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Hazardous contaminants in plastics contained in compost and agricultural soil / Scopetani, Costanza; Chelazzi, David; Cincinelli, Alessandra; Martellini, Tania; Leiniö, Ville; Pellinen, Jukka. - In: CHEMOSPHERE. - ISSN 0045-6535. - 293 133645:(2022), pp. 1-9. [10.1016/j.chemosphere.2022.133645]

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30/04/2026 04:15

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Journal Pre-proof

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PII: S0045-6535(22)00138-2

DOI: <https://doi.org/10.1016/j.chemosphere.2022.133645>

Reference: CHEM 133645

To appear in: *ECSN*

Received Date: 11 October 2021

Revised Date: 7 January 2022

Accepted Date: 13 January 2022

Please cite this article as: Scopetani, C., Chelazzi, D., Cincinelli, A., Martellini, T., Leiniö, V., Pellinen, J., Hazardous contaminants in plastics contained in compost and agricultural soil, *Chemosphere* (2022), doi: <https://doi.org/10.1016/j.chemosphere.2022.133645>.

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Credit authorship contribution statement

Costanza Scopetani – Experimental design, Sampling, Samples treatment, Measurements, MPs characterization, Data analysis and interpretation, Figures, Writing of the article

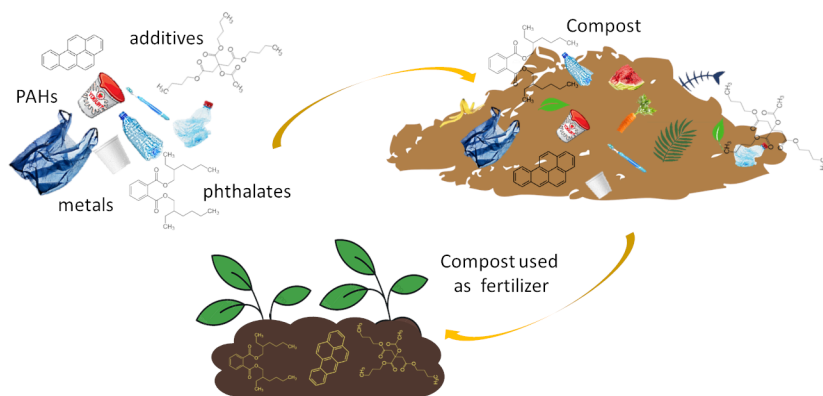
David Chelazzi – MPs characterization, Data analysis and interpretation, Revising Text and Figures

Tania Martellini – Revising Text and figures

Alessandra Cincinelli– Revising Text and Figures

Ville Leiniö – Macroplastics characterization

Jukka Pellinen – Experimental design, Data discussion and Revising Text and Figures



1 **Hazardous contaminants in plastics contained in compost and** 2 **agricultural soil**

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12

13 **Abstract**

14 Macro-, meso- and microplastic (MAP, MEP, MP) occurrence in compost is an environmental issue
15 whose extent and effects are not yet understood. Here, we studied the occurrence of MAPs, MEPs
16 and MPs in compost samples, and the transfer of hazardous contaminants from plastics to compost
17 and soil. MAPs/MEPs and MPs concentrations in compost were 6.5 g/kg and 6.6±1.5 pieces/kg; from
18 common recommendations for compost application, we estimated ~4-23x10⁷ pieces MPs and 4-
19 29x10⁴ g MAPs/MEPs ha⁻¹ per year ending into agricultural soils fertilized with such compost.
20 Regarding contaminants, bis(ethylhexyl) phthalate, acetyl tributyl citrate, dodecane and nonanal were
21 extracted in higher concentrations from plastics and plastic-contaminated compost than from compost
22 where MAPs/MEPs had been removed prior to extraction and analysis. However, some contaminants
23 were present even after MAPs/MEPs removal, ascribable to short- and long-term release by
24 MAPs/MEPs, and to the presence of MPs. DEHP concentration was higher in soils where compost
25 was applied than in fields where it was not used. These results, along with estimations of plastic load
26 to soil from the use of compost, show that compost application is a source of plastic pollution into
27 agricultural fields, and that plastic might transfer hazardous contaminants to soil.

28

29 **1. Introduction**

30 Plastic is one of the most used and produced materials in the world (Geyer et al., 2017, Plastic Europe
31 2020), and has been pointed in the last decades as an emerging pollutant by the scientific community
32 (Carpenter & Smith, 1972; Thompson et al., 2009). Plastic debris is commonly classified in four main
33 categories according to their size, i.e. macroplastics (MAPs ≥ 25 mm) mesoplastics (MEPs, 5-25 mm)
34 (U.S. Environmental Protection Agency, 2011), microplastics (MPs, 1-5000 μm) (Hartmann et al.,
35 2019) and nanoplastics (<1 μm) (Sendra et al., 2020). Plastics are ubiquitous pollutants, found in the
36 atmosphere, marine, freshwater and terrestrial environments and inside living organisms (Bläsing &
37 Amelung, 2018; Gago et al., 2018; Gasperi et al., 2018; Ivleva et al., 2017; Scopetani et al., 2018;
38 (Piarulli et al., 2019; Scopetani et al., 2020). However, while several studies have focused on MPs
39 aquatic pollution (Avio et al., 2017; Bergmann et al., 2015; Cincinelli et al., 2021; Ivleva et al., 2017;
40 Li et al., 2018; Scopetani et al., 2019; Suaria et al., 2020), very little is known about their presence
41 and effects in terrestrial environments (de Souza Machado et al., 2018; Fuller & Gautam, 2016;
42 Kuoppamäki et al., 2021; Scheurer & Bigalke, 2018).

