

Impact of Ultra Violet Radiation on Polyethylene Packaged Water Exposed at Varying Conditions: Are we Drinking Micro-Plastics?

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Abstract: The public is worried about how their health is impacted by microplastics in sachet water. This awareness has repercussion for water producing factories and public health organization in managing health hazard associated with microplastic ingestion. Many countries in Africa, with concern to Nigeria, the act of packaging water in polyethylene materials by water producing factories has become accustomed. These packaged products are popularly called pure water or sachets water. The packaged materials are made of antioxidants, stabilizers, plasticizers, lubricants, antimicrobials, anti-static and anti-blocking agents. Heat hindrance agents are usually incorporated to improve functionality of polyethylene. It was conceived that the polyethylene material if exposed to sun light radiation over a period of time, its components may leach into the potable water due to photolytic, photo-oxidative and thermo-oxidative reactions caused by fragmentation of the polyethylene material. This research was designed to evaluate the water quality/safety of polyethylene sachet-packaged exposed to direct sunlight at different days. Eighteen polyethylene packaged water were sampled from three different factories - Lion water, Galaxy and Ashor water in Nsukka area of Enugu State. They were exposed to sunlight at 31 to 33°C for (24, 48, 72, 96 and 120 h), respectively. Agilent gas chromatography tandem mass spectroscopy was used to analyze the exposed and unexposed (control) packaged water. Organic compounds such as low molecular weight substituted hydrocarbons, polycyclic aromatic hydrocarbons, trichloromethane, benzene, limonene, xylene, toluene and 2-hexanone were detected in all the exposed samples. These micoplastics which leached into water has been listed by the Agency for Toxic Substances and Disease Registry as potential human carcinogens. The concern now is whether we are drinking micoplastics. The only way out of this perceived health risk is to change the packaging material to prevent ingestion of microplastics in water.

Keywords: Microplastics, Polyethylene, Sunlight, Radiation, Water Quality, Exposure

Introduction

The threat to public health associated with ingestion of water exposed to ultra violet radiation at varying interval is an issue for research and discussion. Human life is surrounded by a wide range of Volatile Organic Compounds (VOCs), including o-xylene causing harmful health effects (Kamal *et al.*, 2015). There have been concerns about the presence of microplastics,

which are defined as plastic particles with a diameter less than 5 mm, without lower limit. They are derived from progressive fragmentation of larger debris, or are deliberately made for use in personal care products, medicines and industry (Sandrine *et al.*, 2017). In addition to physical impacts of the plastic particles microplastics are associated with complex mixture of chemicals that may be transfer to humans upon exposure. These chemicals include additives, residual

monomers and sorbed ambient chemical substances, which are endocrine-disrupting and can adversely affect human health if ingested (Kole *et al.*, 2017).

Polythene products are identified as low (very low, linear low, medium, linear medium) and high density resins. They are used in the production of boil-in food packages, heat-sealed films, heat-sealed pouches, production of grocery bags, squeezable bottles, cable insulators and production of flexible food packaging, shrink-wrap, stretch film and overwrap film (NOVA, 2012). Polythene and Polyvinylchloride (PVC) is the most frequently used polymer in packaging, an important prerequisite for food packaging (Thorbjörn and Bengt, 2000). During the production of polyethylene, chemical additives are usually incorporated to improve functionality, they include: Antioxidants, stabilizers, plasticizers, lubricants, antimicrobials, anti-static and anti-blocking agents, "slips," or heat resistance agents (Kanishka *et al.*, 2013). More so, polythene used for packaging potable water are often exposed to heat, either by conventional heating or solar radiation. Heating causes the rapid degradation of polythene into different microplastics (Lin *et al.*, 2000; Carsten *et al.*, 2015). Varieties of synthetic polymers absorb ultraviolet radiation and undergo photolytic, photo-oxidative and thermo-oxidative reactions (Scott, 2000). Photo-oxidative causes the breaking down of polymer chains, radical formation and reduction of molecular weight, leading to leaching of microplastics (Bottino *et al.*, 2004; Emad and Raghad, 2013). These microplastics have health, economics and environmental implications on the final consumers. Traders (hawkers) usually carry these polythene-sachets water about for business, while some marketers abandon the packaged water exposed to sunlight radiation due to lack of space for storage. Microplastics have become prevalent in waters, soil, food webs, indoor and outdoor air. Public concerns about microplastics are mounting as a result of their unknown effects at the micromolecular level and the consequences for human health. There is an expanding knowledge base on microplastics for marine and ecosystems, but the understanding of the human health effects from exposure to microplastics reveals a knowledge gaps. Humans can be exposed to plastic particles via consumption of seafood and terrestrial food products, drinking water and via the air. However, the level of human exposure, chronic toxic effect concentrations and underlying toxicological mechanisms by which microplastics elicit effects are poorly understood to make a full assessment model of the risks to human's exposure. Since sunlight radiation is accompanied with heat, which cause the degradation/fragmentation of polyethylene material and leaching of microplastic into water, investigation was designed to interval.

