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Determination of Acetaldehyde and Formaldehyde in Drinking Water Pet Bottles

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Abstract:

In recent years, polyethylene terephthalate (PET) has become the primary packaging material for many food goods, particularly carbonated beverages, and bottled water, as well as chemical industry packaging of various hygiene maintenance agents, insecticides, solvents, and so on. PET provides excellent strength and permeability capabilities for packaging beverages, as well as excellent chemical resistance and transparency. During the thermoforming of PET containers, acetaldehyde (AA) and formaldehyde (FA) are produced. Acetaldehyde and formaldehyde remain trapped in the walls of a PET container after cooling and may migrate into the water after filling and storing. The primary goal of this paper is to investigate the amounts of acetaldehyde and formaldehyde residues in polyethylene terephthalate bottled water and their implications for consumer health. The migration of acetaldehyde (AA) and formaldehyde (FA) from PET bottles into mineral carbonated water was determined in this study using a UV double-beam spectrophotometer method. A default test is also performed to check the pH of the water to determine whether the water is acidic or basic solution, a TDS of water test is performed to determine the total concentration of dissolved substances in drinking water, and an electrical conductivity test is performed to determine how well the water.

Keywords: Formaldehyde, acetaldehyde, polyethylene terephthalate, migration, solvents.

1. Introduction:

The issue of plastic waste is not limited to people. Consuming microplastics may endanger marine species, and the plastic problem extends beyond eating; animals can become entangled or suffocated by trash, which can harm, debilitate, or even kill them. According to some projections, if we continue to consume plastic at this rate, there will be more plastic in the ocean than fish by 2050. Finally, most of the plastic is never recycled. About half of all plastic is used for single-use products, which are intended to be used once and then discarded. Because less than 10% of plastic gets recycled, while recycling is necessary, it is even more important for each of us to refuse [1]**.** Polymerization of ethylene glycol and terephthalic acid yields polyethylene terephthalate (PET). Terephthalic acid is a crystalline solid derived from xylene, whereas ethylene glycol is a colorless liquid derived from ethylene. When ethylene glycol and terephthalic acid are heated together in the presence of chemical catalysts, they generate PET in the form of a molten, viscous material that can be spun directly into Fibers or solidified for further processing as plastic [4]. In chemical words, ethylene glycol is a diol, an alcohol with two hydroxyl (OH) groups in its molecular structure, while terephthalic acid is a dicarboxylic aromatic acid, an acid with a wide six-sided carbon (or aromatic) ring and two carboxyl's (CO2H) groups in its molecular structure. The hydroxyl and carboxyl groups react with heat and catalysts to generate ester (CO-O) groups, which serve as the chemical linkages that connect numerous PET units to form long-chain polymers. ***Figure 1*** shows the chemical structure of polyethylene terephthalate [1].

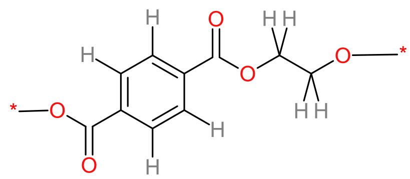


Figure 1:Molecular structure of Polyethylene Terephthalate

The first stage involves the transesterification of dimethyl-terephthalate with ethylene glycol or esterification of terephthalic acid with ethylene glycol, which results in the creation of bis(2-hydroxy-ethyl) terephthalate and minor amounts of bigger oligomer. The second step involves the polycondensation of esters and the creation of polyethylene terephthalate (PET), which is then processed into pellets or Fibers. Acetaldehyde and formaldehyde may occur as unwanted by-products in the creation of PET packaging during the heat degradation of polyethylene terephthalate. Because of the high volatility of acetaldehyde and formaldehyde, there is a considerable possibility of migration from a PET bottle to the packed contents (drinking water), which could result in a change in taste and Odor of the bottled drinking water [4]. The minimal acetaldehyde levels that can be detected in bottled drinking water range from 10 to 20 g/kg. Because the taste of acetaldehydes is hidden by the presence of additional scents, the presence of acetaldehydes in alcoholic and soft drinks that already include acetaldehyde does not pose an issue. The permissible level of acetaldehyde migration from a PET bottle should not exceed 6 mg/kg, and formaldehyde migration should not exceed 15 mg/kg, according to Directive 2002-72-EEC. To determine the amounts of formaldehyde and acetaldehyde, spectrophotometric and chromatographic procedures should be used. The double beam Spectrophotometry is concerned with the measuring of light's interaction with materials. Light can be reflected, transmitted, scattered, or absorbed, and a substance can emit light because of absorbing some light and remitting it, because of gaining energy in another way (e.g., electroluminescence), or because of its temperature [5].