43 The knowledge gap is even larger when it comes to data about the amount of plastics and associated
44 contaminants that agricultural fields receive by the application of recycled fertilizers, like bio-waste
45 and sewage sludge compost (Bläsing & Amelung, 2018; Braun et al., 2021; Weithmann et al., 2018;
46 Zhang et al., 2020). Plastics end up in bio-waste compost due to improper waste disposal and waste
47 management, such as the use of non-biodegradable plastic bags for bio-waste collection (Bläsing &
48 Amelung, 2018; Braun et al., 2021). Sewage sludge retains MPs from water in plants (Carr et al.,
49 2016; Estahbanati & Fahrenfeld, 2016; Mason et al., 2016; Mintenig et al., 2017), and ends up
50 containing around 1000–24,000 plastic items kg^{-1} (Du et al., 2020; Mahon et al., 2017; Mintenig et
51 al., 2017); when sewage sludge is used into agricultural soil, a yearly load of 63,000–430,000 and
52 44,000–300,000 tons of MPs may end up to European and North American agricultural fields,
53 respectively (Nizzetto et al., 2016). The use of sewage sludge exceeding the legal limits in terms of

54 harmful substances is prohibited but there is no indication about MPs in the EU 86/278/EEC
55 regulation nor in the Code 503 of the USA (Nizzetto et al., 2016). Even in Germany, which has,
56 according to Braun et al. (2021), one of the strictest regulations globally regarding recycled fertilizers
57 (“Düngemittelverordnung”), particles smaller than 2 mm are not regulated.

58 Overall, considering that general recommendations in agricultural practices suggest a compost
59 application range from 7 to 35 t compost ha⁻¹ for agricultural fields, and from 6.48 to 19.44 t ha⁻¹ for
60 horticultural soils, Braun et al., (2021) estimated a load of plastics from compost ranging between
61 84,000-1,610,000, and 77,770-894,240 plastic items ha⁻¹ per year, respectively.

62 Plastic pollution risks are not only linked to soil alteration (de Souza Machado et al., 2018; Wan et
63 al., 2019) and impact on biota (Lin et al., 2020), (Huerta Lwanga et al., 2016; Lei et al., 2018; Zhu et
64 al., 2018) but are also connected to the adsorbed toxic substances that polymers may transport and
65 release during their life cycle. Some of the plasticizers, antioxidants, pigments, flame-retardants and
66 other additives contained by plastic materials, pose a hazard to the environment and human health
67 (Hahladakis et al., 2018). Besides, hydrophobic organic pollutants tend to sorb on plastics from the
68 environment (Hüffer et al., 2018), and might be transported and released to other habitats (Bergmann
69 et al., 2015), or into organisms (Browne et al., 2013; Scopetani et al., 2018). Given the number of
70 plastics that agricultural fields receive by the application of recycled fertilizers, and considering the
71 lack of regulations on plastic content in the latter, it is essential to understand the impact that plastics
72 pollution has on the terrestrial environment, evaluating the possible output of hazardous compounds
73 from plastics.

74 Coping with these issues, the present research aims to study the occurrence of plastics and MPs in
75 compost and soil samples, as well as the transfer of contaminants from plastics in the compost to the
76 compost itself and to soil. The investigated contaminants were selected based on their documented
77 presence in plastics and on their potential environmental and human toxicity (Cruz, 2013), and
78 comprised PAHs (polycyclic aromatic hydrocarbons), phthalates (especially bis(2-ethylhexyl)

79 phthalate, DEHP), acetyl tributyl citrate (ATBC), cobalt, cadmium and lead, as well dodecane and
80 nonanal (which should not be in plastics but were found in the samples during preliminary screening).
81 PAHs are a group of compounds considered mutagenic and/or carcinogenic (Andersson & Achten,
82 2015) and plastic can be listed as a source of these contaminants since PAHs have been found in
83 virgin polystyrene foam with a concentration ranging from 79 to 97 ng/g (Coffin et al., 2020). Plastic
84 can also sorb PAHs from the environment; Indeed, post-consumer plastics fragments were collected
85 in selected ocean sites in California, Hawaii and Mexico to be analyzed for organic contaminants and
86 the total concentration of PAHs ranged from 39 to 1200 ng/g (Rios et al., 2007). Prenatal exposure to
87 Benzo[a]pyrene (B[a]P), recognized as one of the most toxic PAHs, impairs brain development
88 (McCallister et al., 2008); when B[a]P is inhaled by male adult rats, it significantly reduced the
89 components of the steroidogenic and spermatogenic compartments of the testis, decreases testis
90 weight, and reduces plasma total testosterone concentration (X. Chen et al., 2011; Ramesh et al.,
91 2008). PAHs presence in soils is a serious environmental concern so that in the European Union the
92 cost for soil remediation from PAH is estimated to be up to two billion euros (Luo & Schrader, 2021).
93 The same applies to heavy metals, of which soil is a major sink, that is cytotoxic and able to cause
94 adverse effects on organisms, even at a low concentration level (Long et al., 2021; Lu et al., 2010).
95 Pb, Sn, Ba, Cd, Co, Cu and Zn are commonly added in plastic products as heat stabilizers or organic
96 pigments (Hahladakis et al., 2018).

97 DEHP is a widely used plasticizer, especially in the production of polyvinylchloride; it forms non-
98 covalent bonds with the polymers and thus, its migration is facilitated over time (Sun et al., 2022).
99 DEHP is recognized as an endocrine disruptor, able to impair the reproduction system and to affect
100 kidney, testicular, ovary, renal and liver function (Liu et al., 2021). Liu et al. (2021) showed that
101 DEHP affects ovarian hormone production and antral follicle development of offspring in lactating
102 mice, while Sun et al. (2022) demonstrated that DEHP exposure to mice disrupts placental growth.
103 Furthermore, it seems that prenatal low-dose DEHP exposure could induce later obesity and

104 metabolic syndrome (Fan et al., 2020). ATBC is a common plastic additive present in food, medical
105 toys and cosmetic plastics, able to leach 10 times more rapidly than DEHP (Malarvannan et al., 2019;
106 Rasmussen et al., 2017). ATBC was born as a safer and more environmentally friendly alternative for
107 phthalates in plastic products, but some toxicology studies showed that it might produce detrimental
108 effects on the ovary of mice and suggested that further studies are needed to deepen its impact on the
109 reproductive system (Rasmussen et al., 2017).

110 Dodecane is a major fuel component (Herbinet et al., 2007) and it is not used as a plastic additive but
111 it was found on plastics recovered from marine waters (Rios et al., 2007) and it shows a strong affinity
112 for polyethylene (Castleman et al., 2021).

