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MICROPLASTIC CONTAMINATION IN COMMERCIAL SEA SALT OF VIET NAM

Dang Thi Ha

Faculty of Chemistry and Food Technology, Ba Ria - Vung Tau University, 80 Truong Cong Dinh, W3, VungTau City, Ba Ria – Vung Tau province

Email: *leha1645@yahoo.com*

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Abstract. This is the first study which assessed the microplastic contamination of sea salt products from Viet Nam. The results obtained from 9 iodate fine table sea salt and 4 raw sea salt samples collected from different salt production regions along Viet Nam showed that microplastics were present in 9/9 of the salt samples. The mean abundance of microplastics was 878 ± 101 items/kg and 340 ± 26 items/kg for raw and fine sea salts, respectively. For both raw and fine sea salts, fibers were the predominant type of microplastics, accounting for more than 60 % of total microplastic particles. In addition, three types of polymer were detected in 12 microplastic particles by FTIR, including polyethylene (PE), polypropylene (PP) and polystyrene (PS), among which, the most common is PE (accounting for 67 %). The results, gathered from this research, evidence the microplastic contamination within Vietnamese sea salt, and thus, become crucial to develop more future research, leading to a better understanding of the risks associated with salt consumption as well as measures to reduce microplastic contamination in sea salt.

Keywords: microplastic, contamination, sea salt, Viet Nam.

Classification numbers: 1.4.4, 3.3.2.

1. INTRODUCTION

Plastics and products from plastics has changed people's life with its durability, convenience and low price, especially in a developing country as Viet Nam [1, 2]. Plastics have become an indispensable part of modern people's life when it appears in almost everyday things like bags, food containers, drinks, household appliances, synthetic textiles, etc. Along with the dominance of plastics, plastic waste has caused extremely dangerous consequences for the environment and ecosystems [1 - 4]. According to the statistics of the United Nations Environment Program published at the International Consulting Workshop on the Development of national action plan on marine plastic debris management in December 2018, every year, Viet Nam discharges to the ocean from 0.28 to 0.73 million tons of plastic waste (~ 6 % of the world total), ranked the 4th in the world, after China, Indonesia and Philippines [1]. Despite growing awareness of the dangers of this contamination, knowledge of the sources and pathways of marine plastics is still quite limited.

Marine plastics in particularly and waste plastics in general can be divided into two groups: macroplastics (≥ 5 mm in size), which usually enter the marine environment in their manufactured sizes; and microplastics – MiP (< 5 mm in size), which originate from the degradation of macroplastics, or are directly released into the environment in the form of small particles (*e.g.*, microplastic particles are added voluntarily to products such as scrubbing agents in toiletries and cosmetics) [2 - 5].

The plastic particles persist for a very long time in the environment due to its degradation process is slow. In addition, the plastic particles can adsorb different contaminants (*e.g*., persistent organic pollutants - POPs, non-metals, additives/monomers, and heavy metals, etc.) from the water and transfer them to the aqua-products (fish, bivalve organisms or salt, etc.) [4 - 6]. Therefore, plastic particles in the environment can result in negative impact on the organisms, such as health risk associated with ingestion of the pollutants; or the microplastic indigestion could also decrease energy reserves, inhibition or reduction of feeding/filtering activity, disrupt the endocrine and reproductive systems, translocation to the circulatory system, and increase toxic load in smaller organisms [6 - 8]. Finally, microplastics can reach the human organism through numerous types of plastic contaminated food, such us salt, fish and mussels, etc.

The presence of microplastics in the sea salt will be a threat to food safety. Many recent studies have showed that the commercial salt from marine origin was contaminated by microplastics [*i.e*., 3 - 9]. Renzi *et al*. [3] reported that levels of MiP in different table sea salts from Italy were 300 items/kg and the annual amount ingested by humans from marine salt consumption ranges between 131 - 580 items/year. A research by Iniguez *et al*. [5] showed that the MiP content found in Spanish table salt of marine origin was 80 - 280 MPs/kg, being polyethylene-terephthalate (PET) - the most frequently found polymer, followed by PP and PE. A study by Lee *et al*. [9] reported that 94 % of salt products tested worldwide contained microplastics, with 3 out of 27 polymer types (PET, PP and PE) accounting for the majority of all particles. Considering a dose of 5 g of salt per day (according to WHO, [10]), human therefore annually ingest several hundreds of microplastic particles from salt alone.

