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Airborne emissions of microplastic fibres from domestic laundry

dryers

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- 43 Graphical Abstract:



52 Mandatory Highlights:

53

- 54 1. Emissions of microplastic fibres from residential dryers into air investigated.
- 55 2. Mechanical drying of polyester emits microplastic fibres into the surrounding air.
- 56 3. Over 20 mins operation, MP emissions for a 660 g blanket were $\sim 1.6 1.8$ fibres/ m³.
- 57 4. Lint of 77 ± 22.4 mg \approx 1.1 x 10⁶ ± 3.2 x 10⁵ fibres captured by inbuilt filtration.
- 58 5. Lint emissions were approximately 0.012% of the blanket mass/ wash.

59

61 Abstract:

62 An emission source of microplastics into the environment is laundering synthetic textiles and 63 clothing. Mechanical drying as a pathway for emitting microplastics, however, is poorly understood. 64 In this study, emissions of microplastic fibres were sampled from a domestic vented dryer to assess 65 whether mechanical drying of synthetic textiles releases microplastic fibres into the surrounding air 66 or are captured by the inbuilt filtration system. A blue polyester fleece blanket was repeatedly 67 washed and dried using the 'Normal Dry' program of a common domestic dryer operated at 68 temperatures between 56 and 59 °C for 20 minutes. Microfibres in the ambient air and during 69 operation of the dryer were sampled and analysed using microscopy for particle quantification and 70 characterisation followed by Fourier-Transform Infrared Spectroscopy (FTIR) and Pyrolysis Gas 71 Chromatography-Mass Spectrometry (Pyr-GC/MS) for chemical characterisation. Blue fibres 72 averaged 6.4 \pm 9.2 fibres in the room blank (0.17 \pm 0.27 fibres/ m³), 8.8 \pm 8.5 fibres (0.05 \pm 0.05 73 fibres/m³) in the procedural blank and $58 \pm 60 (1.6 \pm 1.8 \text{ fibres/m}^3)$ in the sample. This is the first 74 study to measure airborne emissions of microplastic fibres from mechanical drying, confirming that 75 it is an emission source of microplastic fibres into air – particularly indoor air. 76 77 Key words: 78 Microplastic, emission, air, fibre, dryer, Pyr-GC/MS 79

- 80
- 81

82 1. Introduction

83 Microplastics are small pieces of synthetic polymers with their size commonly defined as being 84 between 1 μ m and 5 mm (Dris, 2016; Henry et al., 2019). While typically defined by the length of 85 their longest dimension, they have also been defined according to their diameter (Napper and 86 Thompson, 2016) or ratio of dimensions (Obbard et al., 2014) for characterisation. Microplastic 87 fibres have been reported as prevalent in all environmental compartments including atmospheric air 88 and deposition (Cai et al., 2017; Dris et al., 2015; Dris, 2016), on beaches (Claessens et al.), surface sea water (Cózar et al., 2014), the water column (Dai et al., 2018), marine sediments (Cole et al., 89 90 2011; Van Cauwenberghe et al., 2013), sea ice (Obbard et al., 2014), freshwater lakes (Eerkes-91 Medrano et al.; Eriksen et al., 2013; Gasperi et al., 2014), sediments (Horton et al., 2016; Klein et al., 92 2015) and soils (Hurley and Nizzetto, 2018; Nizzetto et al., 2016). Microplastic fibres have been 93 detected in both urban (Dehghani et al., 2017; Dris et al., 2015; Dris et al., 2016) and remote regions 94 (Allen et al., 2019; Free et al., 2014), including the Arctic (Lusher et al., 2015) and Antarctic (Cincinelli 95 et al., 2017) and at high altitudes (Ambrosini et al., 2019) suggesting the capacity for long range 96 atmospheric transport (Allen et al., 2019). Fibres are the most commonly identified microplastic 97 shape reported in the gastrointestinal tracts of biota at all trophic levels; present within food 98 (Barboza et al., 2018) and deposited onto food destined for human consumption (Catarino et al., 99 2018). Microplastic fibres have also been identified within the human lung (Pauly et al., 1998) and 100 examined in simulated respiration models operated within indoor environments (Vianello et al., 101 2019), therefore, potentially posing a risk to human health through inhalation and ingestion 102 exposure (Gasperi et al., 2018; Prata, 2018; Wright and Kelly, 2017).

103

Despite the environmental prevalence and research importance of microplastic fibres (Henry et al., 2019), little is known about their emission sources. Point source laundry emissions of wet lint from simulated industrial (Cocca et al., 2018; De Falco et al., 2018) and residential laundry effluent have been examined using laboratory based (Carney Almroth et al., 2018; Cocca et al., 2018; De Falco et

108 al., 2018; Hernandez et al., 2017; Jönsson et al., 2018) and commercially available (Browne et al., 109 2011; Folko, 2015; Hartline et al., 2016; Karlsson, 2014; McIlwraith et al., 2019; N. Bruce, 2016; Pirc 110 et al., 2016; Sillanpää and Sainio, 2017) laundering equipment. Early studies found that >1900 fibres 111 are emitted per wash of a single blanket/ fleece or shirt garment, equating to >100 fibres/ L of 112 captured laundry effluent (Browne et al., 2011). Subsequent research has calculated that each 113 blanket releases approximately 1.1 x 10⁵ fibres per wash (Carney Almroth et al., 2018). Total laundry 114 emissions could release anywhere from 7 x 10⁵ fibres per 6 kg load (Napper and Thompson, 2016) 115 $(1.16 \times 10^{5} \text{ fibres/ kg})$ to between 6 – 17.7 x 10⁶ fibres $(1.2 - 3.5 \times 10^{6} \text{ fibres/ kg})$ based on 0.43 to 116 1.27 g lint weight from a 5 kg load (De Falco et al., 2018). Variability could be attributed to mesh 117 sizes used in different studies, material composition, weave and fibre structure (De Falco et al., 118 2018), or the use or absence of chemical confirmation methods.