113 Nonanal is used as a flavor agent and similarly to dodecane it should not be contained in plastics but
114 it was found in cling-films for retail use in a concentration ranging from 46.29-66.48 $\mu\text{g/g}$ (Panseri
115 et al., 2014) and in plastic debris collected from coastal beaches in South Korea (Rani et al., 2015)

116 In the present study, we analyzed plastic pollution in compost made of bio-waste and sewage sludge,
117 and soil samples utilizing Fourier transform infrared spectroscopy (FTIR), and then the contaminants
118 were extracted and quantified using gas chromatography-mass spectrometry, to determine if plastics
119 contained in the compost transfer associated contaminants to the soil. We coupled our experimental
120 results with current data on plastic pollution in compost and compost application to soils, providing
121 evaluations for the overall impact of contaminants potentially transferred from compost. To the best
122 of our knowledge, this is the first time that such an estimation is carried out, and we hope that our
123 data might also provide the basis for following up studies that will have to check if these hazardous
124 substances move further up to the top of the food chain, possibly posing risks for human health.

125

126 **2. Experimental**

127 2.1 Sampling

128 A Finnish waste treatment company that collects and treats bio-waste from households, restaurants
129 and industry, and sludge from wastewater treatment plants, provided the compost samples. Their
130 product is a mixture of bio-waste and composted sewage sludge. The name of the company cannot
131 be given because of anonymity reasons. The biowaste to the composting plant comes from
132 households, restaurants, grocery stores, and food industry. The compost, after going through the
133 hygienisation and maturation of composting steps, is transferred to outdoor piles and kept there up to
134 12 months. There is a steady stream of material to the composting plant throughout the year. Since
135 the quality of biowaste and sewage sludge is relatively constant and the maturing period is so long,
136 seasonal variations are then considered to be low.

137 Soil samples were collected in November 2020, at the same time as the compost, from four fields in
138 rural areas in Orimattila and Kärkölä, Finland. Two of the selected fields, “BeanC” and “BarleyC” (a
139 horse bean and a barley field), were fertilized in 2020 with the compost produced by the same
140 company that provided us the compost for all the analyses. The fields have been fertilized once a
141 year. A third field, “BarleyS” (a second barley field) was fertilized some years ago with sludge from
142 a wastewater treatment plant in Helsinki. No fertilizers have been used in the fourth field, “Green
143 pea” (pea cultivation). Detailed information about the locations cannot be given because of anonymity
144 reasons. No detailed information about the amount of fertilizers applied to the fields was available.
145 All the samples were collected using a metal shovel and kept in metal buckets previously rinsed with
146 ultrapure water. Soil and compost samples were preserved in a cold room at 5 °C prior to analysis.

147

148 2.2 Chemical reagents

149 Phthalate mixture (EPA 506 Phthalate Mix) was purchased from Merck (Darmstadt, Germany), the
150 PAH mixture (naphthalene (NAP), chrysene (CHR), anthracene (ANT), and benzo[a]pyrene (B[a]P))
151 from Phenova (Denver, USA), while dodecane, nonanal and acetyl tributyl citrate (ATBC) from TCI
152 Europe (Zwijndrecht, Belgium). Deuterated solutions of DEHP-d₄ (Sigma-Aldrich), dodecane-d₂₆

153 (Toronto Research Chemicals), acetyl tributyl citrate-d₃ (Toronto Research Chemicals), chrysene-d₁₂
154 (Phenova) were used as internal standards. Metal standards for Al, As, Be, Cd, Co, Cr, Cu, Fe, Mn,
155 Ni, Pb, Se, V and Zn and the internal standard (In) were purchased from VWR International, as well
156 as hexane and acetone. Glass microfiber filters (GF/A, 45 mm diameter, Whatman) were used to filter
157 the samples after the extraction.

158

159 2.3 Extraction and analysis of organic contaminants

160 A portion of the compost was sieved with a 5 mm mesh metal sieve to remove MAPs and MEPs and
161 collect them for further analyses; henceforth, the acronym for the portion of compost without
162 MAPs/MEPs is CompostW/O while the acronym for the compost left with MAPs and MEPs is
163 CompostW.

164 Five replicates of the soil samples (BeanC, BarleyC, BarleyS and Green pea), of CompostW/O,
165 Compost/W and the MAPs/MEPs were analyzed for DEHP, dodecane, nonanal, ATBC, CHR, ANT,
166 B[a]P, and NAP determination.

167 2 g of each replicate was extracted following the procedure described by Aparicio et al. (2007), with
168 slight modifications. Briefly, the samples were lyophilized and transferred to 50 ml glass bottles, and
169 then 20 ml of hexane was added. The bottles were stirred for 30 minutes (180 rpm) and then sonicated
170 for 60 minutes. The extraction methodology was repeated thrice. Internal standards were added, and
171 then the combined extracts were filtered through glass fiber filters and evaporated with a gentle flow
172 of nitrogen down to 1 mL in a volumetric flask.

173 The samples were then analyzed with gas chromatography–mass spectrometry (Shimadzu GC–MS-
174 QP2010 Ultra) system equipped with an AOC-20i autoinjector and a 30-m ZB-5MS column (0.25
175 mm i.d., 0.25 µm film thickness). The instrument operation conditions were as follows: 250 °C
176 injection temperature, split-less injection mode, 1 µl injection volume, He carrier gas. The
177 temperature program was initially 60 °C hold for 1 min, ramped at 10 °C min⁻¹ to 280 °C and

178 maintained for 6 min. The recovery range of the target compounds was 95-105%. The instrumental
179 limit of quantification (LOQ) was 3.4 ng/g for ANT, 7 ng/g for CHR, 2.4 ng/g for NAP, 8.8 ng/g for
180 B[a]P, 173.6 ng/g for DEHP, 44.1 ng/g for ATBC, 8.7 ng/g for dodecane, and 95.3 ng/g for nonanal.
181 The limit of detection (LOD) was 1 ng/g for ANT, 2.1 ng/g for CHR, 0.7 ng/g for NAP, 2.7 ng/g for
182 B[a]P, 52.6 ng/g for DEHP, 13.4 ng/g for ATBC, 2.6 ng/g for dodecane, and 28.9 ng/g for nonanal.

