.1.

360

361 Series 3: Aerosolization in function of concentration



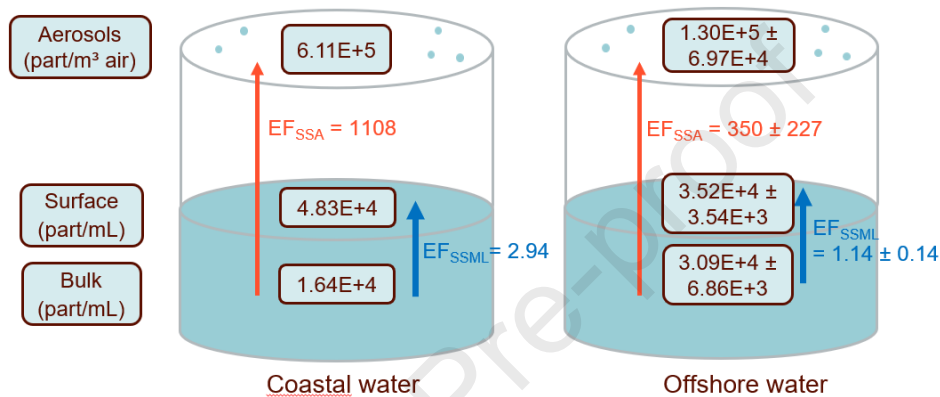
362

363 *Figure 3: The mean (of the replicates) plastic concentrations in the bulk water samples, surface water samples and aerosol*
 364 *samples for two plastic concentrations of TAF particles. The red arrows show the mean enrichment factors of the sea spray*
 365 *aerosols (EF_{SSA}) and the blue arrow shows the mean enrichment factor of the SSML. For the high concentration experiments*
 366 *10^9 plastics were added, while in the low concentration experiments 10^5 plastic particles were added to the experimental*

367 setup (800mL). For the experiments performed with the TAF particles of 1-5 μm , the mean $[EF]_{SSA}$ value is 1556. For replicate
 368 experiments 1 and 2, the number of TAF particles found on the filters was lower than the detection limit and the $[EF]_{SSA}$ could
 369 thus not be calculated. If for both experiments, an $[EF]_{SSA}$ value of zero is considered, the mean $[EF]_{SSA}$ is 519. If the limit of
 370 detection value is used for the number of particles on the filters, the mean $[EF]_{SSA}$ becomes 759 ± 720 . The mean $[EF]_{SSA}$
 371 value will thus be between 519 and 1556.

372 A concentration dependent effect was observed where the enrichment factor was higher if lower concentrations
 373 of microplastics were added (Figure 3). For the experiments performed with high concentrations of TAF the
 374 mean $[EF]_{SSA}$ value is 350 ± 227 . The mean $[EF]_{SSML}$ is 1.14 ± 0.14 , indicating only a small enrichment of
 375 microplastics in the surface layer compared to the bulk water. For the experiments performed with low
 376 concentrations of TAF, the data is the same as the TAF results of series 2 and thus ranges between 519 and 1556.
 377 The data used for the calculation of the enrichment factors can be found in SI B.1.

378
 379 Series 4: Aerosolization in function of type of seawater



380
 381 *Figure 4: The mean (of the replicates) plastic concentrations in the bulk water samples, surface water samples and aerosol*
 382 *samples for two different seawater types: coastal water and offshore water. The tests were performed with TAF 1-5 μm*
 383 *particles. The red arrows show the mean mean enrichment factors of the sea spray aerosols (EF_{SSA}) and the blue arrow shows the*
 384 *mean enrichment factor of the SSML.*

385 A seawater dependent effect was observed (Figure 4): the enrichment factor was higher if coastal water
 386 (including the SSML) was used, compared to offshore water (without the SSML). The $[EF]_{SSA}$ of the experiment
 387 performed with coastal water that included the natural SSML layer is 1108, the $[EF]_{SSML}$ is 2.94. For the
 388 experiments performed with the same polymer type and size of microplastics, but with offshore seawater the
 389 mean $[EF]_{SSA}$ value is 350 ± 227 . The mean $[EF]_{SSML}$ is 1.14 ± 0.14 . The $[EF]_{SSML}$ thus also indicates a higher
 390 enrichment of microplastics in the surface water when coastal water is used compared to offshore water. The
 391 data used for the calculation of the enrichment factors can be found in SI B.1.