Materials and Methods

Sample Collection

Eighteen sachets water samples; six clusters from three different brands of polyethylene packaged-water were obtained from factories: Lion, Galaxy and Ashor, in Nsukka, Enugu State.

Chemicals/Reagents

All chemicals were of analytical grade.

Experimental Design

Fifteen Sachet water, five each from the three brands (Lion, Galaxy, Ashor) were exposed to sun light radiation at 31 to 33°C for day 1, 2, 3, 4 and 5 or (24, 48, 72, 96 and 120 h), respectively. At the end of each day, sachet-water samples were collected with sterilized 200 mL Amber bottle, this was continued to the day 5. Three samples were used as control (unexposed). The samples were analyzed with gas chromatography tandem mass spectroscopy.

Intervals of Exposed Samples:

- Sample a-exposure on day 1 (24 h) at 31 to 33°C
- Sample b-exposure on day 2 (48 h) at 31 to 33°C
- Sample c-exposure on day 3 (72 h) at 31 to 33°C
- Sample d-exposure on day 4 (96 h) at 31to 33°C
- Sample e-exposure on day 5 (120 h) at 31 to 33°C
- Sample f-control (sachet water sample unexposed to sunlight) 2 to 15°C

Preparation of Samples for GC-MS Analysis

Water samples were transferred into glass containers with Teflon lined septa. The sample container was filled just to overflow in order to void air bubble passing through the bottle. The sample bottles were sealed with screw caps to prevent air bubbles entrapment. Gas chromatography-Mass Spectroscopy with purge and trap technique was performed using Agilent Technologies 7890 A and 5975 N mass selective detector procedure. The samples were placed in a sealed vessel and arranged in auto-sampler. The transfer line was connected directly into the GC injector by a needle. Injector mode was used in the split-split less injector. The samples were purged with inert gas concentrator, causing Volatile Organic Compounds (VOCs) to sweep out of the samples. The VOCs were retained in an analytical trap to allow purge gases to pass through the vent. The VOCs were then desorbed by heating the trap injected into the GC by back flushing the trap with carrier gas. Microplastics were identified by comparing measured retention data with structure-retention correlations, mass spectra and interpreted via matching the results with library spectra.

Condition of GC-MS

The following were the condition of the GC-MS with Purge and Trap technique:

Injection: purge and trap split (split ratio 35:1), Liner: 1mm Split (cat.# 20972-214.1)

Inj. Temp.: 200°C, Purge and Trap, instrument: OI 4660 Eclipse Purge and Trap, Trap Type: #10 (Tenax®/silica gel/carbon molecular sieve), Purge: 11 min, flow 40 mL/min, Desorb Preheat Temp.: 185°C, Desorb: 0.5 min @ 190°C, flow 35 mL/min, Bake: 8 min @ 210°C, Interface Connection: injection port, Transfer Line Temp.: 150°C, Oven Temp.: 40°C (hold 4 min) to 90°C at 16°C/min to 220°C at 32°C/min (hold 5 min), Carrier Gas: He, constant flow, Linear velocity: 34 cm/sec @ 40°C, Detector: MS, Mode: Scan, Transfer Line Temp.: 150°C, Ionization Mode: EI, Scan Range: 35-300 amu, Column: Rtx®-VMS, 30 m, 0.25 mm ID, 1.40 µm (cat.# 19915), 502.2 Calibration Mix #1 (gases), 2000 µg/mL in methanol (cat.# 30042), Diluents: 25 mL water.

Results

The chromatograms and Tables bear's the concentration and percentage composition for all compounds detected, while chromatograph can be found at the appendicies. The analysis was ran under the same conditions to compare results of varied exposure days.