183

184 2.4 Extraction and analysis of metals

185 CompostW/O, Compost/W, and MAPs/MEPs samples were analyzed in five replicates for Al, As,
186 Be, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, V and Zn, via inductively coupled plasma mass
187 spectrometry (Perkin–Elmer Elan DRC II ICP-MS). Acid digestion was performed following an
188 adapted version of the method US EPA 3050B (Vedolin et al., 2018). Briefly, 200 mg of CompostW/
189 CompostW/O and MAPs/MEPs samples were digested with 15 ml of concentrated nitric acid and
190 5 ml of H₂O₂ in a Mars 6 microwave device (CEM corporation) (plant material method). 1 ml of the
191 sample was then diluted with 4 ml of ultrapure water, and 50 µl of indium (1 mg l⁻¹) was added to
192 the diluted sample as the internal standard.

193

194 2.5 Extraction of MAPs/MEPs and MPs

195 MAPs and MEPs were extracted from the compost, sieving 2 kg of compost with a 5 mm mesh size
196 sieve. MAPs and MEPs were visually identified, collected, weighted, and analyzed with FTIR
197 spectroscopy.

198 MPs were extracted from 5 replicates following the method described by Scopetani, et al. (2020).
199 Briefly, 10 g of compost for each replicate was mixed with ultrapure water in polytetrafluoroethylene
200 (PTFE) cylinders. 3 mL of olive oil were added, and after shaking the systems were left to settle for
201 2 hours before being frozen at -40 °C. The ice columns, and the oil layers, were pushed out to filtering

202 funnels and filtered through glass microfiber filters (GF/A, 90 mm diameter, Whatman). After
203 removing the oil traces by rinsing the filters with hexane, the filters were dried in a desiccator and
204 analyzed with FTIR spectroscopy. Each replicate underwent the extraction thrice to maximize the
205 recovery.

206

207 2.6 FTIR-ATR analysis

208 MAPs and MEPs (particle sizes from 5 mm to 15 cm) were investigated with an Agilent Cary 630
209 FTIR Spectrometer equipped with a diamond crystal ATR (Attenuated Total Reflection) unit. The
210 analyses were carried out in the 4000-650 cm^{-1} spectral range, with a spectral resolution of 4 cm^{-1} ,
211 acquiring 32 scans for each spectrum in absorbance mode.

212

213 2.7 Microscope FTIR analysis

214 For the analysis of MPs, the dried filters were investigated through 2D imaging FTIR, using a Cary
215 620-670 FTIR microscope equipped with an FPA (Focal Plane Array) 128×128 detector (Agilent
216 Technologies). This instrument allows FTIR analysis to be carried out directly on the MPs-containing
217 filters, with no pre-treatment. FPA detectors are widely used for the detection of MPs, thanks to their
218 high spatial resolution (Andrades et al., 2018; Harrison et al., 2012; Mintenig et al., 2017; Scopetani
219 et al., 2020; Simon et al., 2018; Tagg et al., 2015). We operated the system in reflectance mode using
220 an open aperture and a spectral resolution of 8 cm^{-1} , acquiring 128 scans for each spectrum. Each
221 analysis produces a map of $700 \times 700 \mu\text{m}^2$ (128×128 pixels), where each pixel has a dimension of
222 $5.5 \times 5.5 \mu\text{m}^2$ and provides an independent spectrum. The detection limit of the FPA detector is in
223 the order of $0.02 \text{ pg}/\mu\text{m}^2$ (Mastrangelo et al., 2020). On each filter, MPs were detected and identified
224 in five randomly chosen squares ($2 \times 2 \text{ cm}^2$), so as to cover 31.4% of the filter area.

225

226 2.8 Contamination control

227 To avoid contamination from organic contaminants, especially from phthalates, all the glassware was
228 rinsed with ultrapure (18.2 M Ω) water, hexane and acetone, and then heated at 200 °C overnight. The
229 risk of MPs self-contamination was also taken into account: items and clothes able to release MPs
230 were avoided, according to Scopetani et al., (Scopetani et al., 2020), both during sampling and
231 analyses. All the tools, including the metal buckets, were rinsed with ultrapure (18.2 M Ω) water
232 before covering them with aluminum foil. Field and laboratory blanks were set up in parallel with the
233 samples to check for potential airborne contamination. No MPs were found in any of the blanks.
234 Procedural blanks for the organic contaminants and metals were performed throughout all steps of
235 the analysis to check for laboratory contamination and interferences. Furthermore, to avoid phthalates
236 contamination, all tools and glassware were rinsed first with ultrapure (18.2 M Ω) water, then acetone
237 and hexane. Since the analytical procedure was free of contamination, no procedural blank correction
238 was applied.

239

240 2.9 Statistical analysis

241 IBM SPSS Statistics version 25 (2017) was used to performed statistical analysis. All data were
242 analyzed with Shapiro-Wilks and Levene's test to check for normality and homogeneity. One-way
243 analysis of variance (ANOVA), followed by Tukey's test, was employed when the data were
244 normally distributed. For not normally distributed data, Dunnett's C test was used to detect
245 differences amongst the treatments. Data were divided in two distinct sets, soil samples (BeanC,
246 BarleyC, BarleyS and Green pea), and compost or plastic samples (CompostW/O, Compost/W and
247 MAPs/MEPs). The two sets were statistically analyzed separately. The results were considered
248 significant at a p value of 0.05.

249

250 **3 Results and Discussion**

251 3.1 Organic contaminants

252 MAPs/MEPs displayed the highest contaminant concentrations in comparison to soil and compost
253 samples, clearly indicating that plastics are a possible source of pollutants to the compost, and then
254 to agricultural soil. The results are grouped below according to the pollutants. The mean concentration
255 of each contaminant is reported in Table 1.