392 393 3.2 Estimation of human exposure via inhalation

394 It was estimated that between 6.67×10^{-6} and 1.05×10^{-4} plastic particles with a size between 1 and 5 μm will be
 395 inhaled per day spend at the coast. With increasing plastic production and assumed increasing plastic pollution
 396 in time, the aerosolization is consequently assumed to increase as well. Nonetheless, even in 2100 in the
 397 business-as-usual scenario on plastic production, the estimated daily inhalation of microplastics with a size
 398 between 1 and 5 μm is between 1.51×10^{-4} and 2.37×10^{-3} plastic particles/day (Figure 5). The intermediate results
 399 can be found in SI B.2.

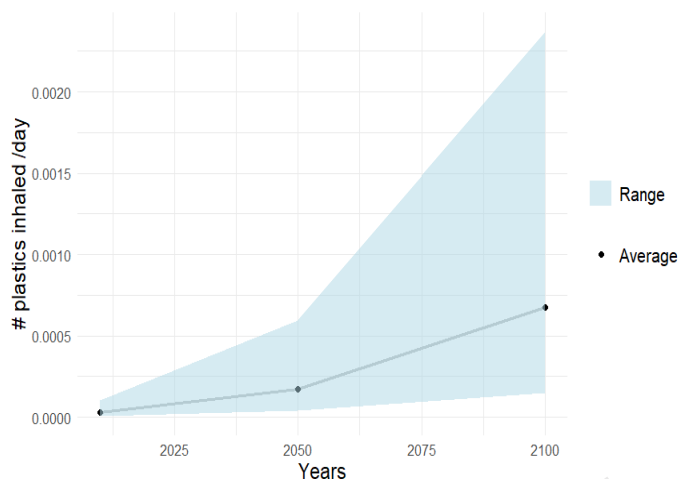


Figure 5: The number of plastics inhaled per day at the coast for 2010, 2050 and 2100. The range indicates the minimum and maximum values.

4. Discussion

The aerosolization of MNPs is a complex process that is influenced by different factors. To estimate the importance of this pathway for human exposure and aerosolization as an MNP transport route, the influence of these factors needs to be investigated. The results of this study highlight that all studied factors here, i.e. MNP characteristics (size, polymer type), concentration and the use of sea surface water, influence the aerosolization process.

Our data indicates that increasing MNP particle size resulted in decreasing aerosolization for PE particles. These results confirm the work of Catarino et al. (2023), showing a similar trend for 0.5, 1, 5, and 10 μm polystyrene particles. Harb et al. (2023) also showed a lower aerosolization for 10 μm PS particles compared to 2 μm particles. Masry et al. (2021) also showed a higher aerosolization for smaller particles but suggested that transfer is limited to particles smaller than 1 μm . The difference with our results can be attributed to the use of natural seawater in our study, while in the study of Masry et al. (2021) ultrapure water was used. In a field sampling study by Kaushik et al. (2024) microplastics in aerosols were reported between 20 μm to 5 mm. A hypothesis is that intense wave action in real ocean environments could cause larger particles to be ejected into the air compared to our experimental setup. In addition, in real environmental conditions, the transfer may depend on particle characteristics but also on the composition of the SSML, the types and concentrations of the present surfactants, as well as the salinity of the water. In future research, it would be useful to be able to measure the size distributions of the particles on the filters and in the water samples. This way, volume based instead of number-based enrichment factors could be calculated and it could be taken into account that within the size range tested, the smaller particles could aerosolize more than the bigger particles.