Discussion

The polyethylene packaged water exposed to sunlight radiation at varying temperature and duration, followed similar trend. The compounds with higher molecular weight gave the highest concentration exemplified in the Tables and Chromatograms. For instance, Cyclotetradecane; 9-Octadecene, (E)-; 2-Tetradecene, (E)- had 12.532 ppm, followed by 8.156 ppm - Cyclotetrasiloxane, octamethyl- and lastly-ethylene oxide 1.243 ppm Table 1. Low molecular weight, typically monomers and residual solvents (e.g., ethylene oxide, methylene chloride, acetic

acid and methane) were quick to elude. According to COWI (2013), monomers such as formaldehyde, vinyl chloride, ethylene oxide and butadiene are gases with tendency to migrate rapidly at ambient temperatures. In addition, solubility also determine the rate of migration of substances i.e., the higher the solubility of the plastic polymer, the less the rate of migration of such substance (COWI, 2013). As temperature increases, the leaching of monomers, oligomers and other compounds increases (Kanishka *et al.*, 2013). As the duration of exposure increases, the internal and/or external chromophoric groups in the packaging material absorb sunlight radiation and produce low molecular weight radicals (R) or polymeric macro radicals (P) through photo-oxidation (Cheshire *et al.*, 2009). Octamethyl, (E)-9-Octadecene and (E)-2-Tetradecene radicals were produced after day 1 exposure to sunlight, Table 1 and 2 (the onset of fragmentation and degradation). This reaction is also referred to as Norrish initiation or photo-degradation reaction (Yousif, 2012). The radicals produced in photo-initiation step proceeds to propagation step, where they undergo reaction with polythene polymer in a chain process to produce low molecular radicals, hydroxyl and polymer alkyl/peroxy/alkyloxy radicals (Soják *et al.*, 2006). This give rise to the production of more radicals on day 3 to 5 exposed samples Table 3 to 5.

Zhang and Song (2005) reported that VOCs containing xylene and formaldehyde may also impair the memory and hearing ability in mice. Also, female workers' in the jewel processing factories are prone to DNA damage in the peripheral blood cells exposed to xylene, benzene and toluene simultaneously (Wei *et al.*, 2009). According to Huang *et al.* (2004), fatigue, conjunctivitis, memory deterioration and hand degreasing is common with individual exposed to microplastics. Low level exposure to mixed - xylene containing benzene and toluene may lead coryza, catamenai disruption, dermatitis and pharyngitis. These mixtures are harmful to reproductive system, nerve system mucous membrane and cuffs (Wang *et al.*, 2004). Carbon dioxide - 1.516 ppm and 78.66% composition can reason injury to human health.

Table 1: Compounds detected on day 1 exposure to sunlight radiation

Peak	Concentration ppm	% Composition	Compounds Identified
1	1.243	68.66	Ethylene oxide; Ethyne, fluoro-
2	1.523	16.82	Methylene Chloride
3	6.998*	5.21	Acetic acid
4	8.156*	5.63	Cyclotetrasiloxane, octamethyl-
5	12.532*	3.68	Cyclotetradecane; 9-Octadecene, (E)-; 2-Tetradecene, (E)-

The microplastics marked with asterisks (*) have concentration above permissible limit
 Chromatogram is found in appendix 1

Table 2: Compounds detected on day 2 exposure to sunlight radiation

Peak	Concentration ppm	% Composition	Compounds identified
1	1.516	78.66	Carbon dioxide; Ethylene oxide
2	1.873	13.59	Methylene Chloride
3	6.419*	7.75	Acetic acid

The microplastics marked with asterisks (*) have concentration above permissible limit
 Chromatogram is found in appendix 2