256

257 PAHs (Polycyclic aromatic hydrocarbons)

258 Naphthalene, chrysene and benzo[a]pyrene concentrations were below the detection limit (LOD) in
259 all samples analyzed. ANT, which has a mean concentration of 651 ± 84 ng/g dw in MAPs/MEPs, is
260 also found at lower concentrations in CompostW and CompostW/O (~110 ng/g dw each, with no
261 significant differences, e.g. $p=0.96$), while it was below LOQ in all the soil samples.

262 The sum of analyzed PAHs in compost samples is largely below the limit of 6 mg/kg of compost set
263 by the Regulation EU 2019/1009 (Regulation EU, 2019), even if this limit concerns the sum of all
264 the 16 PAHs, and our data refer to only four compounds. Brändli et al., (2006) analyzed PAH contents
265 of compost from kitchen and green waste in Switzerland and found PAH concentrations up to four
266 orders of magnitude higher than those detected in the compost analyzed in this study (that is a
267 combination of a mixture of bio-waste and composted sewage sludge). PAHs concentrations in our
268 compost samples were abundantly lower than those found in Poland from raw sewage sludge
269 (Oleszczuk, 2007, 2009) but slightly higher than those detected in composted sewage sludge in Japan
270 by Ozaki et al., (2017).

271

272 DEHP (Bis(2-ethylhexyl) phthalate)

273 DEHP was found in MAPs/MEPs and, at lower concentrations, in all compost and soil samples except
274 for Green pea and BarleyS where the concentrations were lower than the LOD (52.6 ng/g). BarleyC

275 and BeanC samples showed a DEHP concentrations of 931 ± 163 ng/g dw and 1080 ± 209 ng/g dw
276 respectively, and no statistically significant difference was found between them ($p=0.77$).

277 A statistically significant difference was found between Compost/W and CompostW/O ($p=0.043$),
278 where the compost samples with MAPs/MEPs presented, as expected, higher DEHP concentrations.
279 MAPs and MEPs had the higher DEHP concentration with an average of 38200 ± 33900 ng/g dw.
280 Overall, our data point to plastics as one source of DEHP in agricultural soils.

281 The Regulation EU 2019/1009 (Regulation EU, 2019) does not include limitations of DEHP or other
282 phthalates in compost, but concerns have been expressed about the presence of these contaminants in
283 compost and fertilizers (Huygens et al., 2019). DEHP is one of the most common phthalates added
284 as softeners to plastics products and is often found in concentrations exceeding the limit value of 100
285 mg/kg fixed by the EU standard for the land application of DEHP containing sewage sludge (Aparicio
286 et al., 2009; Santos et al., 2007). It is recognized as a persistent organic contaminant and an endocrine
287 disruptor able to cause adverse health effects in organisms (Langdon et al., 2019; Sandeep &
288 Rowdhwal, 2018). As far as we know the Danish Decree for the agricultural use of sewage sludge
289 and waste-derived compost is the only regulation that establishes threshold values for DEHP (50
290 mg/kg) in bio-waste compost. On the contrary, the concentration of DEHP in sewage sludge is
291 regulated by the EU standard for the land application of sewage sludge. The DEHP concentrations
292 we found in compost samples did not exceed the limit value of 100 mg/kg and were lower than those
293 found in the sewage sludge compost produced by a Spanish waste water treatment plant (range 24-
294 124 mg/kg dw and mean 75 mg/kg dw) (Aparicio et al., 2009), but higher than bio-waste compost
295 analyzed by Brändli et al., (2007) ($\sim 280 \mu\text{g kg}^{-1}\text{dw}$). For what concerns soils, our data are comparable
296 to those found by Wang et al., (2013) in suburban vegetable soils in Nanjing (China), but higher than
297 those detected in agricultural soils in the Paris area fertilized with sewage sludge (mean $134 \mu\text{g/kg}$)
298 (Tran & Teil, 2015).

299

300 ATBC (Acetyl tributyl citrate)

301 As for DEHP, we found ATBC in all samples, the highest values expectedly being in MAPs/MEPs
302 (1100±105 ng/g dw). Similarly to what found for DEHP, ATBC concentrations in Green pea and
303 BarleyS samples were below the LOQ (44.1 ng/g). There was no statistically significant difference
304 between Green pea and BarleyS ($p=0.597$). Regarding the compost, ATBC concentration in
305 CompostW was below the LOQ but higher than the LOD (13.4 ng/g), while CompostW/O samples
306 showed an ATBC concentration below the LOD. As for DEHP, the results seem to indicate that
307 plastic debris could transfer ATBC to compost, and later on to soil.

308 As far as we know, the ATBC concentration in compost and agricultural soils is not regulated by any
309 European regulation and there are very few research studies, if any, on its presence in compost and
310 soil. This is probably due to the fact that ATBC is classified as a non-toxic additive (Arrieta et al.,
311 2014; Johnson, 2002), and considered systemically safe up to 1000 mg kg⁻¹ day⁻¹ (Rasmussen, Sen,
312 Liu, et al., 2017). However, recent findings indicate that long-term exposure to ATBC at
313 environmentally relevant concentration (0.5 µg/l) caused a significant adverse effect on the
314 reproductive system of adult zebrafish (Muhammad et al., 2018). There are evidences indicating that
315 ATBC might disrupt mouse antral follicle function (Rasmussen, Sen, Vera, et al., 2017) and be
316 detrimental to mouse ovarian function at low concentration (10 mg kg⁻¹ day⁻¹) (Rasmussen, Sen, Liu,
317 et al., 2017). All these evidences suggest that more information is needed for ATBC risk assessment.

327

328 Dodecane

329 Dodecane was present in all samples, with higher concentrations in MAPs/MEPs and CompostW.

330 There was no statistically significant difference between soil samples ($p = 0.643$), while CompostW
331 presented a significantly higher dodecane concentration ($p=0.001$) than CompostW/O.