Salt is one of the indispensable spices in culinary culture of Vietnamese people. According to statistics of the National Institute of Nutrition in 2018 [11], almost 60 % of Vietnamese people aged 26-64 are absorbing salt twice as much as WHO recommendations ([10], *e.g.* 10 g/day). High salt consumption poses not only a risk to human health (*e.g.* hypertension) but also other potential risks due to the sea salt contamination by microplastics. The previous studies by Lusher *et al.* [12, 13], and Smith *et al*. [14], pointed out that during digestion, less than 10 % of ingested MiP are absorbed into the human bloodstream, and MiP has the potential to bioaccumulate in secondary organs, with possible impacts on the immune system and cell health. However, in Viet Nam, there has not been any published research on microplastic contamination in sea salt.

This paper presents the results of abundance, size, color and nature of MiP in sea salt samples (raw and iodate fine table sea salt) collected from 9 salt production regions along Viet Nam. The aim of this study is to determine levels of contamination and nature of the microplastic in some trademarks of Vietnamese sea salt.

2. MATERIALS AND METHODS

2.1. Sea salt samples

In this study, nice commercial trademarks of iodate fine table sea salt of different origins (Thai Binh, Thanh Hoa, Quang Binh, Quang Ngai, Binh Dinh, Ninh Thuan, Ba Ria – Vung Tau, Ben Tre and Bac Lieu provinces) were collected from supermarkets. In addition, four samples of raw sea salt were selected (from Thai Binh, Thanh Hoa, Ba Ria – Vung Tau and Bac Lieu). Each sample of sea salt is pre-packed (~500 g) and randomly selected on shelfs. All commercial envelopes of salt were made by plastics. However, it should be noted from previous studies that plastic bag does not influence the abundance of microplastics in the salt (*e.g.* [3 - 5]). The commercial names of these products cannot be made public for privacy reasons.

2.2. Laboratory analyses

Sample treatment was based on the protocol of Renzi and Blaskovic, 2018; Kim *et al*., 2018; Iniguez *et al*., 2017; Yang *et al*., 2015 [3 - 5, 7]:

- Step 1 - Preparation of filtered tap water: Tap water was filtered through a 1.6 micrometer pore size filter paper (Whatman GF/A glass microfiber filter papers) and stored in a glass bottle to use for all laboratory procedures.

- Step 2 - Sample preparation and digestion: The salt in each sample package is mixed up by a metal spoon. 100 g of sea salt was placed into a 1.5 L pre-cleaned glass jar and then covered with pre-cleaned aluminum foil. The sample was digested by 100 ml of 17.25 % H_2O_2 solution for 24 hours at 50 °C in order to remove all organic materials.

- Step 3 - Dissolution and filtration: Thereafter, the salt sample was completely dissolved by adding 800 ml of filtered tap water (from step 1), and stirred well with a glass rod until the salt is completely dissolved. The salty solution was then filtered with GF/A glass microfiber filter paper. The filter paper was placed into a clean petri dish with a cover and was dried at 40 °C for 12 hours prior to further microscopic and spectroscopic analysis.

Each trademark of sea salt was analyzed in replicates (n = 3).

- To assure the control quality, specific control samples were established: (i) sieving atmospheric control (SAC, n = 3) consisting of a filter placed on the benchmark and exposed to airborne contamination during digestion and filtration, (ii) observation atmospheric control (OAC, n = 3) consisting of a filter placed on the benchmark and exposed to airborne contamination during stereomicroscope observation. The results obtained showed that no microplastic was found on the filter papers for both SAC and OAC.