2. Analytical Methods Used For Determining Formaldehyde and Acetaldehyde:

Colorimetric-solid phase extraction method: Colorimetric-solid phase extraction (C-SPE) is a new technique gaining popularity in analytical chemistry, notably for detecting volatile organic compounds (VOCs) such as acetaldehyde and formaldehyde in water samples. This novel technology combines colorimetry principles with solid phase extraction (SPE), providing increased sensitivity and selectivity. The C-SPE method involves passing water samples containing acetaldehyde and formaldehyde through a solid phase extraction cartridge, where the target chemicals are selectively absorbed onto a solid sorbent phase. This procedure efficiently concentrates the analytes, allowing for their subsequent detection [17].

Spectrophotometric Method: A spectrophotometric approach provides an accurate and efficient way to measure the amounts of acetaldehyde and formaldehyde in water samples. This approach is based on measuring the light absorption of certain substances at specified wavelengths. To carry out the procedure, a water sample containing acetaldehyde and formaldehyde is first created. Next, suitable reagents are introduced to the sample to aid chemical processes that change the target compounds into species with specific spectrum characteristics. These processes usually include derivatization, in which compounds undergo a chemical transition to produce molecules that absorb light at specified wavelengths. Following derivatization, the absorption of light by the reaction products is measured with a spectrophotometer. The instrument measures the quantity of light absorbed, allowing you to determine the amounts of acetaldehyde [16].

High Performance Liquid Chromatography (HPLC): High Performance Liquid Chromatography (HPLC) is a popular technique for measuring acetaldehyde and formaldehyde levels in water. This approach involves injecting a sample of water containing the analytes (acetaldehyde and formaldehyde) onto a column filled with a stationary phase. The mobile phase, which is a solvent or a solvent combination, is then pushed through the column under high pressure. As the mobile phase passes through the column, the analytes interact with the stationary phase according to their chemical characteristics, causing them to separate. Acetaldehyde and formaldehyde will have varied retention durations depending on how they interact with the stationary phase, allowing for independent measurement [4]

Table 1: Analytical methods for determination of FA and AA concentrations from various sources of drinking water

Type of water	Bottle capacity	Methods used	Experimental exposure condition			Concentration of AA	Concentration of FA	
			Room temperature	Sunlight	Refrigerator			Author
Mineral water	0.5L	Liquid chromatography-Mass spectrometry	25°	Nil	Nil	143.5 ng/ml	31.4 ng/ml	Yutaka Abe, Norihiro Kobayashi
commercial mineral water	100 ML	ATP-based bioluminescent assay and heterotrophic plate count method	40°	Nil	Nil	44.3 µg/g	2.2 µg/g	M. Mutsuga et al.
natural still water, carbonated water, Natural effervescent water	0.5 or 1.5 L	Solid phase microextraction and gas chromatography	14°	Nil	4°	76.2 µg/g	Nil	Andrea Re Depaolini, Elena Fattore,
Water and acetonitrile	50mg(tablets)	HPLC	40°,60°	Nil	Nil	70.32 µg/g	Nil	S. Kumar and R. K. Srivastava
carbonated and noncarbonated water	0.33 L and 2 L	HPLC	Nil	20°-38°	Nil	Carbonated FA-35, Non-carbonated FA-28.25	Carbonated AA-108.82, Non-carbonated AA-78.05	Azra S. Redžepović, Marijana M. Ačanski,

3. Methods:

This review study on determination of Acetaldehyde and Formaldehyde in PET Bottles is used as a systematic technique to acquire trustworthy information from academic sources. The process required a thorough search and study of material available in the Scopus and Web of Science databases. The process involved the following steps:

Literature Research: A comprehensive search utilizing keywords and phrases linked to Determination of Acetaldehyde and Formaldehyde in PET Bottles and related issues are addressed in this review. The search was confined to the Scopus and Web of Science databases. Access to high-quality, peer-reviewed scholarly publications.

Inclusion Criteria: Search results have been checked using established criteria. We only examined research papers, review articles, and conference proceedings from reputable journals or recognized conferences.

Document Selection: Titles and abstracts of search results were analyzed to find relevant documents on Determination of Acetaldehyde and Formaldehyde in PET Bottles. During this process, documents that were irrelevant or duplicate were excluded. The selected documents were then retrieved in full-text format for more analysis.

