TITLE:

Sampling, Identification and Characterization of Microplastics Release from Polypropylene Baby Feeding Bottle during Daily Use

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SUMMARY:

This study detailed a reliable and cost-effective protocol for microplastics collection and detection from the daily use of plastic products.

ABSTRACT:

Microplastics (MPs) are becoming a global concern due to the potential risk to human health. Case studies of plastic products (i.e., plastic single-use cups and kettles) indicate that MP release during daily use can be extremely high. Precisely determining the MP release level is a crucial step to identify and quantify the exposure source and assess/control the corresponding risks stemming from this exposure. Though protocols for measuring MP levels in marine or freshwater

has been well developed, the conditions experienced by household plastic products can vary widely. Many plastic products are exposed to frequent high temperatures (up to 100 °C) and are cooled back to room temperature during daily use. It is therefore crucial to develop a sampling protocol that mimics the actual daily-use scenario for each particular product. This study focused on widely used polypropylene-based baby feeding bottles to develop a cost-effective protocol for MP release studies of many plastic products. The protocol developed here enables: 1) prevention of the potential contamination during sampling and detection; 2) realistic implementation of daily-use scenarios and accurate collection of the MPs released from baby feeding bottles based on WHO guidelines; and 3) cost-effective chemical determination and physical topography mapping of MPs released from baby feeding bottles. Based on this protocol, the recovery percentage using standard polystyrene MP (diameter of 2 µm) was 92.4-101.2% while the detected size was around 102.2% of the designed size. The protocol detailed here provides a reliable and cost-effective method for MPs sample preparation and detection, which can substantially benefit future studies of MP release from plastic products.

INTRODUCTION:

Most types of plastics are non-biodegradable but can break down into small pieces due to chemical and physical processes such as oxidation and mechanical friction^{1,2}. Plastic pieces smaller than 5 mm are classified as microplastics (MPs). MPs are ubiquitous and found in almost every corner in the world. They have become a global concern due to the potential risk to humans and wildlife^{3,4}. To date, significant accumulations of MPs have been found in fish, birds, insects^{5,6} as well as mammals (mouse, in the gut, kidney and liver^{7,8}). Studies found the exposure and accumulation of MPs can damage the lipid metabolism of mice^{7,8}. A risk assessment focusing on fish found that sub-micron MPs can penetrate the blood-to-brain barrier and cause brain damage⁹. It should be noted that to date all MPs risk results have been obtained from animal studies while the specific risk to human health is still unknown.

In the last 2 years, concerns about the MP threat to human health substantially increased with the confirmation of the levels of human exposure to MPs. The accumulation of MPs has been found in the human colon¹⁰, the placenta of pregnant women¹¹ and adult stool¹². A precise determination of MP release levels is crucial to identify exposure sources, assess the health risk and evaluate the efficiency of any potential control measures. In the last few years, some case studies reported that daily-use plastics (i.e., plastic kettle¹³ and single-use cup¹⁴) can release extremely high quantities of MPs. For example, disposable paper cups (with interiors laminated with polyethylene-PE or copolymer films), released approximately 250 micron-sized MPs and 102 million sub-micron-sized particles into each milliliter of liquid following exposure to 85-90 °C hot water¹⁴. A study of polypropylene (PP) food containers reported that up to 7.6 mg of plastic particles is released from the container during a single use¹⁵. Even higher level were recorded from teabags made from polyethylene terephthalate (PET) and nylon, which released approximately 11.6 billion MPs and 3.1 billion nano-sized MPs into a single cup (10 mL) of the beverage¹⁶. Given that these daily-use plastic products are designed for food and beverage preparation, the release of high quantities of MPs is likely and their consumption is a potential threat to human health.

