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Origin of Synthetic Particles in Honeys

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A total of 47 honeys and 22 flowering plants was analysed for their load of synthetic fibres and fragments. In all samples investigated foreign particles were found. These include also black carbon particles which were not enumerated. Fibres and fragments ranged from 10 to 336 kg^{-1} and 2 to 82 kg^{-1} honey, respectively. The data of the flowering plants analysed indicate that a major proportion of the particle load may originate from external sources, *i.e.* these particles are brought into the beehive by the worker bees during nectar collection.

INTRODUCTION

Honey, despite it being an almost non-processed aliment, has been shown to contain contaminants such as trace metals [*e.g.* Formicki *et al*., 2013; Matusevicius *et al*., 2010], pesticides [Al-Waili *et al*., 2012], chlorinated compounds [Matusevicius *et al*., 2010; Rissato *et al*., 2007] and others [Bogdanov, 2006].

Honey has also been reported to contain filth of natural origin [Canale *et al*., 2014]. These authors noted the presence of carbon particles, other inorganic fragments and fragments of animal origin. These included insects, their cuticular fragments, mites and mammal hair. The abundance of this foreign matter showed no difference for honeys from both small and large-sized producers. Liebezeit & Liebezeit [2013] noted the presence of particles of synthetic origin in honeys available in Germany. Here also no differences were observed between small beekeeping enterprises and large producers.

The latter authors also suggested possible pathways by which this contamination may reach the final product. These include external and internal sources. The internal sources can be related to the diligence shown during honey extraction and packaging. In this case, the extent of contamination can be controlled by the processing facility. The external source as was shown by the analysis of flowers is related to the presence of particles in the atmosphere that may deposit on flowers there being held by the pollenkitt.

In this communication we present more data on the presence of synthetic particles in honeys as well as in external sources, *i.e.* flowers.

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MATERIAL AND METHODS

A total of 47 honey samples was obtained from supermarkets as well as directly from German small-scale beekeepers. The honeys investigated are given in Table 1. Liquid honeys were mixed with a an equal volume of 40°C water while solid ones were heated to 40°C until liquified and then diluted. The mixtures were passed through a 40 μm steel sieve. The material remaining on the sieve was copiously rinsed with warm deionised water and, after transfer with 30% H_2O_2 to a 50 mL wide neck Erlenmeyer beaker and cooling to room temperature, treated at room temperature for 72 h. The oxidised samples were then filtered over 0.8 μm grey, gridded cellulose nitrate filters, treated several times with a small volume of 90°C water to remove waxy particles and dried at ambient temperature. One series of 12 samples was analysed in duplicate.

Inflorescences of 22 species were collected from April 2014 to August 2014 (Table 2). Depending on the size of the inflorescence, between 1 and 25 specimens were completely covered with 30% H_2O_2 for 24 h. After removal of the larger parts such as petals the peroxide solution was sieved over a 40 µm steel sieve. After copious rinsing, the remaining particulate material was filtered over 0.8 µm cellulose nitrate filters.

After extensive rinsing with 0.8 µm filtered deionised water, wet filters were covered with 6 mL Rose Bengal (4,5,6,7-tetrachloro-2',4',5',7'-tetraiodofluorescein, 200 mg/L; [Lusher *et al*., 2013; Williams & Williams, 1974] to stain natural organic particles. After 5 min, the dye was filtered and the stained material washed dye-free with filtered deionised water. After drying at ambient temperature, the samples were analysed under a dissecting microscope at up to 80x magnification. No attempts were made to determine fibre lengths or polymer type. Particles that were not stained are regarded as being

TABLE 1. Summary of honeys analysed.

No.	Type	Origin	State
1	mountain	Switzerland	solid
2	not specified	Switzerland	solid
3	not specified	Switzerland	solid
4	chestnut	Switzerland	solid
5	flower	EU/non-EU	solid
6	flower	EU/non-EU	solid
7	forest	Switzerland	solid
8	forest	Switzerland	liquid
9	flower	Switzerland	liquid
10	spring	Switzerland	solid
11	country	EU/non-EU	solid
12	flower	EU/non-EU	solid
13	flower	EU/non-EU	solid
14	flower	Bulgaria	solid
15	flower	Switzerland	solid
16	not specified	EU/non-EU	solid
17	summer flower	EU/non-EU	solid
18	flower	EU/non-EU	solid
19	not specified	Toscana	solid
20	not specified	Switzerland	solid
21	not specified	Germany	solid
22	Millefiori	Italy	solid
23	chestnut	Italy	liquid
24	sunflower	Italy	solid
25	orange	Italy	solid
26	acacia	Bulgaria	liquid
27	eucalyptus	Spain	liquid
28	honeydew	Italy	liquid
29	chestnut	Spain	liquid
30	linden	Bulgaria	liquid
31	coriander	Bulgaria	liquid
32	Lavendulum	Spain	liquid
33	mountain	Spain	liquid
34	not specified	Latin America/E Europe	liquid
35	not specified	EU/non-EU	liquid
36	mountain	Spain/France	liquid
37	flower	EU/non-EU	liquid
38	not specified	EU/non-EU	liquid
39	not specified	Latin America	solid
40	mountain	France	solid
41	flower	France	liquid
42	not specified	EU/non-EU	liquid
43	not specified	France	liquid
44	flower	France	liquid
45	flower	Latin America	liquid
46	flower	Germany	liquid
47	fruit blossoms	Germany	solid