332 There are no threshold limits set by the Regulation EU 2019/1009 (Regulation EU, 2019) for
333 dodecane, a major fuel component (Herbinet et al., 2007). Although it can impair the development of
334 frog embryos (Burýšková et al., 2006) at low doses (0.5 mg/l), and to induce papillomas in mice

335 (Baxter & Miller, 1987), this substance is not considered toxic as per the International Fragrance
336 Association (IFRA) Environmental Standards (Api et al., 2020). To our knowledge, there are only
337 few studies where dodecane occurrence was investigated and detected (but not quantified) in compost
338 tea, green waste compost and sludge mixed with palm waste (El Fels et al., 2016; Ezz El-Din &
339 Hendawy, 2010; Medicinal & Residues, 2014). The same applies to studies regarding dodecane
340 presence in soil and agricultural fields (Barrutia et al., 2011; Hempfling et al., 1991). Given the high
341 affinity of dodecane for plastic (PE in particular) (Castleman et al., 2021), we can speculate that
342 dodecane was adsorbed on the polymers' surface from the environment and that plastic is not a
343 primary source of this contaminant. Our analyses evidenced the presence of several other aliphatic
344 hydrocarbons, mainly alkanes, both in soil, compost and plastic samples. Further research is needed
345 to understand the source of such compounds and the risks associated with their presence in compost
346 products and agricultural fields.

347

348 Nonanal

349 Similarly to dodecane, nonanal was present in all samples, with higher concentrations in MAPs/MEPs
350 and CompostW. There was no statistically significant difference between the soil samples ($p \geq 0.181$)
351 except for Green pea, where nonanal concentration was statistically lower ($p \leq 0.02$) than in other soils;
352 it must be noticed that nonanal is not a plastic additive and, besides being used in perfumery and as a
353 flavoring agent, can also be directly emitted from vegetation and be present in some wax on the
354 surface of plants (Bowman et al., 2003).

355 Overall, the data indicate that plastics pollution might represent a source of contaminant for fertilizers
356 and agricultural soils.

357 As far as we know, the occurrence of nonanal in compost and agricultural soils is not regulated by
358 any European law, and there are no studies regarding its presence in agricultural soils. Published
359 research studies assessing the occurrence of nonanal in compost are scarce but the compound was

360 detected in garden waste compost (López et al., 2016) and in the emissions of municipal solid waste
 361 compost maturation treatment (Dorado et al., 2014).

362

363

	CHR	NAP	ANT	B[a]P	DEHP	ATBC	Dodecane	Nonanal
Green pea	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ (16)	112±3	224±12
BarleyS	<LOD	<LOD	<LOD	<LOD	<LOQ (27)	<LOQ (30)	110±2	274±22*
BarleyC	<LOD	<LOD	<LOD	<LOD	931±163	207±27	113±3	280±12*
BeanC	<LOD	<LOD	<LOD	<LOD	1080±209	102±21	111±5	256±22*
CompostW	<LOD	<LOD	111±34	<LOD	7090±3240*	<LOQ (28)	183±6*	431±64*
CompostW/O	<LOD	<LOD	108±18	<LOD	2610±1290	<LOD	155±8	340±14
MAPs/MEPs	<LOD	<LOD	651±84*	<LOD	38200±33900*	1100±105	815±19*	1380±31*

364 *Table 1 Mean organic contaminants concentrations and standard deviations (n=5) (ng/g dw). * denotes a significant higher*
 365 *concentration compared to the other sets of samples (p<0,05). Values in parentheses show the analytical result that was below the*
 366 *Limit of Quantitation, LOQ.*

367

368 3.2 Metals

369 Metals analyses were performed on compost soil samples and MEPs and MAPs.

370 The mean concentration for each metal is reported in Table 2. As, Se and Cd concentrations were
 371 below the limit of quantification (LOQ) in all samples, while MAPs/MEPs had values below LOQ
 372 for Be, V, Cr, Co, Ni and Pb. Statistical analyses did not show significant differences (p>0.05)
 373 between CompostW and CompostW/O.

374 In comparison to the compost samples, MAPs and MEPs showed a significantly (p<0.05) lower
 375 concentration of all metals analyzed except for Zn, which was found in a significantly higher amount
 376 in plastics (p=0.003). However, as stated before the compost with and without MAPs/MEPs did not
 377 differ significantly in terms of Zn content, indicating that the transfer of Zn from the plastics to the
 378 compost could be negligible. The metals concentrations in all the compost samples were similar to
 379 those found by Dimambro et al., (2007), and below the limit value levels established by the European
 380 Compost Network-European Quality Assurance Scheme for Compost and Digestate (European
 381 Compost Network, 2014). In comparison to plastics, soil samples showed significantly higher
 382 concentrations (p≤0.02) of all metals except for Zn, where the opposite result was found (p<0.001).

383 Cu, Fe and Zn concentrations were significantly lower ($p \leq 0.01$) in soil samples in comparison to
 384 compost samples, while all the other metals concentrations were significantly higher ($p \leq 0.04$) except
 385 for Mn in Barley S samples.

386 Considering these data, it seems that no transfer of metals occurred from the plastics to the compost.
 387 However, we cannot exclude that some of the metals had already transferred from the plastics to the
 388 compost during the composting process before our analyses were performed.

389

	CompostW	CompostW/O	MAPs/MEPs	Green pea	BarleyS	BarleyC	BeanC
Be	0.38±0.13	0.34±0.09	<0.22 (LOQ)	1.28±0.08	0.85±0.06	1.10±0.19	0.95±0.12
Al	6620±1260	7160±182	518±129*	30400±894	23000±1230	28000±1230	21800±2050
V	18±1.67	18±0.55	<1.7 (LOQ)	102±4.77	103±6.07	89±3.97	75±11
Cr	22±3.19	23±1.10	<8.1 (LOQ)	73±2.88	62±3.27	69±2.77	54±4.80
Fe	54600±6660	55600±3980	1820±466*	38400±1140	34400±1817	35600±1520	31800±5020
Mn	422±31	432±18	14±3.10*	764±80	396±17	750±65	650±130
Co	6.04±0.46	6.34±0.21	<2.2 (LOQ)	21±3.96	13±0.89	17±1.67	15±2.61
Ni	16±1	16±0.55	<6.6 (LOQ)	34±1.22	29±1.30	32±0.89	26±2.35
Cu	128±11	132±16	11±1.73*	29±1.22	32±1.22	36±1.52	37±4.32
Zn	492±90	582±234	1240±354*	152±4.47	<151 (LOQ)	182±8.37	136±11.4
As	<4.2 (LOQ)	<4.2 (LOQ)	<4.2 (LOQ)	<8.4 (LOQ)	<8.4 (LOQ)	<8.4 (LOQ)	<8.4 (LOQ)
Se	<25 (LOQ)	<25 (LOQ)	<25 (LOQ)	<50 (LOQ)	<50 (LOQ)	<50 (LOQ)	<50 (LOQ)
Cd	<1.3 (LOQ)	<1.3 (LOQ)	<1.3 (LOQ)	<2.5 (LOQ)	<2.5 (LOQ)	<2.5 (LOQ)	<2.5 (LOQ)
Pb	9.30±0.42	9.76±1.32	<2.8 (LOQ)	18±0.84	13±0.84	17±0.45	17±2.17