Studies on MP release from household plastic products (i.e., plastic kettle¹³ and single-use cup¹⁴) are at an early stage, but it is expectable that this topic will receive increasing attention from researchers and the general public. The methods required in these studies are significantly different from those used in room temperature marine or freshwater studies where well-established protocols already exist¹⁷. In contrast, studies involving the daily-use of household plastic products involves much higher temperature (up to 100 °C), with in many cases repeated cycling back to room temperature. Previous studies pointed out that plastics in contact with hot water can release millions of MPs^{16,18}. In addition, the daily-use of plastic products may over time change the properties of the plastic itself. It is therefore crucial to develop a sampling protocol that accurately mimics the most common daily-use scenarios. The detection of micro-sized particles is another major challenge. Previous studies pointed out that MPs release from plastic products are smaller than 20 µm^{16,19,20}. Detection of these types of MPs requires the use of smooth membrane filters with small pore size. In addition, it is necessary to distinguish MPs from possible contaminants captured by the filter. High sensitivity Raman spectroscopy is used for chemical composition analysis, which has the advantage of avoiding the need for high laser power that is known to easily destroy small particles²⁰. Hence, the protocol must combine contamination-free handling procedures with the use of optimal membrane filters and for a characterization method that allows fast and accurate MP identification.

The study reported here focused on PP-based baby feeding bottle (BFB), one of the most commonly used plastic products in daily life. It was found that high number of MPs are released from plastic BFB during formula preparation¹⁸. To benefit further study of MPs release from daily plastics, the sample preparation and detection method for BFB is detailed here. During sample preparation, the standard formula-preparation process (cleaning, sterilizing and mixing) recommended by the WHO²¹ was carefully followed. By designing the protocols around the WHO guidelines, we ensured the MPs release from BFBs mimicked the baby formula preparation process used by parents. The filter process was designed to accurately collect the MPs released from BFBs. For the chemical identification of MPs, the working conditions for Raman spectroscopy were optimized to obtain clean and easily identified spectra of MPs, while at the same time avoiding the possibility of burning the target particles. Finally, the optimum test procedure and applied force to allow accurate 3-dimensional topography mapping of MPs using atomic force microscopy (AFM) was developed. The protocol (**Figure 1**) detailed here provides a reliable and cost-effective method for MPs sample preparation and detection, which can substantially benefit future studies of plastic products.

PROTOCOL:

1. Hot water preparation

1.1) For all hardware that comes into contact with the samples, use clean glass made of borosilicate 3.3 to prevent any potential contamination. Thoroughly clean all the glassware.

Caution: The pre-existing scratches or imperfection spots on glassware can release particles during heating and shaking process. We suggest the users check the glassware and avoid the use

of the scratched glassware. Our comparison of glassware made of different glasses (such as soda-lime and borosilicate) showed that borosilicate 3.3 releases the lowest quantity of glass particles (can be screened for by Raman spectroscopy), and we recommend the use of borosilicate 3.3 glassware in all tests.

1.2) Pour 360 mL of DI water into a glass beaker. Cover the beaker with a clean glass disk, and then move it into a brand-new microwave oven and heat for 2.5 minutes at full oven power. After gently shaking to remove any potential temperature gradients due to uneven heating, the temperature of the water inside of the beaker is 70 °C and ready for sample preparation.

1.3) Prepare 95 °C water for BFB sterilization by pouring 1 L of DI water in glassware and heating in a microwave oven for 14 minutes.

Caution: Never use plastic kettles to prepare hot water. The plastic kettle itself releases millions of MPs into the hot water during the boiling process¹³.

2. MPs release during formula preparation

NOTE: Carefully following the standard formula-preparation process (cleaning, sterilizing and mixing) recommended by the WHO²¹, the MPs release from BFBs during formula preparation is mimicked in the following 3 steps.

2.1) Collect brand-new BFB products from pharmacy stores and clean them thoroughly after removing the product from its packaging. Wash each BFB using detergent water (repeat 3 times at room temperature-RT) and distilled water (repeated 3 times, RT). Finally, rinse the BFB 3 times using DI water at RT.

Caution: Do not clean the BFB using sonication. Though sonication is widely used in laboratories for mixing and cleaning, the sonication of BFB can severely damage the bottle surface and cause the MPs release from PP products within 1 minute.