119

120 Despite previous studies identifying that laundering clothing is a significant point source for 121 emissions of microplastics (Carney Almroth et al., 2018; De Falco et al., 2018; Hernandez et al., 2017; 122 McIlwraith et al., 2019; Pirc et al., 2016; Salvador Cesa et al., 2017; Sillanpää and Sainio, 2017; 123 Zambrano et al., 2019), as far as we are aware, only two studies have examined the process of 124 mechanical drying of clothing and textiles. Limited to microfibre emissions captured within the 125 internal filtration mechanisms (Pirc et al., 2016), one study established that mechanical drying 126 resulted in greater microplastic emissions captured in emitted microfibres than fibres released into 127 laundry effluent (Pirc et al., 2016). The second study examined laundering of clothing with wash and 128 dry cycles, however dryer emissions were unreported (Zambrano et al., 2019). The hypothesis was 129 that residential dryers contribute microplastic fibres into the surrounding atmospheric environment. 130 The aim of the current study was to determine whether microplastic fibres are captured in inbuilt 131 filtration or are emitted into the surrounding indoor/ outdoor atmospheric environment during the 132 mechanical drying of synthetic textiles.

133

134 2. Materials and Methods

135

136 **2.1 Sampling details**

A blue coloured fleece blanket, labelled by the manufacturer as 100% polyester (fleece) and suitable
for mechanical drying, was purchased from a prominent Australian retail outlet. The blanket,
measuring 152 cm x 203 cm in size, was repeatedly laundered alone consecutively for five individual
wash and dry cycles. Gravimetric analysis was conducted both prior to and post laundering cycles
using a laboratory balance (Metter Toledo, New Classic MS Balance). The average (± standard
deviation) of the dry blanket mass prior to and during consecutive laundry processing was 665 ± 6.73

143 g.

144

145 Laundering was performed within an 8 kg sensor washing machine (Bosch Australia, Clayton, VIC) 146 using a standard 45-minute-cycle at 40 °C and 1400 revolutions per minute (RPM). Samples were 147 mechanically dried for 20 minutes using a 6.5 kg sensor dryer (Electrolux, Alexandria, NSW) 148 representative of the Australian market. The dryer was operated using the 'Normal Dry' program 149 described by the manufacturer as "suitable for everyday fabrics". Technical details of the drying 150 program are unable to be provided by the manufacturer as the internal program varies the RPM and 151 temperature automatically based on a combination of exhaust and ambient air temperature. 152 However, internal maximum temperatures were monitored by using a temperature button 153 (iTemperature, Instrument Choice) during operation both with and without a blanket, with 154 temperatures ranging between 56 and 59 °C, achieved at approximately 10 min of dryer operation. 155 The dryer was installed within a room approximately 21 m³ in size, with no active ventilation. Access 156 to the room during the sampling program was prevented to minimise air flow and potential 157 contamination. The only air exchange occurred when opening and closing the door immediately 158 prior to and post sampling.

159

160 <u>2.1.1 Airborne particulate matter and inbuilt dryer filter sampling of polyester microfibres</u>

161 Airborne particulate matter was sampled using a high volume total suspended particle air sampler 162 (Komoto, Japan) with a sampling volume of 55 m^3 / hour to collect all airborne particles 163 indiscriminate of size. Samples were collected onto a Whatman GF/A glass filter (1.6 µm), muffle 164 furnaced at 500 °C for 4 hours prior to use. Based on the extraction efficiency of the air sampler and 165 room volume, sampling was limited to the first 20 minutes of dryer operation to avoid over sampling 166 the air. Air samples were collected prior to the experiment as an ambient air room blank, during operation of the empty dryer as a procedural blank and whilst mechanically drying a blanket as five 167 168 replicate samples (n=5). Microfibres were collected by manually wiping the inbuilt dryer filter after 169 procedural blanks and samples with paper towel, then stored wrapped in aluminium foil. At least ten 170 minutes between each sample collection of air and lint was provided to allow time for deposition of 171 resuspended particles (Cheng et al., 2016).

172

173 **2.2 Microscopy**

174 Analysis of the blanket, total particle count (fibres and fragments) and characterisation (colour, 175 morphology and size) was undertaken using an Olympus SZ-CTV microscope coupled with a Motic 176 Images Plus (Software Version 3.0) camera. To facilitate representative sub sampling, the samples 177 (filters) from two room blanks, one procedural blank and one sample were divided into quarters and 178 microscopically analysed separately to test homogeneity within the sample. Applying an ANOVA test 179 to the null hypothesis that there was no variance between quarters for blue fibre count resulted in 180 an f statistic of 0.173 respectively, with a significance of p = 0.913. A Brown-Forsyth test assessing 181 homogeneity within the sample indicated a significance of p = 0.912 for blue fibres inferring 182 moderate homogeneity between the quarters – both confirming the null hypothesis. Quartile 183 analysis of the entire filter was acceptable, consistent with the recommendation of Koelmans et al

184 (Koelmans et al., 2019). Quarter 3 (respective to sampler orientation) was selected for sub sampling

as it demonstrated the least variability between sample types and replicates.

186

187 2.3 Chemical composition

188

189 <u>2.3.1 Fourier-transform infrared spectroscopy (FTIR)</u>

190 The blanket, both prior to and post laundering, and inbuilt dryer filter contents were characterised

191 using Fourier-Transform Infrared Spectroscopy (FTIR) (Spectrum 2, Perkin Elmer) against a plastic

192 specific spectral library (ATR Polymer Introductory Library, Perkin Elmer). After obtaining background

193 spectra, samples were analysed in absorbance mode from 4000 cm⁻¹ to 400 cm⁻¹ at 300 scans/

194 minute, data interval of 0.2 cm⁻¹ and resolution of 16 cm⁻¹. A random sub sample of larger visible

195 blue fibres were manually extracted from the filter papers to examine for fibre composition analysis

196 purposes to match to the blanket source. Identification was based on the library spectra with the

197 highest percentage match to the sample. Only matches above 80% were used for positive

198 identification.