of synthetic origin and will be referred to in the following as microplastics using the definition for this material as being particles smaller than 5 mm but larger than 1 μm.

As the filters used were found to be occasionally contaminated with fibres, presumably from the production process, they were rinsed with deionised filtered water and checked under the microscope prior to use. To avoid airborne contamination [Liebezeit & Liebezeit, 2013] the filtration unit and all other glassware used was covered during the whole workup procedure. Maximum exposure time to the atmosphere including microscopic analysis was 20 to 30 min. Blank filters carried through the complete scheme gave a maximum of 2 fibres/filter. Laboratory air sucked through a previously rinsed filter for 1 h at 50 L/min had 2 fibres, 0 fragments and 3 small granular particles [Liebezeit & Liebezeit, 2014]. Hence, contamination from this source can be neglected. All water used as well as the peroxide solution were filtered over 0.8 μm cellulose nitrate filters prior to use.

Some pollen types and also some string-like fibers were not stained under the conditions given above. Nevertheless these can be easily distinguished by their morphology from the more irregular microplastic particles. The same holds for chitin which is also not stained by Rose Bengal. Here again morphology and discolouration can be used to exclude these particles from the microplastic counts.

As quartz grains were observed in the flowers analysed as well as in some cases in honey samples only fibre and fragment counts will be reported here.

RESULTS AND DISCUSSION

In all 47 honey samples analysed both fibres and fragments were found ranging from 10 to 336 fibres/kg and 2 to 82 fragments/kg (Figure 1). Fibres are clearly dominating (Figure 2) with lengths from 40 μm up to several millimeters (Figure 3). Fragments were generally smaller in size reaching only some ten micrometers. For the 12 honeys investigated in duplicate (Figure 1) relative variations between samples ranged from 0 to 57.2% for fibres and 0 and 55.5% for fragments indicative of an inhomogeneous distribution of particles in the packaged product.

In addition, black carbon particles were frequently encountered. Most of these were granular in shape and are hence not reported in detail here. These findings, however, corroborate the data by Canale *et al*. [2014] who also found large number of carbon particles in their samples of Italian honeys.

Comparing samples from smaller beekeepers with those available from large scale producers did not show significant differences. This indicates that honey harvesting, processing and packaging do not contribute to a large extent to the foreign particle load. Canale *et al*. [2014] reported insects and mammal hair in some of their samples providing evidence for poor operating conditions during honey processing and packaging. In the present suite of samples no intact insects or insect remains were found while in two samples one hair each was noted. This might be due to the fact that the larger producers screen their products through 200 μm sieves, a practice which, at least in Germany, is also exercised by small beekeepers.

In all flowering plants analysed particles were found that could not be stained by Rose Bengal and hence can be considered to be of synthetic nature (Table 2; Figure 4). Furthermore, in several filaments of untreated lily blossoms (*Lilium* sp.) fibres were observed (Figure 4). As in the honeys analysed fibres were dominant also in all plant species (Table 2) making up on average 77.9±19.0% (n=27). This is direct evidence for the deposition of foreign particulate matter on flowers there being held by the pollenkitt and other pollen coatings [Hesse, 2010]. That this foreign material is transferred to the hive by pollen collecting bees is also evident from the finding of fibres in their pollen load. Here of six investigated specimens two were carrying fibres.