Table 3: Compounds detected on day 3 exposure to sunlight radiation

Peak	Concentration ppm	% Composition	Compounds identified
1	1.506	22.23	Carbon dioxide; Ethylene oxide
2	1.629	6.74	Formic acid hydrazide; Hydrazine, 1,1-dimethyl-
3	1.818	6.04	Methane, dimethoxy-
4	2.288	1.41	Trichloromethane; Methane, bromodichloro-; Trichloromethane
5	4.548	19.70	Toluene
6	6.265*	0.96	Acetic acid
7	6.648*	2.56	Benzene, 1,3-dimethyl-; o-Xylene
8	7.062*	0.70	o-Xylene; Benzene, 1,3-dimethyl-
9	7.734*	1.21	1R-.alpha.-Pinene; Bicyclo[3.1.1]hept-2-ene, 2,6,6-tetramethyl-, (+/-)-; .alpha.-Pinene
10	8.162*	0.89	Benzene, 1-ethyl-2-methyl-
11	8.561*	3.39	1-Decene; Cyclopropane, nonyl-
12	8.615*	2.54	Benzene, 1,2,3-trimethyl-, 1,2,4-trimethyl-
13	8.844*	3.11	3-Carene
14	9.033*	1.02	Benzene, 1-methyl-4-(1-methylethyl)-, 1-methyl-2-(1-methylethyl)-
15	9.088*	1.10	Limonene; D-Limonene
16	10.924*	14.30	1-Dodecene; 2-Dodecene, (Z)-; 1-Undecanol
17	11.004*	0.76	Dodecane; Hexadecane; Undecane
18	11.062*	1.73	6-Dodecene, (Z)-; 5-Dodecene, (Z)-; 6-Dodecene, (E)-
19	11.162*	1.28	2-Dodecene, (Z)-; 3-Dodecene, (Z)-; 2-Dodecene, (E)-
20	12.078*	0.71	Naphthalene, 2-methyl-, 1-methyl-
21	12.843*	7.62	Cyclopropane, nonyl-; 1-Tetradecene; 2-Tetradecene, (E)-

The microplastics marked with asterisks (*) have concentration above permissible limit
 Chromatogram is found in appendix 3

Table 4: Compounds detected on day 4 exposure to sunlight radiation

Peak	Concentration ppm	% Composition	Compounds identified
1	1.493	22.87	Carbon dioxide; Ethylene oxide
2	1.616	7.77	Formic acid hydrazide; Ethyne, chloro-; Thiirane
3	1.792	6.32	Methane, dimethoxy-
4	2.259	1.15	Trichloromethane; Ethane, 1,2,2-trichloro-, 1,1-difluoro-
5	4.503	5	Toluene; 1,3,5-Cycloheptatriene
6	6.477*	0.7	Ethylbenzene
7	6.625*	3.21	o-Xylene; p-Xylene
8	7.043*	0.83	Benzene, 1,3-dimethyl-; o-Xylene; p-Xylene
9	7.725*	1.36	alpha.-Pinene; 1R-.alpha.-Pinene; Bicyclo[3.1.1]hept-2-ene
10	8.152*	1.01	Benzene, 1-ethyl-4-methyl-, 1-ethyl-3-methyl-, 1-ethyl-2-
11	8.557*	4.12	1-Decene
12	8.612*	3.22	Benzene, 1,2,3-trimethyl-, 1,2,4-trimethyl-
13	8.75*	0.69	cis-3-Carene; 4-Decene; trans-4-Decene
14	8.84*	3.9	3-Carene; 1S-.alpha.-Pinene
15	9.03*	1.19	Benzene, 1-methyl-4-(1-methylethyl)-, 1-methyl-3-(1-methylethyl)-, 1-methyl-2-(1-methylethyl)-
16	9.085*	1.29	Limonene; D-Limonene
17	9.918*	0.83	Undecane
18	10.927*	19.32	1-Dodecene; 5-Dodecene, (E)-; 2-Dodecene, (Z)-
19	11.008*	0.97	Dodecane; -octadecanesulphoyl chloride
20	11.062*	2.22	5-Dodecene, (Z)-; 3-Dodecene, (E)-; 6-Dodecene, (Z)-
21	11.162*	1.64	2-Dodecene, (Z)-; 2-Dodecene, (E)-
22	12.843*	9.66	1-Tetradecene; 2-Tetradecene, (E)-; 1-Dodecene
23	12.956*	0.73	5-Tetradecene, (E)-; 3-Tetradecene, (E)-

The microplastics marked with asterisks (*) have concentration above permissible limit
 Chromatogram is found in appendix 4