199

200 <u>2.3.2 Pyrolysis- gas chromatography -mass spectrometry (Pyr-GC/MS)</u>

201 Pyrolysis gas chromatography mass spectrometry (Pyr-GC/MS) was used for chemical

202 characterisation to verify the blanket material and confirmation/ assist in determining the

203 composition of collected fibres.

204

Pyrolysis was undertaken using a multi-shot micro-furnace pyrolyser (EGA/ PY-3030D) equipped with an auto sampler (AS-1020E) (Frontier Lab Ltd., Fukushima, Japan) coupled to a GC/MS – QP2010-Plus (Shimadzu Corporation, Japan). Being for identification and confirmation purposes only against the reference material of the blanket and PET analytical standard, the pyrolyser was operated in single shot mode. Based on the optimisation parameters in Okoffo et al (Okoffo et al., 2020), the furnace

210	was programmed to 650 °C with pyrolysis occurring for 12 seconds. The pyrolyser interface and GC
211	injection port temperature was set at 300 °C with the samples injected at a split of 1:5 onto an Ultra
212	Alloy $^{ extsf{B}}$ 5 capillary column (30 m, 0.25 mm I.D., 0.25 μ m film thickness) (Frontier Lab). The GC oven
213	temperature was held at 40 °C for 2 minutes, increased to 320 °C at 20 °C min $^{-1}$, then held for 14
214	minutes. Helium was used as a carrier gas at 1.0 mL/ min with a constant linear velocity. The mass
215	spectrometer was operated in full scan mode, acquiring data from 40 to 600 m/z, and the pyrogram
216	was recorded from 2 to 30 minutes.
217	
218	2.4 Statistical Analysis
219	All quantification figures are based on a confidence interval of 95%, being ± two times the standard
220	deviation of the mean calculated in Microsoft Excel version 16. Microscopy quantification of one
221	quarter of the sample were extrapolated to the whole filter (number of fibres detected/ m^3 based on
222	Quarter 3 of the sample multiplied by 4) for whole of sample quantifications.
223	
224	2.5 Contamination controls, QA/ QC
225	
226	2.5.1 Room Control QA/ QC
227	Efforts to reduce contamination were undertaken during all procedures. A restricted access room
228	was utilised, cleaned thoroughly with Ethanol 70%, wiping all surfaces with paper towel and
229	vacuuming the rubber floor twice two days prior to commencement. Access was prohibited during
230	sampling episodes. However, contamination was possible through people entering the restricted
231	room on days between sampling episodes; one small open but not operational air vent and an
232	exposed section of 2.6 m x 0.15 m between the upper levels of the building which could have
233	contributed particles to the results.
234	
235	2.5.2 Sampling and Storage QA/QC

236 Sampling and laboratory contamination controls included placing the sample inside of a white cotton 237 pillowcase when not in use and stored within a metal container. The samples were extracted from a 238 new mechanical dryer, wiped clean with paper towel and Milli-Q water prior to each use (Napper 239 and Thompson, 2016). The dryer was operated empty between the room blank and sample. The 240 internal drum was also wiped clean with paper towel between replicates and the inbuilt dryer filter 241 was wiped clean after each procedural blank and sample to remove potential carry over. Workspace 242 controls included wiping all surrounding laboratory surfaces with Ethanol 70% and paper towel prior to use and operating without gloves where possible. Particle free gloves were used where necessary. 243 244 Green cotton laboratory coats were worn and the clothing of attendees was noted during each 245 sampling process. Subtraction was undertaken for any coloured fibre which was not blue, matching 246 the blanket. The filter papers were stored within aluminium foil and during microscopy blank filter 247 papers were exposed to determine whether baseline subtraction of laboratory contamination was 248 required (Dris, 2016). Contamination mitigation for pyrolysis analysis included using new sample 249 cups for each sample, collecting fibres using Ethanol 70% and MilliQ cleaned forceps, wiped with 250 paper towel.

251

252 2.5.3 Analysis QA/ QC

Blank Whatman GF/D (2.7 μm) filters were exposed at all times during microscopy. Although the
samples themselves were covered while not directly being examined, establishing baseline
deposition contamination during microscopy analysis was prudent.

256

Particles were collected immediately prior to pyrolysis analysis and deposited into the pyrolysis
sample cups under microscopy, confirming particle discharge from the forceps into the cup and
correct placement for analysis. Samples were covered with aluminium foil during the physical
transfer to the automatic sampler and while loading the samples. The pyrolysis unit itself also
features a plastic guard which protects samples from atmospheric deposition during the duration of

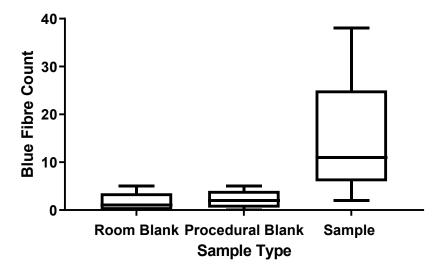
- processing time.
 3. Results
 3. Results
 3.1 QA/ QC Results
 Contamination during microscopy varied between 0 8 particles, averaging across all room blanks,
 procedural blanks and samples to be 2.6 ± 2.5 (95% Cl) fibres of various colours. No blue fibres were
 detected in the room blank, 0.4 ± 1.1 blue fibres in the procedural blank and 1.7 ± 2.2 blue fibres in
- the sample. This is considered to be negligible compared to the average number of blue fibres (8.8 ±
- 8.54 fibres) in the procedural blank and in the sample (58 ± 60 fibres), and therefore no baseline
- 272 subtraction of contamination during analysis was undertaken. Contamination during microscopy
- 273 consisted of particles which were 87% fibre shaped and 14% fragments, with the most dominant
- colour being black. Over 70% of the contamination particles were <50 μm in size. The size fractions
- and colour characteristics of the contamination is detailed in Figures S1 S2 of the SI.
- 276

277 3.2 Air concentration

278 The number of blue particles on the analysed quarter of each sample was 1.6 ± 2.5 fibres in the

room blank, 2.2 ± 2.3 fibres in the procedural blank and 14.6 ± 16.96 in the sample. (Figure 1)

Blue fibre emissions



280

Figure 1: Counts of blue particles per sample type.