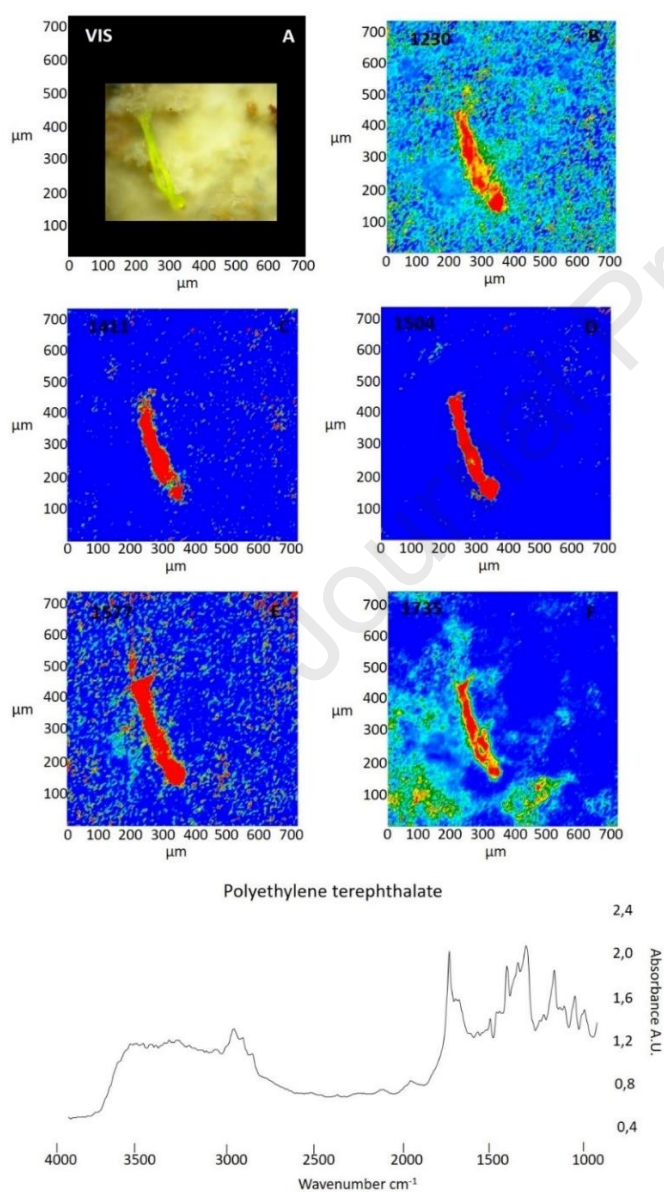
390 Table 2 Mean metals concentrations (mg/kg dw) and standard deviation (N=5). * in MAPs/MEPs data denotes significant difference
 391 compared to the compost ($p < 0.05$)

392

393 3.3 MPs, MEPs and MAPs FTIR analysis

394 36 MAPs/MEPs were collected from the compost and identified. Polypropylene (PP) (58.3%) and
 395 polyethylene (PE) (36.1%) were the most abundant polymers found in the samples, followed by
 396 acrylonitrile butadiene styrene (ABS) (2.8%) and polyethylene terephthalate (PET) (2.8%).

397 MAPs+MEPs concentration in the compost was 6.53 g/kg dw. These data are in agreement with those
 398 found by Watteau et al., (2018). The authors analyzed plastics >5 mm in two municipal solid waste
 399 compost samples, finding concentrations ranging between 1 and 15.3 g/kg dw (Watteau et al., 2018).
 400 MPs were detected in all the compost replicates. The relative abundance of each type of plastics
 401 analyzed was as follows: polyethylene terephthalate (PET) (44.2%), PE (25%), acrylates (9.6%), ABS
 402 (7.7%), PP (5.8%), polystyrene (PS) (3.9%), acrylonitrile (1.9%), polyurethane (PU) (1.9%). MPs
 403 mean concentration found in the compost was 6.6 ± 1.5 items/g dw.



404

405 Fig 1. 2D FITR imaging of a plastic fiber found in the compost. (A) Visible light image of the fiber. (B-F) 2D FTIR Imaging maps
 406 (700 × 700 μm²), showing the intensity of the following bands: (B) 1230 cm⁻¹ (CO stretching); (C) 1411 cm⁻¹ (aromatic skeleton

407 stretching); (D) 1504 cm^{-1} (aromatic C=C stretching); (E) 1577 cm^{-1} (aromatic C=C stretching); (F) 1735 cm^{-1} (CO stretching). The
408 absorbance intensity of the bands is shown in false colors: blue < green, < yellow < red. The bottom panel shows the FTIR Reflectance
409 spectrum of the plastic fiber, relating to a single pixel ($5.5 \times 5.5 \mu\text{m}^2$) of the 2D Imaging map.

410 Fig. 1 shows an example of a yellow fiber (ca. 300 μm long) that was analyzed through FTIR
411 microscopy using the FPA detector, and identified as polyethylene terephthalate due to intense
412 absorption peaks at 3000-2800 (aromatic and aliphatic CH stretching region), 1735 (C=O stretching),
413 1230 cm^{-1} (C-O stretching), 1577 and 1504 (aromatic C=C stretching), 1411 (aromatic skeleton
414 stretching) (Z. Chen et al., 2012; Jung et al., 2018; Pereira et al., 2009).