Table 5: Compounds detected on day 5 exposure to sunlight radiation

Peak	Concentration ppm	% Composition	Compounds identified
1	1.510	29.23	Carbon dioxide; Ethylene oxide
2	1.623	6.43	Formic acid hydrazide; Hydrazine, 1,1-dimethyl-
3	1.822	9.73	Methane, dimethoxy-
4	2.291	2.23	Trichloromethane
5	4.539	19.80	Toluene
6	6.632*	2.32	Benzene, 1,3-dimethyl-; p-Xylene
7	6.757*	0.90	Acetic acid, anhydride with formic acid
8	8.554*	2.24	Cyclodecane; 1-Decene; Cyclopropane, 1-methyl-2-octyl-
9	8.609*	2.22	Benzene, 1,2,3-trimethyl-, 1,2,4-trimethyl-, 1,3,5-trimethyl-
10	8.841*	2.04	1R-.alpha.-Pinene; 3-carene
11	10.921*	12.97	1-Dodecene; 2-Dodecene, (Z)-; 1-Undecanol
12	11.062*	1.41	3-Dodecene, (E)-; 5-Dodecene, (E)-
13	11.162*	0.99	2-Dodecene, (Z)-; Cyclopropane, 1-butyl-2-pentyl-, cis-; 6-Dodecene, (Z)-
14	12.844*	7.50	4-Tetradecene, (E)-; 2-Tetradecene, (E)-; 1-Tetradecene

Chromatogram is found in appendix 5

The microplastics marked with asterisks (*) have concentration above permissible limit

The control samples have no peaks (F). No microplastics in the control samples.

Furthermore, as the durations of exposure increases to day 3, 4 and 5, more microplastics progressively leached into the water appendix 3 and Table 3. Naphthalene - 12.078 ppm (0.71%) composition and tetradecene - 12.843 ppm had the highest concentration, but varies in composition and concentration with o-xylene, dodecene and limonene. In Table three and appendix three, it was observed that many different microplastics leached into the packaged water. Therefore, it was concluded that upon exposure of Sachet water, the day three leaching was most significant. Thus, day four and day 5 the microplastic level begins to decrease due to volatilization and evaporation, while day 1 and day 2 was assumed to be the onset of fragmentation/degradation.

Photo-degradation/fragmentation process causes the leaching of unreacted monomers, oligomers and additives incorporated in the plastic polymer (Carsten *et al.*, 2015). COWI (2013), states that hydrazine, methylene Chloride, formamide, dimethylformamide, limonene and D-limonene, naphthalene, 2-methylnaphthalene, ethylene oxide, cyclopropane, ethane, trichloromethane, ethylbenzene. Ethylene oxide, ethane, cyclopropane, trichloromethane are volatiles, while ethylbenzene, limonene and D-limonene are monomers. Guo *et al.* (2010) reported that hydrazine is an intermediate in production of blowing agents in thermoplastics, flame retardants for nylon and smoke suppressant additives for polyurethane foam; formamide and dimethylformamide are used as solvent and plasticizer in consumer products. Also, naphthalene, 2-methylnaphthalene and 1-methylnaphthalene are impurity in plasticisers (BFR, 2015). The leaching of these additives under different study conditions was reported by (Biedermann *et al.*, 2008). Some of these additives are harmful to humans, even at the worst nadir value (Fasano *et al.*, 2012). Many of this compounds detected are among the Priority List of hazardous substances by Agency for Toxic Substances and Disease Registry (ATSDR, 2015). These includes: trichloromethane (Chloroform), naphthalene, benzene,

xylene, toluene, 2-hexanone, methane, Methylene chloride, hydrazine, 1, 2-dichloroethane, Acetone, formaldehyde, 2-butanone, 2-Butene, Silanes, Ethylene oxide, hexanal, propanal, decane, dodecane, cycloheptatriene, cyclohexadecane, undecan. Naphthalene has been listed by the US Environmental Protection Agency as priority environmental pollutants (US EPA, 2014). Naphthalene and hydrazine have been confirmed by U.S. Environmental Protection Agency (EPA) as a group C and group 2B, possible human carcinogen (US EPA, 2016).