2.2) Soak the BFB in 95 °C DI water (Section 1.3) to sterilize the bottle. To avoid the floating of the BFB, slightly press the exterior of the BFB using a stainless-steel tweezer and ensure the whole bottle body immersing in the water.

2.2.1. After 5 minutes, take out the bottle and move it to a clean glass disk. During the air dry step, invert the bottle on the glass disk until there is no evidence of droplets.

2.3) Pour 180 mL hot DI water (70 °C, obtain from Section 1.2, corresponding to WHO guidelines) into the air-dried bottle. Then cover the bottle immediately using a glass petri dish and place it into a shaking bed.

2.3.1. To simulate the formula mixing process, shake the bottle at a speed of 180 rpm for 60 seconds. After shaking, move the bottle to a clean glass plate and allow it to cool down.

3. Sample preparation for MPs identification and quantification

3.1) Sonicate and thoroughly rinse all parts of the glass filter (diameter of 25 mm, glass funnel, fritted glass support base and receiver flask) using DI water.

3.1.1. Place a piece of gold-coated polycarbonate-PC membrane filter (pore size of 0.8 μm , Au coating layer thickness of 40 nm) in the middle of glass base.

3.1.2. Assemble the glass funnel and stainless-steel clamp to fix the membrane filter. Finally connect the assembled glass filter to a vacuum pump (**Figure 2**).

Caution: To ensure the membrane smoothly sticks onto on the surface of the glass base it is important to keep the glass base wet. If necessary, 1-2 drops of DI water should be dropped on the surface of the glass base before placing down the membrane filter.

3.2) Carefully mix the cooled water sample in the BFB (obtain from Section 2.3), and then transfer a certain amount of the water sample to the glass funnel using a glass pipette. Switch on the vacuum pump to allow the water sample to filter through the membrane filter slowly.

3.2.1. After filtering, wash the interior of the glass funnel using DI water to ensure there are no particles sticking on the funnel.

Caution: To avoid the overlap of the particles on the surface of the membrane filter, it is important to carefully choose the correct volume of water that is passed through the filter. BFBs release large number of particles, so that 3-5 membrane filters are needed to filter the entire volume of the water sample.

3.3) Disconnect the vacuum pump and disassemble the glass filter. Then carefully take out the membrane filter using a stainless-steel tweezer and move it to a clean cover glass. Fix the membrane filter on the cover glass using a small piece of paper tape. Immediately store the sample in a clean glass petri dish.

4. Sample preparation for AFM topography characterization

4.1) Prepare a clean silicon wafer. Drop a 50 μL water sample (obtain from Section 2.3) on the surface of the silicon wafer and dry it in an oven at a temperature of around 103 $^{\circ}\text{C}$. Repeat this process if the MPs level in the water sample is low.

4.2) After 1-hour drying time, move the wafer to a clean glass Petri dish and allow it to cool down in a desiccator.

4.3) After the wafer has cooled, store the sample in a dry place.

5. MP identification and quantification using Raman spectroscopy

5.1) Calibrate the Raman system using zero-order correction and silicon wafer. Ensure the peak location of silicon wafer is at 520.7 cm^{-1} and peak intensity is higher than 6000 a.u. when the laser intensity is at 100%.

5.2) Set up the parameters of the Raman system to obtain high signal-to-noise MP spectra while avoiding the burning of MPs. Set the system as follows: 532 nm excitation laser, remove cosmic ray, laser intensity of 10% (laser power of 0.18 mW), spectral resolution of 1.5 cm^{-1} , exposure time of 10-20 seconds, accumulations of 10-40 times and spectral range of 200-3200 cm^{-1} . **Figure 3** showed typical spectra of MPs with accumulation times from 1s to 400s.

Caution: Do not test particles using 100% laser directly to avoid the rapid-burning (can be burned in 1 minute if the particle is small). Use low intensity (10-50%) to conduct the test first.