- Microplastic analysis

The filters were observed using a Leica S9i Stereo Microscope (range of magnification 0.61$^{\times}$ - 5.5$^{\times}$). The potential microplastic particles in the filter paper were counted and measured for physical characteristics (such as shape, size and color) using the measuring tool of the image analysis software LAS X (Leica Application Suite X). Colors were identified in 7 classifications: white, yellow, red, green, grey, blue and black. Shapes were identified in three categories; fragment (irregular shape with an uneven surface), fiber (thin, straight, and often cylindrical shape), and pellets (rounded shape). Fibers were defined as elongated line being equally thick, not tapered towards the ends, having a three-dimensional bending; and fragments were defined as irregular shaped hard particles having appearance of being broken down from a larger piece of litter, or flat flexible particle with smooth or angular edges. All must have an absence of visible cellular or organic structures and being homogeneously colored [3 - 5]. Straight and transparent fibers were being ignored in order to exclude biological or organic origin. Size of MPs was ranged between 50 - 5,000 μm for fiber, and between 250 - 25,000,000 μm^2 for fragment [3 - 5]. All particles were photographed.

The nature of microplastics was determined on small subsamples from each site using a FTIR-ATR iS50 Thermo Fisher Scientific® at Center for Analysis Service of Experiment in

HoChiMinh city (CASE). The polymers matching more than 70 % of reference spectra were accepted as suggested in previous studies (*e.g.* [9, 15, 16]). It's important to note that only the largest fibers and fragments were analyzed by FTIR-ATR. The smallest size was very challenging to retrieve, and consequently, the determination of the nature of microplastics was not representative of the whole sample statistically.

- Data processing

In this study, one-way analysis of variance (ANOVA) was used to test for significant statistical differences between the abundance of microplastics of each salt type. Homogeneity and normality tests were applied to the data to validate the tests. All analyses were performed with a significance level of 0.05.

3. RESULTS AND DISCUSSION

The results of microplastic abundance (in items/kg) in raw and iodate fine table sea salt samples are summarized in Table 1.

Table 1. Microplastic abundance (in items/kg) in raw and iodate fine table sea salt samples. Data were presented as mean ± standard deviation (SD). The values with different superscript letters (a-d) are statistically significant difference ($\alpha = 0.05$).

	Raw sea salt								
Sample	R1	R2	R3	R4					
MiP concentration (items/kg)	(840±112)^{b}	(863±169)^{b}	(1057±174)^{c}	(723±196)^{a}					
	Iodate fine table sea salt								
Sample	F1	F2	F3	F4	F5	F6	F7	F8	F9
MiP concentration (items/kg)	(327±148)^{d}	(337±122)^{d}	(253±164)^{d}	(383±97)^{d}	(377±173)^{d}	(389±82)^{d}	(385±195)^{d}	(302±183)^{d}	(312±74)^{d}

Note: R-Raw sea salt (n = 4 samples with 3 replicates), F-Fine sea salt (n = 9 samples with 3 replicates).

3.1. Abundance, size, and color of MiP in raw sea salt

The abundance of MiP in raw salt samples collected at different provinces in Viet Nam varied between 723 ± 196 items/kg and 1057 ± 174 items/kg (Table 1). In addition, the results of Anova - Single Factor analysis showed that significant difference the abundance of microplastics in raw sea salt samples ($\rho = 0.05$), showing the strong spatial variability of MiP content in raw sea salt samples. The mean abundance of MiP in Vietnamese raw salt was 878 ± 101 items/kg.

In a total of 594 MiP particles observed on the filter paper of four raw sea salt samples, fibers were the predominant type of microplastics, corresponding to roughly 83 % of the total MiP particles (Figure 2). Fragments accounted for 17 % of the total MiP.

MiP occurred in raw sea salt samples with a variety of colors, including red, blue, grey, white, black, yellow and green (Figure 3). The most common colors of MiP particles found in raw sea salt were blue (29 %), black and grey colors accounted for about 23 and 24 %, respectively. The other colors were only found at a small ratio (red - 5 %, white - 9 %, and yellow – 10 %). The color distribution order was quite similar to all raw sea salt samples.