Clausen *et al.* (2001 - quoted from CICAD 1998) reported that 72% of *d*-limonene metabolites were excreted in male rabbit urine 72 h after oral administration, while 7% was found in the faeces. In humans, about 25-30% of an oral dose of *d*-limonene was found in urine as *d*-limonene-8,9-diol (M-II) and its glucuronide (M-VI); about 7-11% was eliminated as perillic acid (M-III) and its metabolites (Dietrich and Swenberg (1991a) - quoted from CICAD 1998). *D*-Limonene-8, 9-diol is most probably formed via *d*-limonene-8,9-epoxide (Dietrich and Swenberg, 1991b; EMEA 2002-both quoted from CICAD 1998). Cyclodecane, dodecene, methane, hydrazide and hydrazine were detected in day 5 exposed samples at different composition and concentration. Ethylene oxide is a known toxicant, which is listed among the 30 chemicals of concern due to its implication on reproductive and developmental consequences. IARC, (1994) concluded that ethylene oxide is carcinogenic to humans. US EPA has classified ethylene oxide as a Group B1, probable human carcinogen. Therefore this research has set up a lead way to avoiding the use of exposed polyethylene packaged water for consumption at temperature range of 31 to 33°C, as its future health effect may be detrimental to the public. Finally figure 1 to 5 are annexed as appendices. There are chromatograms with retention time and % area data compound identified. Each chromatogram was run under the same conditions to allow for comparison between results of different days of exposure.

They begin from appendix or figure 1, 2, 3, 4 and 5, respectively. Representing chromatogram for sample a, b, c, d and e, sequentially.

Conclusion

The presence of microplastics in water justified the knowledge gap that leaching do occur when polythene-sachet water are exposed to sunlight radiation. Indeed, the public is consuming microplastics. Based on findings some of these microplastics are among the priority list of hazardous substances as potential endocrine disruptor and repro-toxic substances.

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Authors Contributions

Okpashi Victor Eshu: Design the work, write the manuscript and supervised the work.

James Matthew Anayo: Did the field work and laboratory analysis.

Ikechukwu N.E. Onwurah: The lead research/Supervisor.

Ethics

The authors hereby declare no conflict of interest regarding the publication of this article.