282 *Box indicates 25th and 75th percentiles, line is the median and whiskers represent minimum and

283 maximum values. *Procedural Blank is the operation of the empty dryer without the blanket

sample.

285

286 Extrapolated to whole filter calculations, the room blank contained 6.4 ± 10.3 blue fibres, 8.8 ± 9.6

287 blue fibres in the procedural blank and 58.4 ± 67.9 blue fibres in the sample.

288

289 An ANOVA grouping the room blank and procedural blank against the sample demonstrated an f

statistic of 8.9 and a p value = 0.01, indicating with statistical significance that the blanket emits

291 microplastic fibres into the airborne environment.

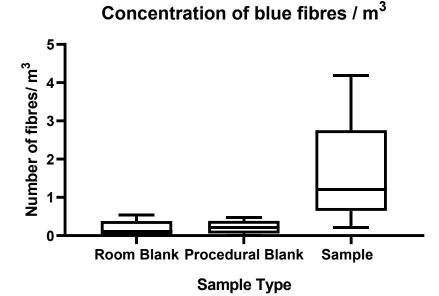
292

293 <u>3.2.1 Particle concentration in the air</u>

294 Normalised against the operation of the sampler, the number of blue fibres in the air was calculated

to be 0.17 \pm 0.27 fibres/ m³ in the room blank, 0.5 \pm 0.5 fibres/ m³ in the procedural blank and 1.6 \pm

296 1.8 fibres/ m³ in the sample. (Figure 2)



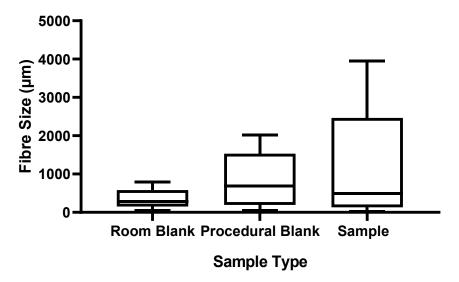




299 *Procedural Blank references the operation of the empty dryer without the blanket sample.

- 300
- 301 3.3 Air Particle Characterisation
- 302 <u>3.3.1 Size Classification</u>
- All blue particles were fibre shaped. The length of the blue fibres varied from $43 799 \,\mu\text{m}$ in the
- room blank (average $341 \pm 273 \mu$ m), $47 2016 \mu$ m (average $844 \pm 819 \mu$ m) in the procedural blank
- and $19 3948 \,\mu\text{m}$ (average 764 ± 940 μm) in the sample. (Figure 3)

Size distribution of blue fibres



306

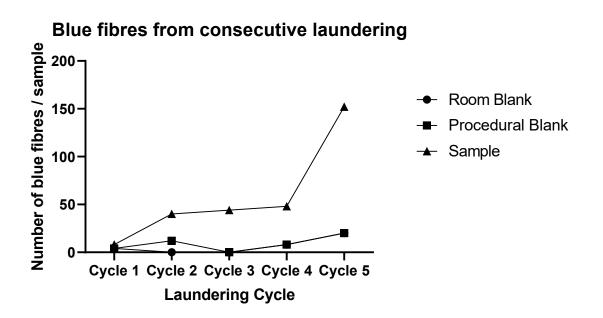
307 Figure 3: Blue fibre size comparison between sample types.

308

309 <u>3.4 Particle counts and consecutive laundering</u>

310 The number of blue fibres in the atmosphere increased over consecutive laundering. No stability of 311 fibres release was reached over time from five replicates. This is in contrast to Pirc et al (Pirc et. al; 312 2016), who demonstrated that shedding of a PET blanket during laundering and mechanical drying (captured lint in inbuilt filtration) decreased over consecutive laundering, reducing substantially post 313 wash 3 to account for new garment shedding and stabilising after wash 7. Carney Almroth et al 314 315 determined that aged garments shed more fibres than new garments. However, overall shedding of 316 fibres from PET fabric during laundering decreased over time, reaching stability for one type of PET weave after wash 5 whilst no stability was reached for a second PET weave after 10 washes (Carney 317 318 Almroth et al., 2018). Hernandez et al found that laundered PET fabric released decreasing amounts 319 of microplastic fibres, stabilising after wash cycle 3 to 4 depending on laundry surfactant (Hernandez 320 et al., 2019). It is unknown what attributed to the increased shedding of fibres into the atmospheric 321 environment in these results. It is hypothesised that the increase of fibres into the air could be a 322 result of degradation of the fibre from mechanical drying, possibly from the physical abrasion of the 323 fabric within the internal drum of the dryer during rotation or the weakening of the fibres from the

- heat generated internally whilst mechanically drying the blanket. The cause remains unknown as
 does the impact of repeated laundering past 5 replicates, which should be examined further in
 future research. (Figure 4).
- 327