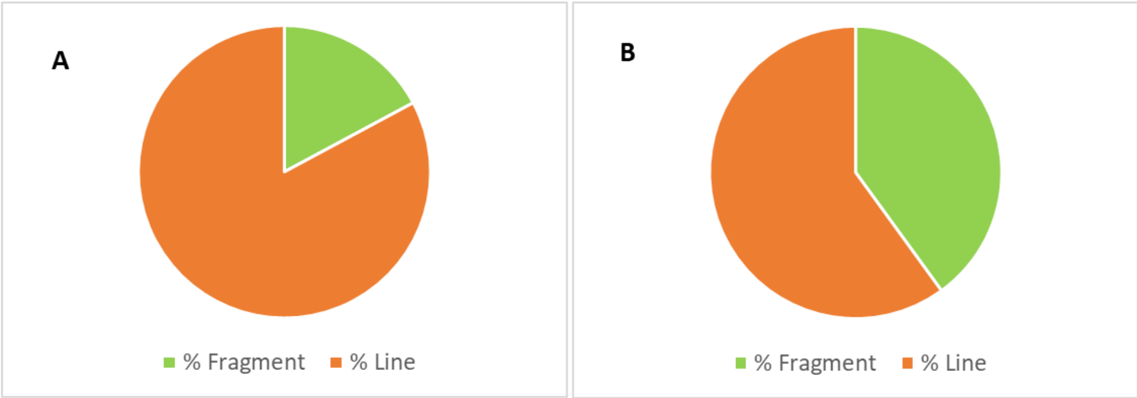

Figure 2. Contribution of the different shapes of microplastic particles in sea salt samples. (A: Raw sea salt, B: Iodate fine table sea salt).

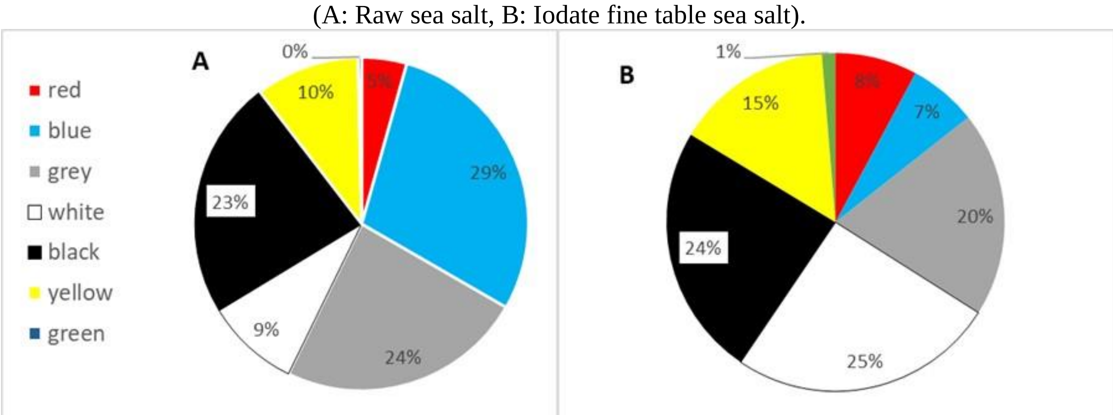

Figure 3. Contribution of the different colors of microplastic particles in sea salt samples. (A: Raw sea salt, B: Iodate fine table sea salt).

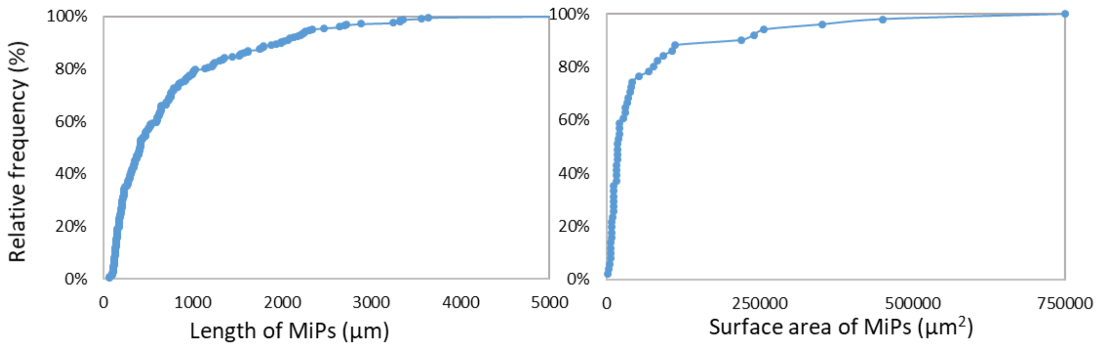

Figure 4. Relative frequency of MiP length and surface area by size class cumulated for raw sea salt samples.