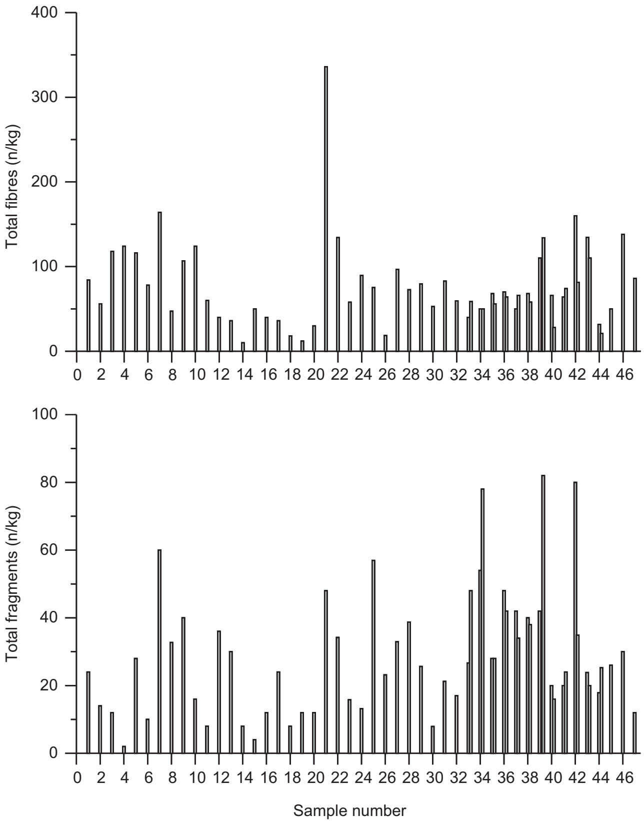

FIGURE 1. Fibre and fragment numbers in 47 honey samples. Numbers refer to Table 1.

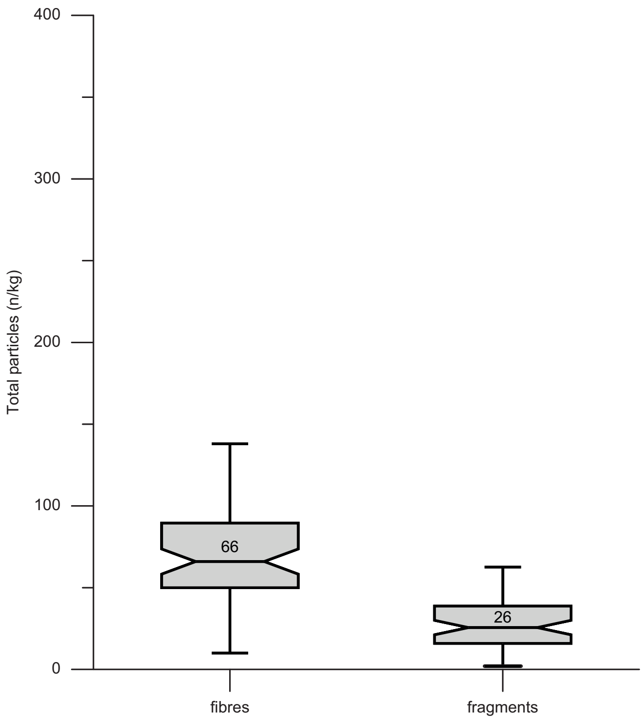

FIGURE 2. Median, upper and lower quartiles, 95% values and outliers for 47 honey samples.

Our results clearly indicate that fibres and, to a lesser extent, fragments are already present in the bees' feed and are transferred from the blossoms to the hive by the insects. Particles in the atmosphere are derived from a variety of sources including abrasion of clothing. Also synthetic particles used to improve horticultural soils such as polystyrene peat or hygro peat may become airborne by outblowing after dry periods. The same may hold for synthetic fibres present in sewage sludge used as fertilizer in agriculture [Habib *et al*., 1998; Zubris & Richards, 2005].

Little information is available on the content of organic fibres, both natural and synthetic, in ambient air. Altree-Williams & Preston [1985] found contents from <1000 to 63,000 fibres m^{-3}. Data provided by Schneider *et al*. [1996], using optical microscopy, support the finding that organic fibers in indoor air occur in concentrations larger than those of "other inorganic" fibres.

Synthetic organic fibers include polyethylene, polypropylene, polyvinylalcohol, polyester, polyamide and polytetrafluoroethylene fibers [Hodgson, 1993]. Fragmentation of macroplastic litter due to the action of sunlight, oxygen, humidity and temperature [Barnes *et al*., 2009] or agricultural films may lead to the formation of fragments in the sub-millimetre range. These will, as other particles in this size range, become easily airborne. Especially for agricultural films the claim that these are degraded is based on the fact that some physical properties such as tensile strength or elongation at break change as films age. The formation of final degradation products such as water or carbon dioxide is only rarely assessed.