328

- 329 Figure 4: Blue fibre emissions from consecutive laundering.
- 330 *Extrapolated whole of filter calculations.
- 331 Chemical and Visual Characterisation of Material and Fibre Composition
- 332
- 333 <u>3.5 FTIR analysis</u>
- FTIR characterised the blanket as poly (1,- cyclohexanedimethylene terephthalate) with 80 % and 78
- 335 % accuracy (Figure S3) prior to processing, and poly (1,- cyclohexanedimethylene terephthalate) with
- 80 % accuracy (Figure S4) for the inbuilt dryer filter contents. Examination of randomly selected
- visible individual blue fibres proved unsuccessful based on difficulties ensuring correct placement of
- the particle for examination and library comparisons of very low accuracy.
- 339

340 <u>3.6 Pyr-GC/MS analysis</u>

341 Fibres from the blanket that were unlaundered; washed and oven incubated at 60°C for 20 minutes,

342 along with pools of three to six individually picked blue coloured fibres from the samples to 343 determine if they originated from the blanket fibres. Pyrograms from the blankets demonstrated 344 consistent peaks in the chromatograms between replicates in total ion chromatograms (TIC) and 345 indicator compounds/ ions regardless of laundering effects. The chemical composition of the blanket 346 was identified to be polyethylene terephthalate (PET) based on the presence of benzene, vinyl 347 benzoate and benzoic acid. Insufficient indicator compounds were present for the positive 348 identification of Poly (1,-cyclohexanedimethylene terephthalate), identifying discrepancy between 349 FTIR and Pyr-GC/MS. Pyrograms following oven exposure for 20 minutes at 60 °C were consistent 350 with PET. Pyrograms of pools of blue fibres indicated a PET composition in two replicates. All 351 pyrograms are presented in the Supporting Information (Figure SI 5 - SI 7). 352 353 3.7 Microfibre emissions from mechanical drying captured by inbuilt filtration 354 Drying one 660 g polyester blanket generated 77 \pm 22 mg of lint which equates to ~1.1 \pm 0.3 x 10⁶ 355 fibres into the inbuilt filter. This is approximately 0.012% of the blanket mass. Additionally, 54 ± 60 356 fibres would be released into the air. It is noted these calculations have many associated 357 uncertainties and assumptions, including that the per fibre weight is the same as that used in the calculations by de Falco et al (De Falco et al., 2018) of 7 x 10^{-8} g/ fibre: 6 x 10^{6} to 17.7 x 10^{6} fibres 358 359 corresponding to a lint weight of 0.43 - 1.27g of lint = $1.27g/17.7 \times 10^6 = 7 \times 10^{-8} g/$ fibre. Inherent 360 variability in dryer emissions also exists regarding dryer characteristics such as different load 361 capacities/ venting options and different shedding propensities of the load composition and fabrics 362 based on manufacture differences and fibre characteristics. As such, future estimates of microplastic 363 emissions from laundering of synthetic materials should include drying as an emission source, 364 however emission amounts require refinement and further research.

365

366 4. Discussion

367 Airborne emissions of 58 ± 60 fibres per 660 g blanket sample were higher than the amount of blue

368 fibres present in the ambient air, being 6.4 ± 9.21 fibres. Subtracting the room blank and procedural 369 blank values, mechanical drying contributes ~2 fibres/ m³ into the surrounding atmospheric 370 environment. Working from the average of 58 fibres/ 660 g blanket, drying 1 x 660g blanket/ once 371 per week (consistent with Australian dryer operation (Statistics, 2008a; Statistics, 2011)) could emit 372 3×10^3 airborne fibres into households and/ or the atmospheric environment per household, per 373 annum. A whole 6.5 kg polyester blanket load (being 6.5 kg wet weight capacity/ average blanket 374 wet weight of 830 g = 7.8 blanket capacity per drying load) could emit 406 ± 468 airborne fibres/ load operated with exclusively polyester blankets. These figures do not consider discretionary dryer 375 376 usage, differences in fabric, variability in usage both nationally and internationally or mechanical 377 variations between dryer types.

378

379 As many of these fibres escape inbuilt filtration or are released when cleaning the filter (Cheng et al., 380 2016), human health implications should also be considered and examined. When vented indoors, 381 these fibres are likely to accumulate in dust and contribute to microfibre abundance being reported 382 in dust, as well as exposure via dust. Dryer placement within the household, clothing composition 383 and frequency of use are expected to vary between countries. In Australia, 56% of households own a 384 mechanical dryer and operate their dryer once per week (Statistics, 2008a; Statistics, 2008b), 385 however frequency of operation would vary nationally, with differing climates and dryer ownership 386 and operation frequency varies internationally. For example, in 2018, 58% of UK households owned 387 a dryer and in 2017, 42% of households in Germany. In some countries such as the UK, the dryer is 388 located within or adjoining the kitchen (Wendy Wills, 2013) which could potentially increase human exposure to airborne particulates and/ or result in deposition onto food (Catarino et al., 2018) and 389 390 food preparation surfaces, providing another avenue of human ingestion exposure as well as 391 respiratory exposure.

392

393 **<u>5. Conclusion, limitations and future work</u>**

The major limitations of this work are the small sample size, the examination of one type of polyester blanket and one type of domestic dryer. Limitations also include the inability to calculate mass loss of the blanket and mass of the airborne fibres/ filters, making gravimetric mass balance unachievable. Calculations of fibres per m³ are approximate, based on variability of the air volumes sampled. Drying time was limited during treatments to the first 20 minutes of the cycle in consideration of room capacity to avoid over sampling the air. Retained water could have impacted the mass based outcomes.

401

Future work is required to gain an understanding of airborne dryer emissions, including the influence of technical specifications such as dryer composition (condenser/vented), temperature, RPM variability, textile materials or their length and weave composition impact on the prevalence of emissions into the environment (De Falco et al., 2018), as well as considering general laundry load composition or commercial laundromat emissions. These specifications could be applied into lifestyle or policy adaptations to reduce environmental and human exposure to microplastic fibres.