Microplastics in raw sea salt of Viet Nam had a wide range of sizes with an average length of 733 ± 102 μm (min-max: 63 μm - 5000 μm) and an average surface area of 69110 ± 37504 μm^2 (min-max: 1557 μm - 750000 μm). The mean length and surface area of MiP in four raw sea salt samples were quite similar to the most frequent size of fibers, *i.e.* in the range of 100 - 900 μm (accounted for 78 % of total microplastics observed). The predominant MiP's surface area was from 1000 to 70000 μm^2 (76 %). Moreover, the surface area group of 1500 - 20000 μm^2 was the most common size in Vietnamese raw sea salt, which accounted for 30 % of the total MiP (Fig. 4).

Table 2. Type and abundance of MiP in commercial salt in the world.

Origin	Type of salt	MiP (items/kg)	Type of MiPs	Sources
France	Fine Sea salt	0-2	PE, PET, PP	[8]
Italy	Fine Sea salt	22-294	PE, PP	[3]
	Raw Sea salt	50-400	PET, PE, PP	[4]
Spain	Fine Sea salt	80-280	PE, PET, PP	[5]
Portugal	Fine Sea salt	0-10	PET, PP	[8]
UK	Fine Sea salt	120	PP, PE, PVC	[6]
USA	Fine Sea salt	300	PE	[7]
India	Fine Sea salt	56-103	PA, PE, PP, PU, PVC	[13]
	Raw Sea salt	800-1100	PET, PE, PP	[4]
Malaysia	Fine Sea salt	0-1	PP	[8]
China	Fine Sea salt	120-718	PE, PET, PV, PU, PP	[7]
	Raw Sea salt	1000-2000	PE, PET, PV, PU, PP	
Chinese Taipei	Fine Sea salt	0-1300	Acrylic, Nylon, PE, PET, PP, PVC, PW	[4]
Indonesia	Fine Sea salt	100	PE, PET, PP	[4]
Japan	Fine Sea salt	0-1	PE, PET	[8]
Korea	Fine Sea salt	100-300	Acrylic, Nylon, PE, PET, PP, PVC	[4]
	Raw Sea salt	100-500	Acrylic, Nylon, PE, PET, PP, PVC	
Thailand	Fine Sea salt	80-600	PE, PET, PP, PVC	[4]
	Raw Sea salt	500-1000	PE, PET, PP, PVC	
Vietnam	Fine Sea salt	189-469	PE, PP, PS	This study
	Raw Sea salt	620-1200	PE, PP, PS	
Germany	Rock salt	2	PET	[5
Italy	Rock salt	80	PE, PET, PP, PVC	[3]
USA	Rock salt	5	PE	[4]
China	Rock salt	0-14	PET, PP, Teflon	[4]
Philippines	Rock salt	120	PE, PET, PP, PVC	[4]
China	Lake salt	28	PE, PET, PP, PS	[7]
USA	Lake salt	113	-	[12]
Turkey	Lake salt	8-102	PE, PET, PP, PU, PA, PVC	[13]
Malaysia	Lake salt	0	-	[8]
Iran	Lake salt	1	PP	[8]

Note: PA - polyamide, PE - polyethylene, PET - polyethylene terephthalate, PP - polypropylene, PVC - polyvinylchloride, PS - polystyrene, PU - polyurethane, PW - paraffin wax.