5.3) Place the filter sample (obtained from Section 3.3) in the middle of the Raman sample stage. Choose four representative spots (2 spots are in the middle area while other 2 spots close to the edge of working area. For details see **Figure 3C**) on the membrane filter to conduct the test (total test area around 1.5 mm^2).

5.4) Observe and photograph the particles on the surface of the membrane filter using an optical microscope (100x) followed by chemical identification using Raman spectroscopy.

5.4.1. Compare the Raman spectrum obtained to the reference standard PP spectra (from bulk material of BFB and previous publication²²).

5.4.2. Determine the particles' chemical property using the intensive peaks in the range of 2780-2980, 1400-1640 and 709-850 cm^{-1} , corresponding to the stretching vibrations of CH/CH₂/CH₃ and C-C groups associated with polymer materials (**Figure 3**).

5.5) Analyze the size and quantity of the identified MPs using ImageJ software package.

5.5.1. Obtain the MPs concentration in water sample based on the tested area, total working area (227 mm^2) and the known filtered sample volume.

5.5.2. Classify the confirmed MPs into 5 groups in terms of the size: 0.8-5 μm , 5-20 μm , 20-50 μm , 50-100 μm and $> 100\text{ }\mu\text{m}$.

5.5.3. Finally, determine the MPs quantity in one litre of water sample based on the filtered sample volume, number of MPs recorded and tested area of the membrane filter.

6. MP topographic characterization using AFM

6.1) Equip the AFM system (NT-MDT) with a tapping mode probe. Calibrate the system using

step height standard (SHS). Set up the system within optimal work conditions: the scan rate is 1 Hz and the scan size is 10-50 μm , the tuning frequency is around 160 KHz, the scan line is 512 pixels.

6.2) Fix the silicon wafer (obtain from Section 4.3) on the AFM sample stage. Observe and photograph the target particles on the surface of the silicon wafer, followed by chemical identification using the method in Section 5.

6.3) Switch system to AFM mode (note: the Raman spectroscopy and AFM are assembled in one system) and test the topography of identified MPs.

6.4) Analyze the 3d data using Gwyddion 2.54 software. Use the option of *profile* to obtain the particle dimensions and average heights while *3D view* to obtain 3D structure.

REPRESENTATIVE RESULTS:

To validate this protocol, the water sample was prepared by adding standard polystyrene microplastic spheres (a diameter of $2.0 \pm 0.1 \mu\text{m}$) to DI water. The MPs quantity added corresponded to 4,500,000 particles/L, which is similar to the MPs release level from BFBs. Following the protocol Sections 2-3, the MPs were successfully collected (**Figure 4A**) and the recovery rate was 92.4-101.2%. This recovery rate is comparable to a previous study on MPs²³. Using ImageJ software, the detected diameter of standard MPs was $2.04 \pm 0.08 \mu\text{m}$ (where $\pm$ represents standard error of the mean value), which is around 102.2% of the designed size ($2.0 \pm 0.1 \mu\text{m}$). Meanwhile, the potential interference from other types of MPs, such as PP and PE, was also tested for but none was found in these standard PS water samples. Hence, the developed protocol avoids contamination and is a reliable test of MP release from BFBs.

This protocol was used to test the MPs release from eight popular BFB products. **Figure 4B** showed the typical MPs collected on the surface of the membrane filter. During the chemical determination using Raman spectroscopy (**Figure 3**), the peaks in the range of $2830\text{-}2970 \text{ cm}^{-1}$ became more and more significant with the increase accumulation time. These peaks reflect the stretching vibrations of $\text{CH}/\text{CH}_2/\text{CH}_3$ groups, which can be used to identify MPs. A high number of MPs were release during the use of BFBs. The MPs levels ranged from 1.31 million to 16.20 million particles per liter (**Figure 5**). This result is 3-5 orders of magnitude higher than the previously reported levels of MPs in drinking water²⁴. It is evident that the vulnerable babies likely experience high levels of MPs exposure.