Data Extraction and Analysis: Relevant information was extracted from carefully selected documents. The main results, techniques, case studies, and emerging trends were identified and reported. The material acquired from the literature was grouped into the various parts of the review study.

Synthesis and Writing: The selected documents' conclusions, insights, and major points were integrated into the review paper's relevant parts. The information was gathered logically to provide a full overview of the issue. This review study used a systematic and rigorous technique for data collection, analysis, and synthesis. This paper's approach used recognized databases and set inclusion criteria to assure exceptional and trustworthy information, adding credibility and validity to the findings.

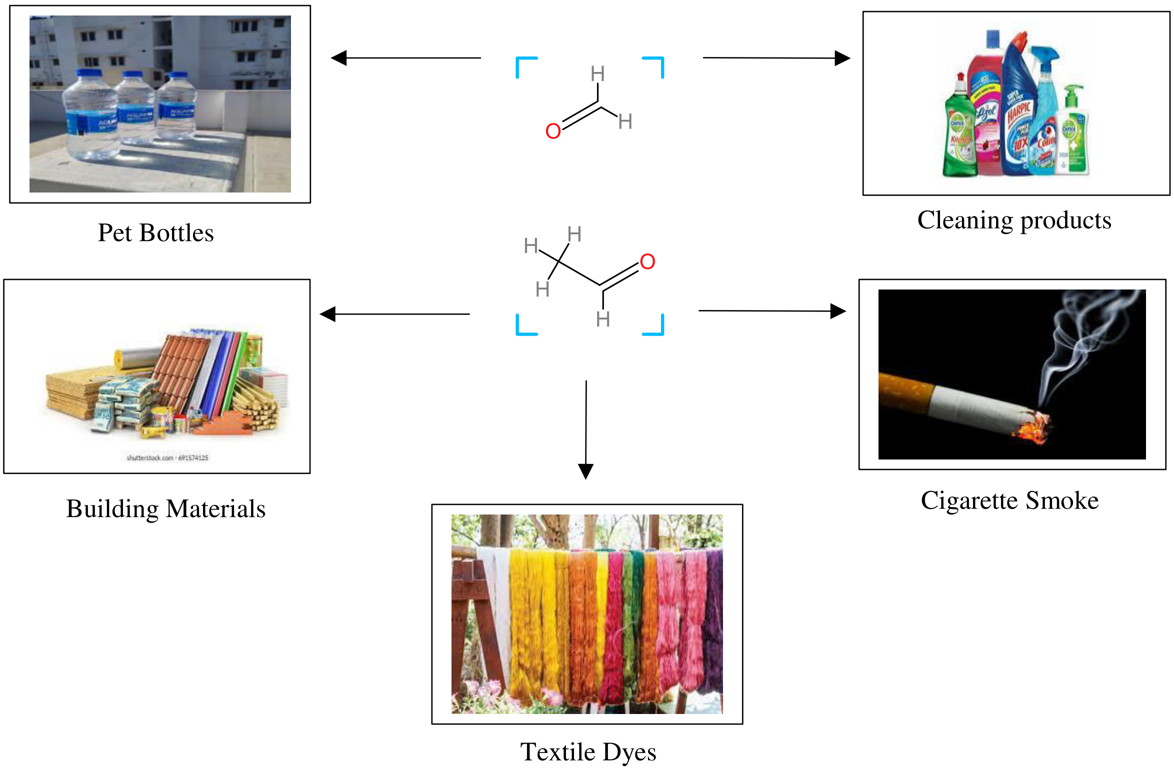
4. Literature Studies on Aldehyde Migrations:

[1] Concentration of migrants, carbonyl compounds from PET containers to packed carbonated and noncarbonated mineral water was detected. The migration may produce chemical and sensory changes in PET-bottled drinking water. PET bottles with varying volumes (0.33 and 2 L) were used to evaluate the migration of formaldehyde and acetaldehyde used in the packing of carbonated mineral water. The water was preserved for one full day (24 hours) or three months. Mineral water was preserved for one day in nine 2 L PET bottles (24 h). Mineral water was preserved for three months in PET bottles of 2 L and in PET bottles of 0.33 L. According to the data, the migration of formaldehyde and acetaldehyde to water begins after 24 hours of storage of mineral water in PET bottles of 2 L. The concentration of formaldehyde in one bottle is always lower than the concentration of acetaldehyde. The concentration of both migrants increased after three months of storage in PET bottles of 2 L. [2]