Despite the limited number of raw sea salt samples collected and analyzed in this study (4 samples), the comparison with other salt studies from other countries allows preliminary assessment of the microplastic contamination level in sea salt of Viet Nam. The abundance of MiP in raw sea salt samples collected from Viet Nam is similar to that observed in India or China, but higher than that observed in Italy or Korea. However, these values are relatively lower than those measured in China (Table 2). Previously published literature on the abundance of MiPs in unrefined sea salt (*i.e*. raw salt) from worldwide rivers showed a significant correlation between MiP abundance and plastic emissions/MiP pollution levels in surrounding seawaters [4, 5, 16, 17]. Research of Kim *et al*. on 25 samples of sea salt worldwide highlighted that the origin of MiP in sea salt from both plastic inputs from rivers and pollution levels in seawaters near the production area of table salt [4]. More recently, a research by Strady *et al*. evidenced that the abundance of MiP measured in Chinese lakes, rivers, and bays was mostly higher than those measured in Viet Nam [2]. Therefore, raw sea salt can be a good indicator of the magnitude of MiP pollution in the surrounding marine environment [*e.g*. 15, 17, 18, 19].

3.2. Abundance, size, and color of MiP in iodate fine table sea salt

The results in Table 1 showed that MiP abundance measured in iodate fine sea salt samples was lower than that in raw sea salt samples. In fact, MiP abundance in 9 samples of fine table sea salt collected along Viet Nam varied from 189 to 469 items/kg (Table 1). However, the results of Anova - Single Factor analysis showed that no significant difference in the abundances of microplastics in fine sea salt samples was observed ($\rho = 0.05$), showing that the MiP abundance among iodate fine table sea salt samples are similar for all regions in Viet Nam. The mean abundance of MiP in fine table sea salt of Viet Nam was 340 ± 26 items/kg.

Concerning the shape of microplastics in iodate fine sea salt, fibers and fragments accounted for 60 and 40 % of the total MPs, respectively (Figure 2). In terms of MiP's color, the color distribution order was slightly different from raw sea salt (Figure 3). In fact, white was the predominant color of microplastics (25 %), black and grey colors accounted for about 24 % and 20 %, respectively. The other colors were only found at a small ratio (green – 1 %, blue – 7 %, red – 8 % and yellow – 15 %; Figure 3).

The microplastic particle size observed in iodate fine sea salt samples was smaller than that in raw sea salt samples with both fragment and fiber shapes (Figure 4, 5). The length of microplastics in fine sea salt samples ranged from 61 to 2196 µm with a mean length of 563 ± 103 µm. In terms of fragment, the surface area of MiP varied from 1034 to 200000 µm^2 with an average of 25332 ± 9403 µm^2. The most frequent size of MiP was in the range of 100 - 700 µm (~ 76 %), and 2000 - 30000 µm^2 (~ 74 %), for fiber and fragment, respectively (Figure 5).

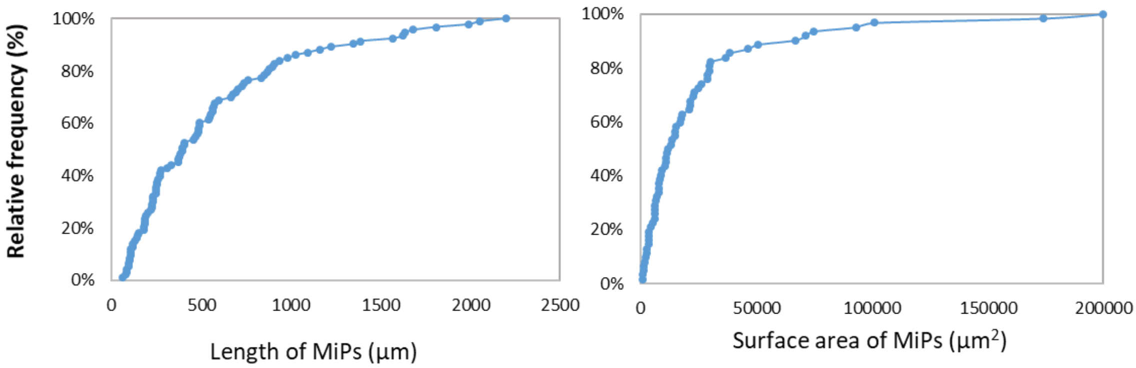

Figure 5. Relative frequency of MiP length and surface area by size class cumulated for iodate fine sea salt samples.