408

409

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420	Microscope.
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- 426 References:
- Allen S, Allen D, Phoenix VR, Le Roux G, Durántez Jiménez P, Simonneau A, et al. Atmospheric
 transport and deposition of microplastics in a remote mountain catchment. Nature
 Geoscience 2019; 12: 339-344.
- Ambrosini R, Azzoni RS, Pittino F, Diolaiuti G, Franzetti A, Parolini M. First evidence of microplastic
 contamination in the supraglacial debris of an alpine glacier. Environmental Pollution 2019;
 253: 297-301.
- Barboza LGA, Dick Vethaak A, Lavorante BRBO, Lundebye A-K, Guilhermino L. Marine microplastic
 debris: An emerging issue for food security, food safety and human health. Marine Pollution
 Bulletin 2018; 133: 336-348.
- Browne MA, Crump P, J Niven S, Teuten E, Tonkin A, Galloway T, et al. Accumulation of Microplastic
 on Shorelines Woldwide: Sources and Sinks. Vol 45, 2011.
- Cai L, Wang J, Peng J, Tan Z, Zhan Z, Tan X, et al. Characteristic of microplastics in the atmospheric
 fallout from Dongguan city, China: preliminary research and first evidence. Environmental
 Science and Pollution Research 2017; 24: 24928-24935.
- 441 Carney Almroth B, Åström L, Roslund S, Petersson H, Johansson M, Persson N-K. Quantifying
 442 shedding of synthetic fibers from textiles: a source of microplastics released into the
 443 environment. Environmental Science and Pollution Research 2018; 25: 1191-1199.
- Catarino AI, Macchia V, Sanderson WG, Thompson RC, Henry TB. Low levels of microplastics (MP) in
 wild mussels indicate that MP ingestion by humans is minimal compared to exposure via
 household fibres fallout during a meal. Environmental Pollution 2018; 237: 675-684.
- Cheng K-C, Zheng D, Tetteh AO, Park H-K, Nadeau KC, Hildemann LM. Personal exposure to airborne
 particulate matter due to residential dryer lint cleaning. Building and Environment 2016; 98:
 145-149.
- Cincinelli A, Scopetani C, Chelazzi D, Lombardini E, Martellini T, Katsoyiannis A, et al. Microplastic in
 the surface waters of the Ross Sea (Antarctica): Occurrence, distribution and
 characterization by FTIR. Chemosphere 2017; 175: 391-400.
- 453 Claessens M, Van Cauwenberghe L Fau Vandegehuchte MB, Vandegehuchte Mb Fau Janssen CR,
 454 Janssen CR. New techniques for the detection of microplastics in sediments and field
 455 collected organisms.
- 456 Cocca M, Di Pace E, Errico ME, Gentile G, Montarsolo A, Mossotti R. Proceedings of the International
 457 Conference on Microplastic Pollution in the Mediterranean Sea. Cham: Springer
 458 International Publishing, 2018.
- 459 Cole M, Lindeque P, Halsband C, Galloway TS. Microplastics as contaminants in the marine
 460 environment: A review. Marine Pollution Bulletin 2011; 62: 2588-2597.
- 461 Cózar A, Echevarría F, González-Gordillo JI, Irigoien X, Úbeda B, Hernández-León S, et al. Plastic
 462 debris in the open ocean. Proceedings of the National Academy of Sciences 2014; 111:
 463 10239.
- 464 Dai Z, Zhang H, Zhou Q, Tian Y, Chen T, Tu C, et al. Occurrence of microplastics in the water column
 465 and sediment in an inland sea affected by intensive anthropogenic activities. Environmental
 466 Pollution 2018; 242: 1557-1565.
- 467 De Falco F, Gullo MP, Gentile G, Di Pace E, Cocca M, Gelabert L, et al. Evaluation of microplastic
 468 release caused by textile washing processes of synthetic fabrics. Environmental Pollution
 469 2018; 236: 916-925.
- Dehghani S, Moore F, Akhbarizadeh R. Microplastic pollution in deposited urban dust, Tehran
 metropolis, Iran. Environmental Science and Pollution Research 2017; 24: 20360-20371.
- 472 Dris R, Gasperi J, Rocher V, Saad M, Renault N, Tassin B. Microplastic contamination in an urban
 473 area: a case study in Greater Paris. Environmental Chemistry 2015; 12: 592-599.
- 474 Dris R, Gasperi J, Saad M, Mirande C, Tassin B. Synthetic fibers in atmospheric fallout: A source of
 475 microplastics in the environment? Marine Pollution Bulletin 2016; 104: 290-293.