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Appendices

Appendix 1

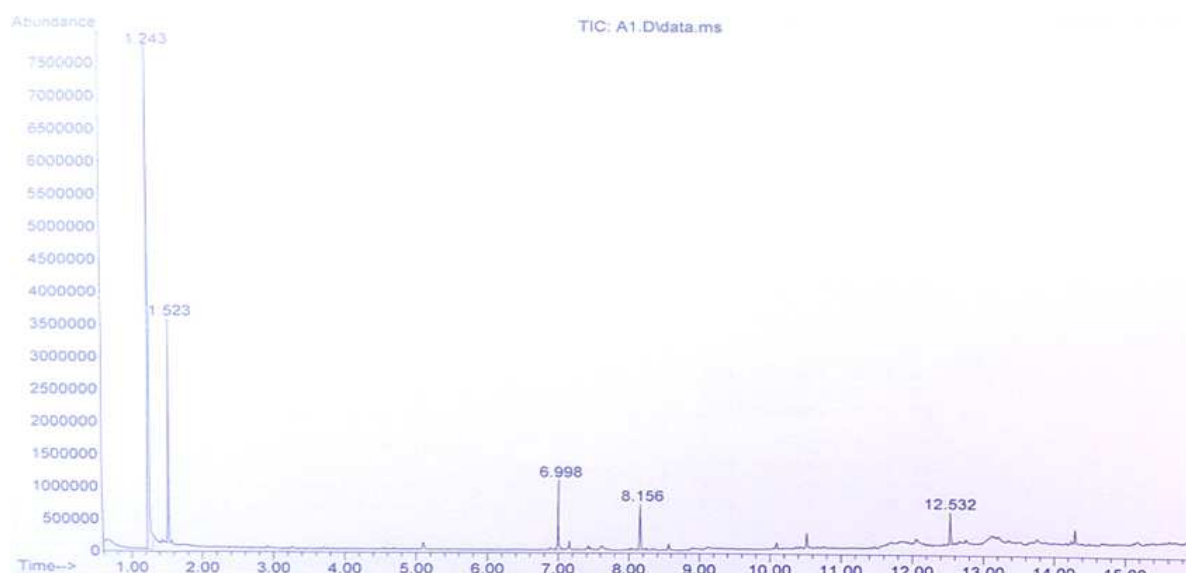


Fig. 1: Chromatogram for sample a, results is presented in Table 1

Appendix 2

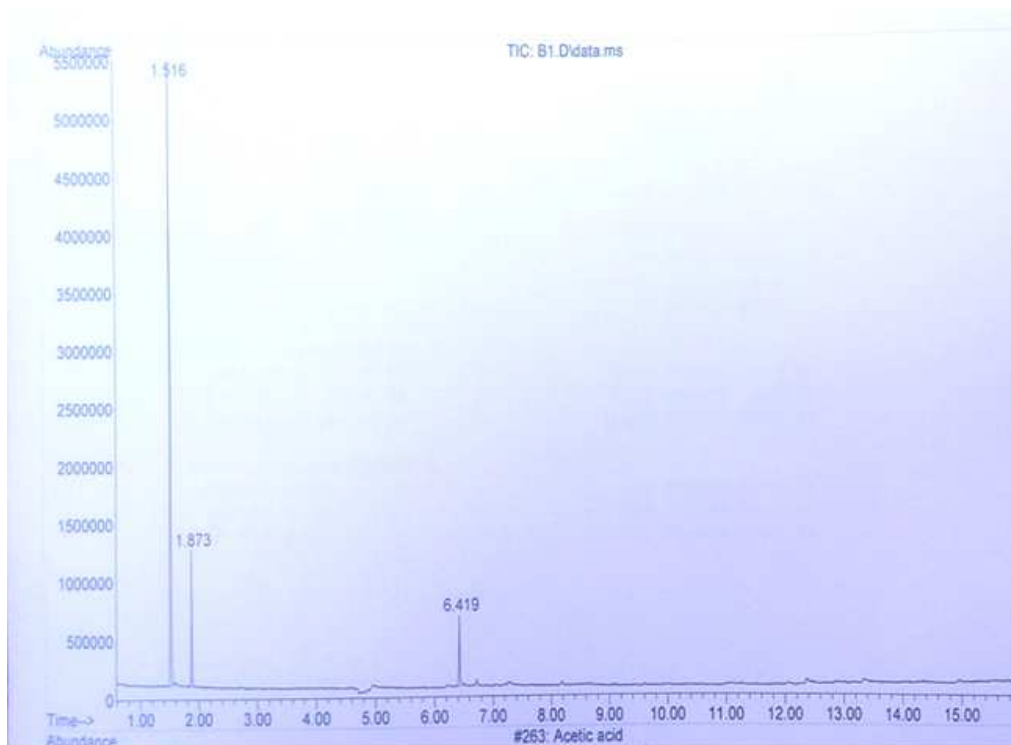


Fig. 2: Chromatogram for sample b, results is presented in Table 2

Appendix 3