Table 2 showed that a similar range of microplastic abundances (100 - 700 items/kg) in fine sea salt to this study was observed in China, Korea, Thailand and the USA. These values are relatively higher than those measured in Italy, Spain, UK, Indonesia and India (ranging between 20 - 300 items/kg). The MiPs abundances measured in other countries (*e.g*. France, Portugal, Malaysia and Japan) were one order of magnitude lower (<10 items/kg) than that in Vietnamese sea salt. In addition, the data in Table 2 also showed that sea salt is more contaminated by microplastics as compared to lake and rock salts. This result is not surprising because the source of microplastic contamination in salt has been proved to be from environmental pollution (*e.g.* seawater).

3.3. Nature of microplastics in sea salt

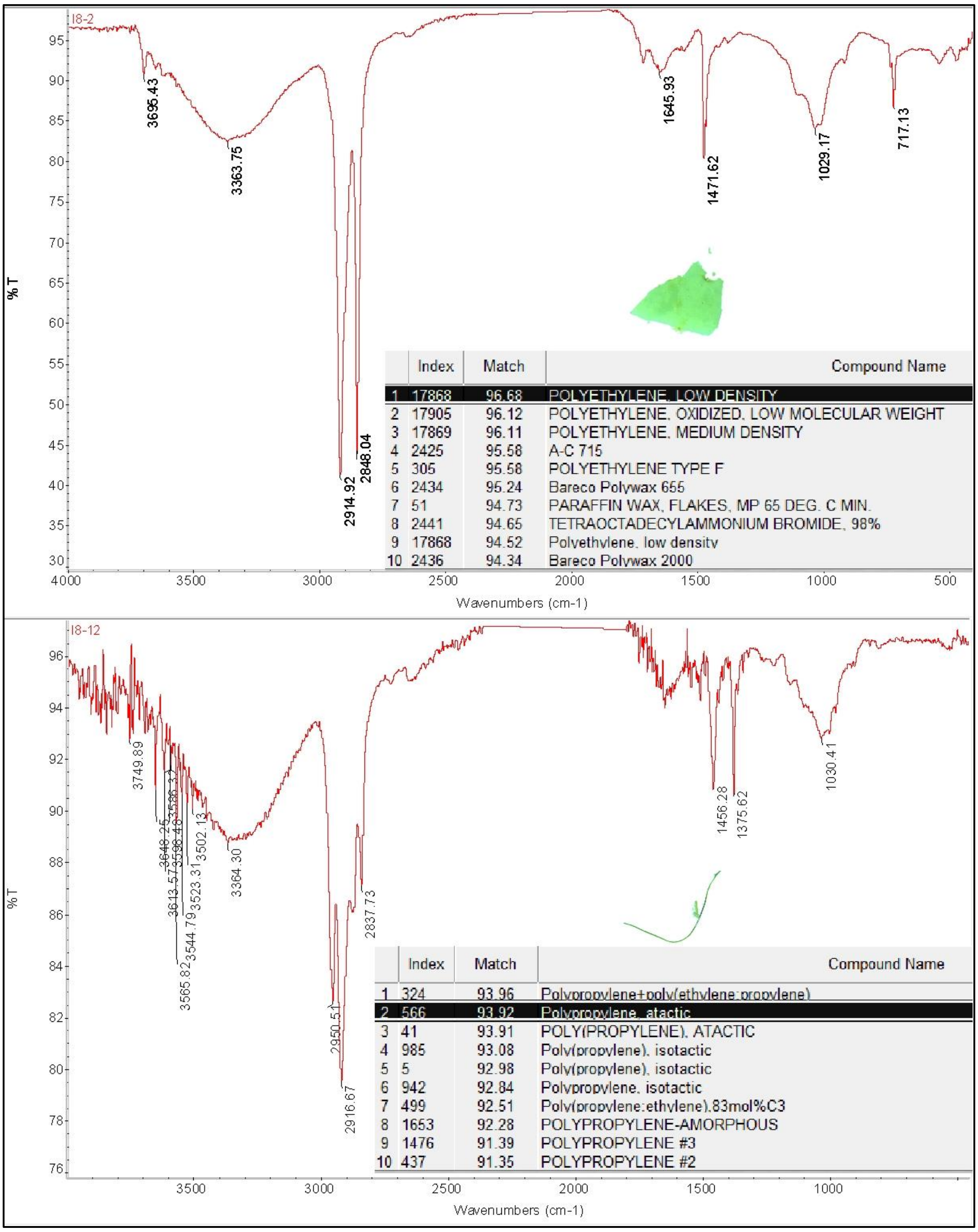

Figure 6. Examples of MiP's FTIR spectra found in sea salt samples.

Fourier Transform Infrared Spectroscopy (FTIR) is one of the most popular methods used to confirm the nature of microplastics [*e.g.* 15 - 18]. In this study, we have chosen 12 microplastic particles (8 fragments within 1000 - 5000 µm of size and 4 fibers within 750 000 - 1 000 000 μm^2) in 6.5 kg of salt from 13 salt products (4 samples of raw salt and 9 samples of iodate fine salt) for FTIR analysis. Based on FTIR results, we detected three different polymer types, including polyethylene (PE), polypropylene (PP) and polystyrene (PS) (Figure 6), among which, the most common is PE (accounting for 67 % of the total analyzed MiP). Although the determination of the nature of microplastics was not statistically representative for the whole sample (12/1986 particles), it is clear that PE and PP plastics detected in the sea salt of Viet Nam are also the two most abundant types of microplastics observed in commercial salt in the world (Table 2). This result completely coincides with the previous studies (*e.g.* [2, 20 - 23]), which showed that the most abundant microplastics observed in the marine environment were PE and PP.

3.4. Ingestion of microplastics via salt consumption