476 Dris R, Gasperi, J., Saad, M., Mirande, C., & Tassin, B. . Synthetic fibers in atmospheric fallout: A 477 source of microplastics in the environment? . Marine Pollution Bulletin 2016; 104(1-2): 290. 478 Eerkes-Medrano D, Thompson RC, Aldridge DC. Microplastics in freshwater systems: a review of the 479 emerging threats, identification of knowledge gaps and prioritisation of research needs. 480 Eriksen M, Mason S, Wilson S, Box C, Zellers A, Edwards W, et al. Microplastic pollution in the surface 481 waters of the Laurentian Great Lakes. Vol 77, 2013. 482 Folko A. Quantification and Characterisation of fibres emitted from common synthetic materials 483 during washing. Kappala. Stockholms Universitet, 2015. 484 Free CM, Jensen OP, Mason SA, Eriksen M, Williamson NJ, Boldgiv B. High-levels of microplastic 485 pollution in a large, remote, mountain lake. Marine Pollution Bulletin 2014; 85: 156-163. 486 Gasperi J, Dris R, Bonin T, Rocher V, Tassin B. Assessment of floating plastic debris in surface water 487 along the Seine River. Environmental Pollution 2014; 195: 163-166. 488 Gasperi J, Wright SL, Dris R, Collard F, Mandin C, Guerrouache M, et al. Microplastics in air: Are we 489 breathing it in? Current Opinion in Environmental Science & Health 2018; 1: 1-5. 490 Hartline NL, Bruce NJ, Karba SN, Ruff EO, Sonar SU, Holden PA. Microfiber Masses Recovered from 491 Conventional Machine Washing of New or Aged Garments. Environmental science & 492 technology 2016; 50: 11532. 493 Henry B, Laitala K, Klepp IG. Microfibres from apparel and home textiles: Prospects for including 494 microplastics in environmental sustainability assessment. Science of the Total Environment 495 2019; 652: 483-494. 496 Hernandez E, Nowack B, Mitrano DM. Polyester Textiles as a Source of Microplastics from 497 Households: A Mechanistic Study to Understand Microfiber Release During Washing. 498 Environmental science & technology 2017; 51: 7036-7046. 499 Horton A, Svendsen C, J. Williams R, Spurgeon D, Lahive E. Large microplastic particles in sediments 500 of tributaries of the River Thames, UK – Abundance, sources and methods for effective 501 quantification. Vol 114, 2016. 502 Hurley RR, Nizzetto L. Fate and occurrence of micro(nano)plastics in soils: Knowledge gaps and 503 possible risks. Current Opinion in Environmental Science & Health 2018; 1: 6-11. 504 Jönsson C, Levenstam Arturin O, Hanning A-C, Landin R, Holmström E, Roos S. Microplastics 505 Shedding from Textiles—Developing Analytical Method for Measurement of Shed Material 506 Representing Release during Domestic Washing. Sustainability 2018; 10: 2457. 507 Karlsson TM. Can microlitter in sediment and biota be quantified? Department of Biological and 508 Environmental Sciences. Degree project for Master of Science (2 years) in Marine Ecology. 509 Univeristy of Gothenburg, 2014. 510 Klein S, Worch E, Knepper TP. Occurrence and Spatial Distribution of Microplastics in River Shore 511 Sediments of the Rhine-Main Area in Germany. Environmental Science & Technology 2015; 512 49: 6070-6076. 513 Koelmans AA, Mohamed Nor NH, Hermsen E, Kooi M, Mintenig SM, De France J. Microplastics in 514 freshwaters and drinking water: Critical review and assessment of data quality. Water 515 Research 2019; 155: 410-422. 516 Lusher AL, Tirelli V, O'Connor I, Officer R. Microplastics in Arctic polar waters: the first reported 517 values of particles in surface and sub-surface samples. Scientific Reports 2015; 5: 14947. 518 McIlwraith HK, Lin J, Erdle LM, Mallos N, Diamond ML, Rochman CM. Capturing microfibers -519 marketed technologies reduce microfiber emissions from washing machines. Marine 520 Pollution Bulletin 2019; 139: 40-45. 521 N. Bruce N, Hartline, S. Karba, B. Ruff, S. Sonar & P. Holden. Microfiber pollution and the apparel 522 industry. IUniveristy of California Santa Barbara, Bren School of Environmental Science & 523 Management 2016. 524 Napper IE, Thompson RC. Release of synthetic microplastic plastic fibres from domestic washing 525 machines: Effects of fabric type and washing conditions. Marine Pollution Bulletin 2016; 112: 526 39-45.

- 527 Nizzetto L, Futter M, Langaas S. Are Agricultural Soils Dumps for Microplastics of Urban Origin? 528 Environmental Science & Technology 2016; 50: 10777-10779. 529 Obbard RW, Sadri S, Wong YQ, Khitun AA, Baker I, Thompson RC. Global warming releases 530 microplastic legacy frozen in Arctic Sea ice. Earth's Future 2014; 2: 315-320. 531 Okoffo ED, Ribeiro F, O'Brien JW, O'Brien S, Tscharke BJ, Gallen M, et al. Identification and 532 quantification of selected plastics in biosolids by pressurized liquid extraction combined with 533 double-shot pyrolysis gas chromatography-mass spectrometry. Science of The Total 534 Environment 2020: 136924. 535 Pauly JL, Stegmeier SJ, Allaart HA, Cheney RT, Zhang PJ, Mayer AG, et al. Inhaled cellulosic and plastic fibers found in human lung tissue. Cancer epidemiology, biomarkers & prevention : a 536 537 publication of the American Association for Cancer Research, cosponsored by the American 538 Society of Preventive Oncology 1998; 7: 419. 539 Pirc U, Vidmar M, Mozer A, Krzan A. Emissions of microplastic fibers from microfiber fleece during 540 domestic washing. Environ Sci Pollut Res Int 2016; 23: 22206-22211. 541 Prata JC. Airborne microplastics: Consequences to human health? Environmental Pollution 2018; 542 234: 115-126. 543 Salvador Cesa F, Turra A, Baruque-Ramos J. Synthetic fibers as microplastics in the marine 544 environment: A review from textile perspective with a focus on domestic washings. Science 545 of the Total Environment 2017; 598: 1116-1129. 546 Sillanpää M, Sainio P. Release of polyester and cotton fibers from textiles in machine washings. 547 Environmental Science and Pollution Research 2017; 24: 19313-19321. 548 Statistics ABo. 4602.0.55.001 - Environmental Issues - Energy Use and Conservation, Mar 2008 White 549 Goods, Canberra, 2008a. 550 Statistics ABo. 4602.0.55.001 - Environmental Issues: Energy Use and Conservation, Mar 2008. 2019. 551 Australian Commonwealth Government, Australia, 2008b. 552 Statistics ABo. B32 Tenture Type and Landlord Type by Dwelling Structure. 2019, Australia, 2011. 553 Van Cauwenberghe L, Vanreusel A, Mees J, Janssen CR. Microplastic pollution in deep-sea 554 sediments. Environmental Pollution 2013; 182: 495-499. 555 Vianello A, Jensen RL, Liu L, Vollertsen J. Simulating human exposure to indoor airborne 556 microplastics using a Breathing Thermal Manikin. Scientific Reports 2019; 9: 8670. 557 Wendy Wills AM, Angela Dickinson and Frances Short. Domestic Kitchen Practices: Findings from the 558 'Kitchen Life' study. University of Hertfordshire 2013. Wright SL, Kelly FJ. Threat to human health from environmental plastics. BMJ 2017; 358: j4334. 559 560 Zambrano MC, Pawlak JJ, Daystar J, Ankeny M, Cheng JJ, Venditti RA. Microfibers generated from the 561 laundering of cotton, rayon and polyester based fabrics and their aquatic biodegradation. Marine Pollution Bulletin 2019; 142: 394-407. 562 563 564 565 566
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Supplementary Information