Figure 6 shows the typical topography maps of MPs recorded using protocol Sections 1, 2, 4 and 6. For the large MP of around $8 \mu\text{m}$ -lateral size (P1 in **Figure 6**), the average thickness is $0.82 \mu\text{m}$. For smaller MP around $3 \mu\text{m}$ in lateral size (P2 in **Figure 6**), the thickness is close to $0.25 \mu\text{m}$. In general, the thickness of MPs release from BFB is around a tenth of the lateral size. It is also noticeable that the surface texture of MPs is rich with nano-sized bumps and valleys, which can substantially increase their absorption capacity. Previous studies found that MPs are the effective carriers for pollutants, such as pesticide^{25,26}. The observed topography of the MPs found here is likely an important contributor to the high carrying capacity of MPs.

FIGURE AND TABLE LEGENDS:

Figure 1: The diagram of sample preparation and test.

Figure 2: Assembly of the glass filter and pump. 1-cooled water sample in BFB; 2-assembled glass filter; 3-glass transfer pipette; 4-vacuum pump; 5- reciprocal shaker.

Figure 3: Typical Raman spectra for MPs determination. (A) The Raman spectra of bulk piece from BFB, membrane filter and MPs on the membrane filter, respectively. (B) The Raman spectra of one potential MP with different acquisition time (1s, 10s, 100s, 400s). (C) The representative spots tested. The total diameter of filter membrane is 25 mm of diameter with 17 mm-diameter real working area. The four white boxes indicate the full representative spots for Raman test. 2 spots are in the middle area while other 2 spots close to the edge of working area. In total, the tested area of four spots is 1.5 mm².

Figure 4: Typical optical image of standard PS MPs and MPs release from BFB, respectively. (A) The optical image of standard PS MPs. The particle inside of red box was confirmed as typical PS MP. (B) The optical image of MPs release from BFBs. The particle inside of red box was confirmed as typical MP.

Figure 5: Quantity of MPs released from plastic BFB products. 8 popular products were chosen at the study. Error bar indicates the standard error of the mean value.

Figure 6: Typical 3D image of MPs release from BFB. (A) AFM image of typical MPs release from BFB. (B) Extracted cross-section profiles of the MPs. (C) The 3D topographic image of the released MPs.

DISCUSSION:

Though the study of MPs in marine and freshwater has been widely reported and the relevant standard protocol has been developed¹⁷, the study of daily-use plastic products is an important emerging research area. The different environmental conditions experienced by household plastic products means that extra care and efforts are needed to obtain reliable results. The study protocol must be consistent with the real daily use scenarios. For example, sonication is widely used in lab-tests to clean samples. However, it was found that 1 minute sonication can severely damage the BFB's surface, resulting in an order of magnitude higher levels of MP release. Similar polymer breakage due to sonication was also reported previously²⁷, which indicates that sonication is not a suitable cleaning method for plastic sample preparation in MP studies.

In addition, potential contamination sources must be identified and eliminated. Kettles are widely used to prepare hot water, which is necessary for the BFB test. However, a single boil can generate up to 30 million particles per liter in a plastic kettle¹³. Microwave ovens are non-contact method to prepare hot water once care is taken to eliminate local heating. For filtration, a glass transfer pipette rather than the plastic one (usually made of PP) is recommended. For the brand-new PP products, it has been reported that a high quantity of MPs is attached to the surface due

to the manufacturing process ¹⁵ so care must be taken to properly clean all products before testing begins. In summary, the researcher must be vigilant to avoid any procedure that can adversely influence the measured levels of MP release from BFBs.

It should be noted that the protocol cannot account for all types of MP release. Due to the use of a filter with 0.8 µm pore size, nanoparticles smaller than 0.8 µm are beyond the scope of this method. In addition, individual parents might not follow the WHO guidelines on which our protocol is based so that in real life the MPs level in prepared formula could be significantly different from that reported here.

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DISCLOSURES:

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