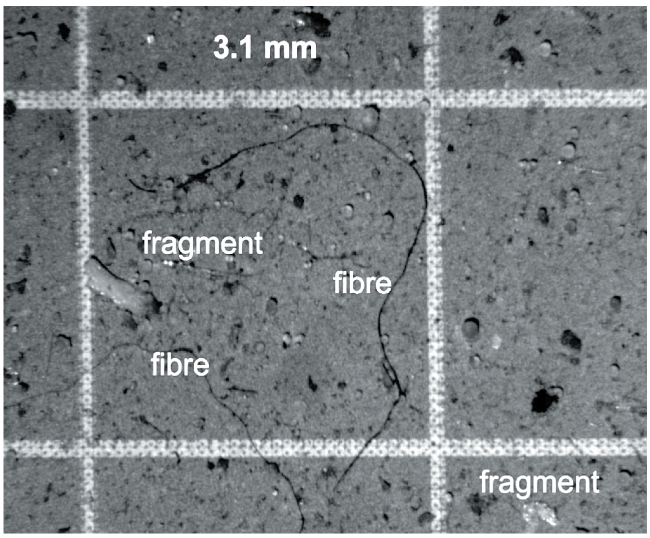

FIGURE 3. Example of fibres and fragments in honey.

TABLE 2. Summary of flowering plants analysed.

Species	Inflorescences analysed	Microplastic particles	
		fibres	fragments
Aquilegia	5	5	2
Aquilegia vulgaris	3	9	3
Daisy *Bellis perennis*	10	12	2
Cornflower	4	28	11
Centaurea cyanus	4	8	4
Cornel cherry *Cornus mas*	10	5	0
Crocus	4	50	4
Crocus sp.	4	40	5
Hazelnut *Corylus avellana*	5	16	6
Hawthorn *Crataegus* sp.	25	0	1
Snowdrop *Galanthus* sp.	8	14	0
Hyazinth *Hyacinthus* sp.	1	9	2
Magnolia *Magnolia* sp.	5	5	1
Daffodil, large *Narcissus* sp.	3	21	4
Forgetmenot *Myosotis* sp.	12	17	4
Daffodil, small *Narcissus* sp.	5	23	6
Primrose *Primula* sp.	4	51	6
Plum *Prunus* sp.	14	14	2
Sour cherry *Prunus cerasus*	10	13	2
Lungwort *Pulmonaria officinalis*	6	38	4
Rose	3	39	2
Rosa sp.	3	6	4
Willow *Salix* sp.	8	24	3
Dandelion *Taraxacum officinale*	5	4	2
Tulip *Tulipa* sp.	3	15	7

Particles >10 μm, referred to as giant or ultragiant particles [cf. Johnson, 1982] have been reported to be present in the atmosphere. For mineral particles >75 μm long range transport has been shown by Betzer *et al*. [1988].

While most studies related to these particles determination for only the elemental composition, de Bock *et al*. [1994] reported on the presence of organic constituents in North Sea particles >20 μm but did not investigate their exact nature. Van Malderen *et al*. [1992] noted a markedly higher abundance of giant organic particles in air masses with a continental origin.

Black carbon or soot particles which were regularly found in the present suite of samples originate from combustion processes such as forest fires or fossil fuel burning. These particles may be transported over very long distances [*e.g.* Ramanathan *et al.,* 2007].

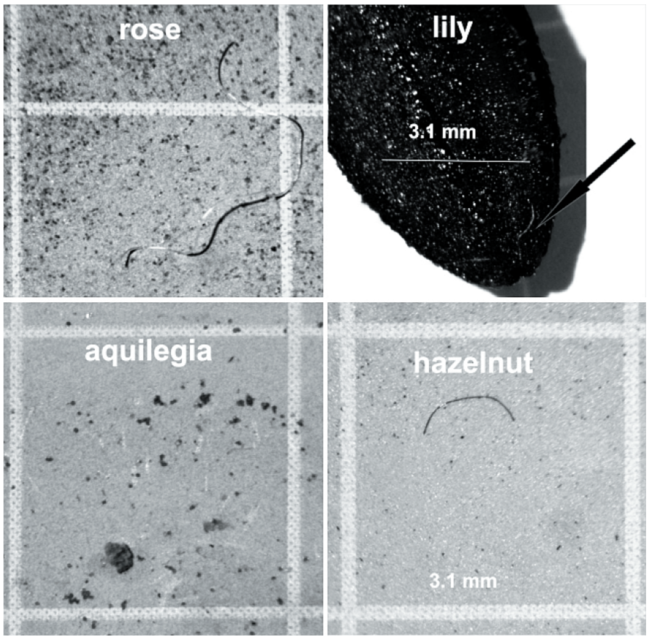

FIGURE 4. Examples of foreign particles in various flowers. The lily filament in the upper right part was photographed directly without further treatment.

These data suggest that the global environment is affected to a large extent by microplastic particles either from direct inputs or from fragmentation of macrolitter.

While an atmospheric source for the particles is proven by our findings the exact contribution to the overall foreign particle load in honey is still to be determined. The finding of Canale *et al*. [2014] of insects or mammal hair or the data on fibre content of indoor air suggest that improper handling of the product or the ubiquitous presence of airborne particles may also contribute to the overall particle burden of honey. This can surely be reduced by the beekeeper using adequate processing techniques.

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