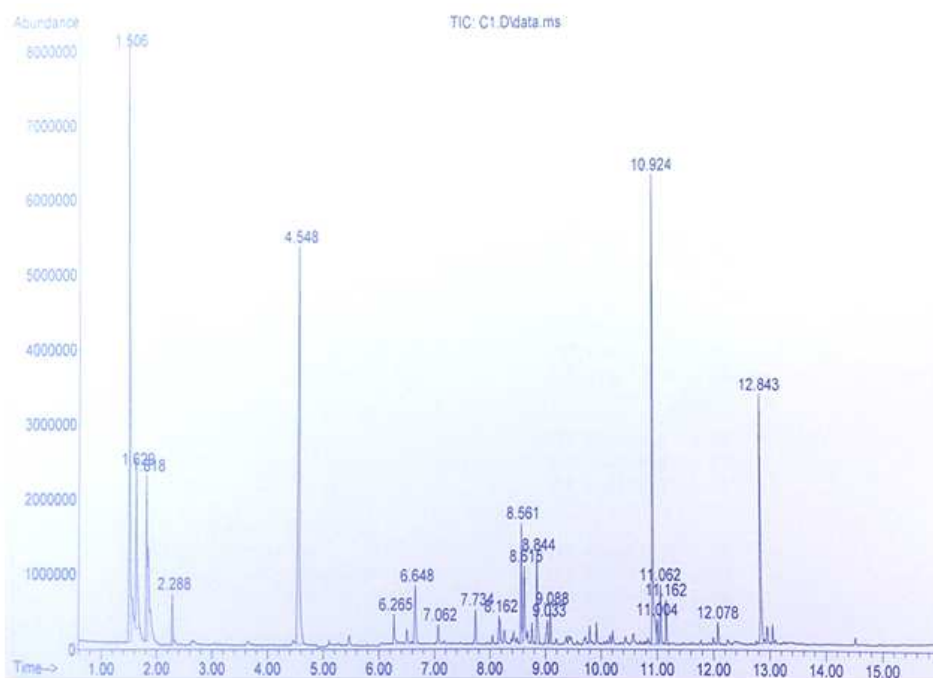


Fig. 3: Chromatogram for sample c, results is presented in Table 3

Appendix 4

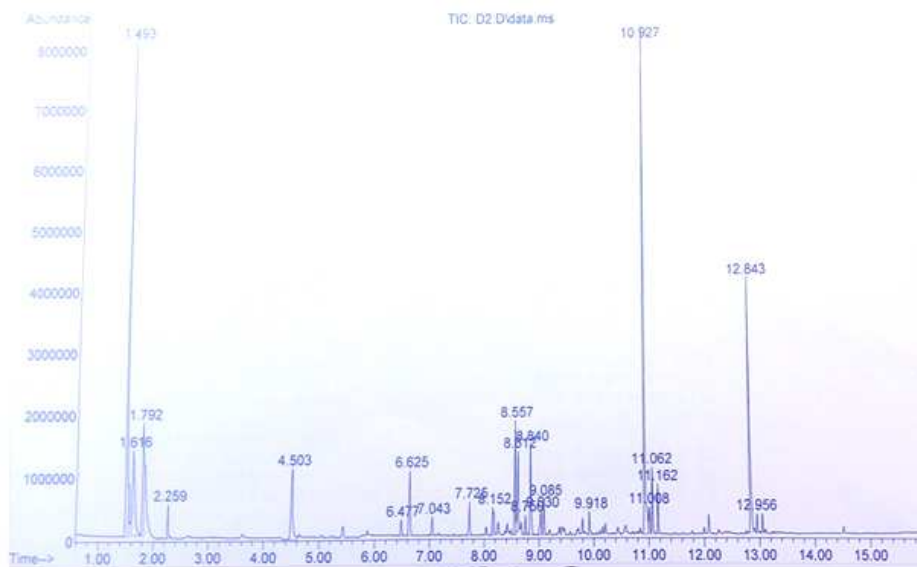


Fig. 4: Chromatogram for sample d, results is presented in Table 4

Appendix 5

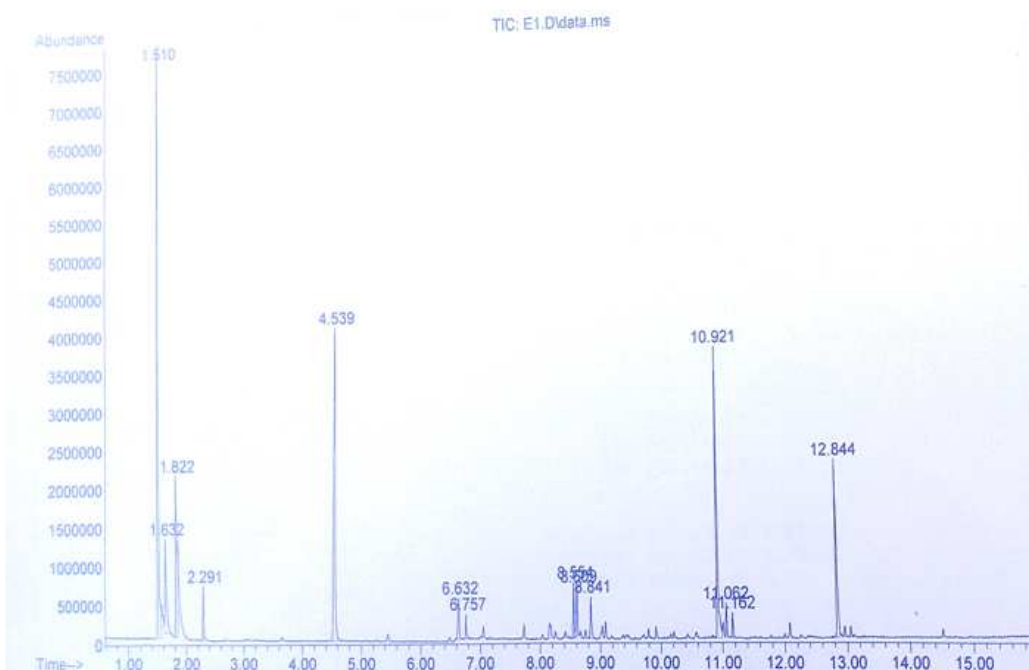


Fig. 5: Chromatogram for sample e, results is presented in Table 5