Second, we observed variability across different polymers. The sinking higher density polymer tested (TAF) showed a higher aerosolization than the floating lower density polymer (PE) tested. Harb et al. (2023) reported that floating PE particles are less effectively aerosolized than PS particles. They reported a significant number of PE aggregates that could be linked to a tendency of PE to aggregate more than PS due to surface properties. This might explain the lower aerosolization of PE particles, which also supports our first results indicating a link between size and aerosolization. It is important to note that in our experimental setup, the bubbles are coming from the bottom. As the denser TAF particles tend to sink, these particles could be taken with the bubbles and interact stronger with the bubbles compared to where a plunging jet might actually partially push them down. In the real environment, different upward and downward forces will act on the particles (gravitational settling, turbulence, the seawater lapping at the surface, currents, ...) and this is not taken into account in our experimental setup. These forces indicate that factors beyond density affect the transport of these materials, potentially leading to their enrichment in the SSML. These results might thus be dependent on the experimental setup used and can be different in real environmental conditions. More research is needed to understand the role of the chemical polymer composition on aerosolization. In addition to the density, other polymer

437 characteristics like hydrophobicity and morphology could have an effect on the aerosolization process and need
438 to be investigated in the future. In this study, we did not distinguish between these specific characteristics.

439 Third, we suggest an effect of concentration. Lower concentrations of microplastics can result in a higher
440 enrichment. A likely reasoning is particle aggregation upon higher concentrations. Wang et al. (2021) showed
441 that aggregation of microplastics is an important physicochemical process dominating the transport behavior
442 and overall fate of microplastics in aquatic environments. This aggregation can occur between microplastics
443 (homoaggregation) or between microplastics and different types of particles (like organisms and organic
444 material) (heteroaggregation). This potential aggregation of microplastics in natural seawater might be a reason
445 for the lower aerosolization at a higher microplastic concentration, as also suggested by Harb et al. (2023). Due
446 to the lower environmental MNP concentrations, mostly heteroaggregation is expected in the field, while in our
447 experimental setup higher concentrations of plastics are used, possibly also resulting in homoaggregation. Other
448 studies also show a higher enrichment in the SSML if the concentrations in the bulk seawater are lower. Higher
449 enrichment was observed for DOC and POC in the SSML, when low concentrations were present in the bulk
450 seawater (van Pinxteren et al., 2012; van Pinxteren et al., 2017). The average EF SSA for TAF 1-5 μm low
451 concentrations is in the range between 519 and 1556. The three experiments show a high variability in EF SSA
452 values. For low concentrations, it can depend on the spatial location of the low number of particles, if they are
453 incorporated in the aerosols. This spatial variation is likely higher at lower concentrations than at higher
454 concentrations leading to more variation in the EF in low concentrations versus high concentration where
455 particles are likely more homogeneously distributed. This could result in a large variation of number of particles
456 in the aerosols and thus in the EF SSA. Further research is needed with realistic polymer mixtures in realistic
457 concentrations, which include among others other natural particles.

458 Fourth, seawater characteristics affect the aerosolization process. Here, we compared natural seawater
459 collected in two ways. Offshore seawater is taken at a depth of 3 m, so no natural SSML layer was sampled. The
460 water taken at the coast included the natural SSML. Our results indicate that the coastal water, with the SSML
461 included, contributes to higher aerosolization of microplastics. The SSML can namely be enriched with organic
462 matter, bioactive molecules, organisms and surfactants, which could influence the aerosolization process (Casas
463 et al., 2020; Flores et al., 2021; Lv et al., 2020). The biogenic surfactant dipalmitoylphosphatidylcholine (DPPC),
464 is for example known to accumulate in the SSML (Van Acker et al., 2021a). In general, the use of natural seawater
465 compared to artificial seawater in aerosolization experiments is suggested to have an effect. Studies using
466 artificial seawater (Catarino et al., 2023; Shiu et al., 2022) showed much lower enrichment factors. Catarino et
467 al. (2023) expects that the presence of microorganisms in the SSML will augment the enrichment factors for
468 MNPs. Yang et al. (2022) on the other hand suggested in their experiments that microplastic transfer processes
469 with natural seawater can be realistically reproduced with synthetic seawater. They, however, filtered the
470 natural seawater over 4.5 μm , removing a lot of the microorganisms. This filtering step will affect the SSML
471 composition and the aerosolization process. Further research is thus needed to elucidate how the biochemical
472 composition of the SSML and seawater in general influences aerosolization.