415 In the supplementary materials, Figures S1-S7 show the different polymers found in the samples.

416 The lack of information on the abundance of MPs in compost is a gap in the scientific literature that
417 needs to be filled (Scopetani et al., 2020). Only few studies have investigated MPs pollution in
418 recycled fertilizers so far. Among them, Gui et al., (2021) studied MPs (0,05-5mm) in compost from
419 rural domestic waste finding an average concentration of 2.4 ± 0.4 items/g dw with polyester, PP and
420 PE being the most common polymers (Gui et al., 2021). These findings comply with our results.

421 El Hayany et al., (2020) quantified MPs in fresh and in dewatered sewage sludge with mean
422 concentrations of 40.5 ± 11.9 particles/g and $36.0 \pm 9,7$ particles/g, respectively (EL Hayany et al.,
423 2020), about one order of magnitude higher than our results. Instead, lower MPs concentrations were
424 detected in compost samples by Schwinghammer et al., (2020) and Braun et al., (2021) ranging from
425 39 to 102 items/kg, and from 12 ± 8 to 46 ± 8 items/kg, respectively (Schwinghammer et al., 2020;
426 Braun et al., 2021). Braun et al., (2021) estimated that compost application to agricultural fields
427 includes a plastic load of 84,000 to 1,610,000 plastic items ha per year (Braun et al., 2021). This
428 calculation was made taking into account the common recommendations in composting practice that
429 establish an application rate ranging from 7 to 35 t compost ha^{-1} per year.

430 Applying the same estimate to the MPs and MAPs/MEPs concentrations we found in compost, we
431 obtained a MPs load of 4.62×10^7 to 2.31×10^8 items ha^{-1} per year and a MAPs/MEPs load of $4.57 \times$
432 10^4 to 2.29×10^5 g ha^{-1} per year. This indicates that the input of plastics coming from the application
433 of compost to agricultural soil might be higher than previously estimated (Braun et al., 2021).

434 Thus, our data shows that compost can be a source of plastic contamination to agricultural fields and
435 that technical strategies aimed to minimize the presence of polymers in recycled fertilizers are needed.

436

437

438 **Conclusions**

439 The purpose of this research was to study the occurrence of MAPs, MEPs and MPs in compost and
440 soil samples, as well as evaluate the transfer of selected contaminants from the plastics contained in
441 the compost to the compost itself, and later on to the soil.

442 MAPs/MEPs and MPs concentrations in compost were 6.5 g/kg and 6.6 ± 1.5 items/kg respectively,
443 based on which we estimated a MAPs/MEPs load of 4.57×10^4 to 2.29×10^5 g ha⁻¹ per year, and a
444 MPs load of 4.62×10^7 to 2.31×10^8 items ha⁻¹ per year into agricultural soils. We can thus consider
445 compost as a source of plastic contamination to agricultural fields. MAPs/MEPs had the highest
446 concentrations of all contaminants (except for metals) in comparison to soil and compost samples,
447 indicating that they are a source of potential pollutants transfer from the plastics to the compost and
448 soil. Indeed, MAPs/MEPs-containing compost had significantly higher concentrations of DEHP,
449 ATBC, dodecane and nonanal than compost where these plastics had been removed before
450 contaminants' extraction and analysis. Besides, higher concentrations of DEHP, ATBC and nonanal
451 were also found in the soil samples that had been fertilized with compost, supporting the hypothesis
452 of a contaminant transfer chain from plastics to compost, and then to soil.

453 A significant transfer of metals from the plastics seems unlikely since plastics had lower metals
454 concentrations than compost and soil samples. However, we cannot exclude that some of the metals
455 had already transferred from the plastics to the compost during the composting process.

456

457 Our data indicate that there are risks associated with the presence of plastics in recycled fertilizers
458 and, therefore, regulatory guidelines are needed to ensure the good quality of the final agricultural

459 products. Furthermore, different crops might have diverse capability of fixating and accumulating
460 contaminants. This aspect should be further investigated in future studies to better understand the
461 risks of the presence of plastics in agricultural fields to humans.

462

463

464 **Acknowledgment**

465 We thank Santeri Savolainen for assisting with the metal analysis. CSGI (Consorzio Interuniversitario
466 per lo Sviluppo dei Sistemi a Grande Interfase) is also gratefully acknowledged for partial funding.

467

468 **Funding**

469 This work was funded by Maa-ja vesitekniikan tuki ry (Land and Water Technology Foundation) and
470 by Onni ja Hilja Tuovisen Säätiö within the CoTraP project. Additional funding was provided by
471 Helsinki-Uusimaa Regional Council/ European Regional Development Fund within the KIEMURA
472 – PROJECT: Circular Economy Solutions for Microplastics and Recycled Plastics. Partners: Lahti
473 University of Applied Science, University of Helsinki, Muovipoli Oy. Partial funding was also
474 provided by CSGI (Consorzio Interuniversitario per lo Sviluppo dei Sistemi a Grande Interfase).

475

476 **Credit authorship contribution statement**

477 Costanza Scopetani – Experimental design, Sampling, Samples treatment, Measurements, MPs
478 characterization, Data analysis and interpretation, Figures, Writing of the article

479 David Chelazzi – MPs characterization, Data analysis and interpretation, Revising Text and Figures

480 Tania Martellini – Revising Text and figures

481 Alessandra Cincinelli– Revising Text and Figures

482 Ville Leiniö – Macroplastics characterization

483 Jukka Pellinen – Experimental design, Data discussion and Revising Text and Figures

484

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845

Highlights

- MAPs/MEPs and MPs concentration in the compost: 6.5 g/kg dw and 6.6 ± 1.5 items/g dw
- MPs load estimation: $4,6 \times 10^7$ to $2,3 \times 10^8$ items ha⁻¹ yr⁻¹ into agricultural soils
- MAPs/MEPs-containing compost had significantly higher concentration of DEHP
- Compost can represent a source of plastic contamination to the agricultural fields

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.

Journal Pre-proof