According to the WHO recommendations, the amount of salt intake in the adult's diet should not exceed 5 g/day, microplastics ingested per day per adult is 1.74 items/day (*e. g.* 635 items/year - calculated according to the results of MiP in Vietnamese fine sea salt). However, Vietnamese people consume salt twice as much as WHO recommendations (up to 10 g/day, [10]), so the annual number of MiP particles ingested per Vietnamese adult varies from 637 to 1270 items/year (using Vietnamese iodate fine sea salt). The occurrence of MiP in food chain in general and in table salt in particularly, not only decreases their quality but also will be possible adverse effects on human health, potential high socio-economic cost, and bring serious economic consequences for sea saltworks [24 - 27].

4. CONCLUSIONS

This research presents the first results on microplastic contamination in commercial sea salt of Viet Nam. The result obtained showed that the abundance of MiP in raw sea salt was higher than that in fine sea salt with an average value of 878 ± 101 items/kg for raw sea salt and 340 ± 26 items/kg for fine sea salt. These results are comparable to the values measured in sea salt from China, Korea, Thailand and the USA. Moreover, we have determined that fibers were the predominant type of microplastics in both raw and fine sea salts with an average length of 733 ± 102 μm and 563 ± 103 μm in raw and fine sea salts, respectively. In addition, MiP occurred in all sea salt samples with variety of colors, including red, blue, grey, white, black, yellow and green; and the most frequently found colors of MiP particles were blue, black, grey and white. The result of FTIR showed that the most common of MiP in sea salt is PE. Finally, we estimated that the annual number of MiP particles ingested per Vietnamese adult varies from 635 to 1270 particles with a mean daily salt consumption of 5 - 10 g/day.

It is clear that the presence of MiP in food chain in general and commercial salt in particular, will be a threat to human health, as well as have a negatively impact on the salt industry, the socio-economy of populations that subsist with their production. Human food safety and health need to be a priority not only for the scientific community but also for the government, aiming at developing appropriate measures and policies to reduce plastic pollution and risks associated with plastic pollution.

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Declaration of competing interest. The author declares that she has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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