473 The MNPs in sea spray aerosols can be inhaled by coastal populations. However, it is unclear how much this
474 pathway contributes to the human exposure to MNPs via inhalation. Here, we made a first step to assess
475 potential human exposure by extrapolating results from the laboratory setting to field. Our first estimate showed
476 that between 6.67×10^{-6} and 1.05×10^{-4} plastic particles with a size between 1 and 5 μm will be inhaled per day
477 spend at the coast. This is negligible compared to reported inhalation exposure in urban context where estimates
478 showed an inhalation of 1.5 particles per m^3 and indoor, where even higher concentrations of 60 particles per
479 m^3 are inhaled (UN, 2021). Our results show a large variation due to the weather influence on the number of
480 aerosols formed. With increasing plastic production and assumed plastic pollution over time, the aerosolization
481 is consequently assumed to increase as well. Nonetheless, following the business-as-usual scenario on plastic
482 production, the daily inhalation in 2100 via SSAs of 1-5 μm microplastics is estimated to still be far below 1
483 particle per day. Furthermore, both urban and indoor particle pollution will similarly increase under the
484 business-as-usual scenario. Based on our first estimate, sea spray aerosols might not relevantly contribute to
485 the total plastic exposure concentration by inhalation.

486 In most plastic-aerosol experiments (including our experiment) MNP concentrations of 10^4 - 10^6 particles/mL are
487 used (Shiu et al., 2021; Yang et al., 2022). Higher MNP concentrations are used in the experiments than in the
488 real ocean situation (18 particles/m³ in 2010, see SI B.2). This is because the concentrations need to be high
489 enough for analytical detection limits. The results of series 3 show that lower concentrations of microplastics
490 result in higher aerosolization levels for TAF particles, but this could thus not be taken into account in the
491 extrapolation. To be able to take concentration into account, concentration response curves are crucial to
492 determine the relationship between the concentration in the water and the number of plastics in the aerosols.
493 Currently, due to the lack of environmental concentrations of nanoplastics and a lack of knowledge on how they
494 behave, it is impossible to do a similar extrapolation for nanoplastics.

495 Although aerosolization of MNPs via sea spray aerosols might not relevantly contribute to human microplastic
496 exposure via inhalation, this process is still important to take into account in the source- sink dynamics of the
497 plastic cycle (Bank, M. S., & Hansson, S. V.; 2019). Indeed, the ocean is not only a sink for MNPs but also a source.
498 This process also contributes to understanding the plastic fluxes to remote regions (Bergmann et al., 2019; Allen
499 et al., 2019). Furthermore, marine aerosols play a vital role in the Earth system and influence the radiation
500 balance of the Earth by scattering and absorbing solar radiation (Rastelli et al., 2017; Schiffer et al., 2018). The
501 potential impact of the presence of MNPs in these aerosols remains to be elucidated.

502 Conclusion

503 This study investigated the transfer of MNPs to the atmosphere via SSAs. The results showed that aerosolization
504 of MNPs is influenced by multiple factors including size, polymer type, concentration and composition of the
505 seawater. Future research on MNP aerosolization should thus take these different factors into account to
506 resemble realistic plastic pollution situations. A first estimate of human exposure to microplastics via the
507 inhalation of SSAs shows that this pathway is negligible for human exposure compared to exposure to
508 microplastics in outdoor urban environments and indoor air. An increase of the exposure of microplastics via
509 SSAs is expected in the future due to increasing plastic pollution. This study shows that aerosolization is a new
510 plastic transport pathway to take into account, but that human exposure to microplastics via SSAs seems
511 negligible compared with other sources.

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517 Competing interest

518 No competing interest.

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Highlights

- Aerosolization increased with decreasing Polyethylene particle size
- Plastic concentration and polymer type influence plastic enrichment in aerosols
- Human plastic exposure via sea spray seems negligible compared to urban and indoor

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Declaration of interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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