Microscopy Contamination Results

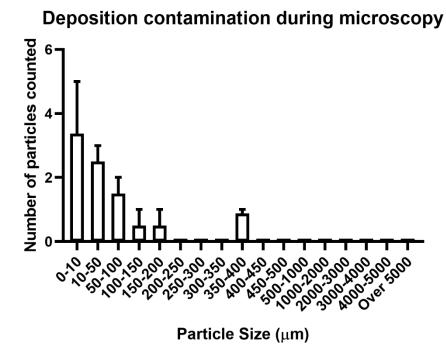
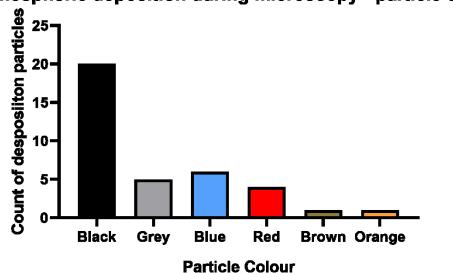
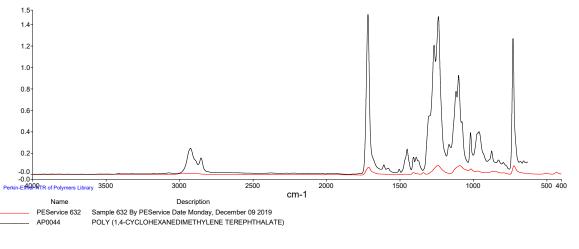


Figure S1: Size fractionation of atmospheric deposition particles within the laboratory environment during microscopy. This was undertaken as part of the QA/QC of sample analysis.



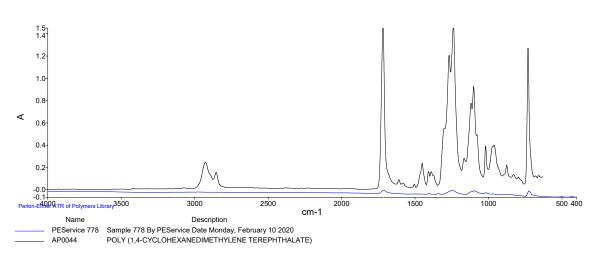
Atmospheric deposition during microscopy - particle colour

Figure S2: Colour composition of atmospheric deposition particles within the laboratory during microscopy.



Search ScoreSearch Reference Spectrum Description0.782612POLY (1,4-CYCLOHEXANEDIMETHYLENE
TEREPHTHALATE)

Figure S3: Fourier Transform Infrared Spectroscopy of the blanket sample showing an identification of Poly (1,4-Cyclohexanedimethylene) from the library search with 78% accuracy.



Search ScoreSearch Reference Spectrum Description0.808173POLY (1,4-CYCLOHEXANEDIMETHYLENE TEREPHTHALATE)

Figure S4: Fourier Transform Infrared Spectroscopy of the lint showing an identification of Poly (1,4-Cyclohexanedimethylene) from the library search with 80% accuracy.

Pyrolysis – GC/MS Results

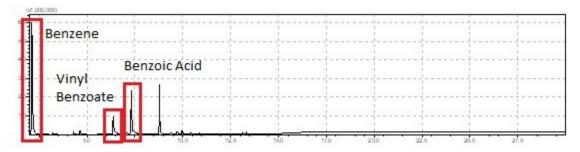


Figure S5: Pyrogram of the laundered blanket using Pyr-GC/MS for chemical characterisation. This was undertaken prior to sampling to obtain the chemical signature of the blanket for chemical identification and comparative purposes with the samples. Indicator compounds of Benzene, Vinyl Benzoate and Benzioc Acid at specific retention times identified the blanket composition to the Polyethylene Terephthalate (PET).

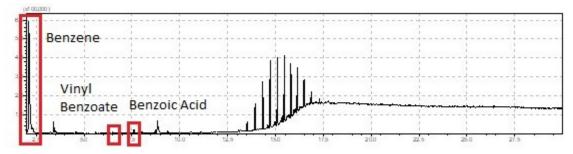


Figure S6: Pyrogram of a pool of 5 visually identified blue fibres using Pyr-GC/MS (n=5). Indicator ions of Benzene, Vinyl Bezoate and Benzoic Acid at the same retention times, characteristic of PET were identified to confirm chemically that the selected blue fibres were sourced from the blanket.

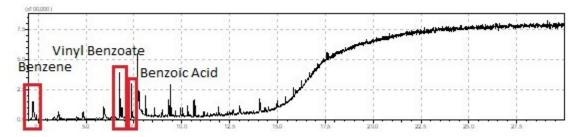


Figure S7: Pyrogram of a pool of 3 visually identified blue fibres from Pyr-GC/MS (n=3). Indicator ions of Benzene, Vinyl Benzoate and Benzioc Acid at the same retention times, again showed consistency that the visually identified blue fibres were also of PET composition.