Sensitive, cost-effective, reproducible high-performance liquid chromatographic method for quantitative detection of acetaldehyde in candesartan cilexetil was developed and validated utilizing the notion of toxicological concern. When acetaldehyde is combined with 2,4-dinitrophenylhydrazine, a Schiff base product with an absorption maximum of 364 nm is formed. On an Intersil ODS 3V, 2504.6 mm, 5 m column with a mobile phase of 40:60 v/v water and acetonitrile at a flow rate of 1.0 ml/min, effective chromatographic separation was accomplished. The injection volume was 30 l, and the column temperature was set to 25°. Within 20 minutes, these conditions resolved the dinitrophenylhydrazine-acetaldehyde product with unreacted dinitrophenylhydrazine, the drug ingredients, and associated impurities, as well as the diluent peak. It was discovered that the risk and quality evaluation report is devoted to estimating the impact of the bottled drinking water package (especially PET one). The work is focused on employing a single integral technique for various risk kinds (factors connected with potential carcinogenicity: concentrations of antimony, formaldehyde, di-ethyl-hexyl-phthalate; and organoleptic factors: turbidity, color, and pH). We suggest the nature of organoleptic (quality assessment) elements that are related to dangerous ones since their indirect influence on chemical polluting power can be amplified. These varieties' tolerable risk limits are set at 10-1 and 10-5, respectively. The study is based on Russian and international (most notably, American) scientific studies and standards. The risk metric is proposed to be calculated in dimensionless numbers (hazard quotient - HQ). In conversion to occurrences per million, HQ can be translated into probabilistic figures (Risk Index - RI and risk of olfactory-reflective effect components, Integral Index of Water Risk). We used the concept of "chronic daily intake" (CDI) in the article as an acceptable risk-free assessment of carcinogenicity factors, which is a suitable evaluation of allowed concentration. A simple and quick analytical method for determining the levels of formaldehyde (FA) and acetaldehyde (AA) in water was devised. FA and AA were derivatized by 2,4-dinitrophenylhydrazine in an LC vial for 20 minutes at room temperature (approximately 25 C) before being analyzed with LC-MS/MS. For FA and AA values ranging from 2-150 ng/mL, the calibration curve demonstrated excellent linearity. Recovery experiments with ultra-pure water and commercially available PET-bottled mineral water samples revealed high precision and trueness. Using this method, we measured the FA and AA contents of 105 PET-bottled mineral water samples from the Japanese market. FA was detected in 61% of the samples at levels ranging from 2.6 to 31.4 ng/mL, whereas AA was detected in 68% of the samples at values ranging from 5.3 to 143.5 ng/ml [3,4]. Formaldehyde (FA) and acetaldehyde (AA) levels in polyethylene terephthalate (PET) bottles and commercial mineral water have been documented. FA (10.1-27.9 mg/l) and AA (44.3-107.8 mg/l) values were found in all water samples bottled in Japan. Eight of the eleven European bottled water samples contained no FA or AA, whereas the remaining three showed measurable quantities of FA (7.4-13.7 mg/l) and AA (35.9-46.9 mg/l). Two of the three North American bottled water samples tested positive for FA (13.6 and 19.5 mg/l) and AA (41.4 and 44.8 mg/l), whereas the third did not. Regardless of provenance, all sterilized water samples included FA and AA, however none of the unsterilized water samples without carbonate contained FA or AA. Three of the carbonated water samples included FA and AA, while one did not. When fortified with FA and AA, the commercial water sample that did not include otherwise detectable FA and AA was able to reduce levels, whereas the commercial water sample that did contain FA and AA was unable to. An ATP-based bioluminescent test and a heterotrophic plate count approach were used to look for bacteria in commercial water samples. [5] This study was to investigate the quantities of acetaldehyde residues in polyethylene terephthalate bottled water and their implications for consumer health. To be representative of the national market, we studied 104 samples obtained throughout Italy. CO2 level, shelf life, weight of the empty bottle, and distance from the manufacturing locations to the point of sale were also obtained. Although the acetaldehyde levels were below the limitations set by Italian regulation, they varied, with concentrations ranging from 0.41 to 76.2 g/L. The level of acetaldehyde in bottled water is unlikely to be of any safety concern for human health, according to an assessment of human health safety using the margin of exposure approach. The acetaldehyde residues were primarily caused by CO2 levels, which influence acetaldehyde solubility in water. Formaldehyde and acetaldehyde concentrations in bottled water were found to be up to 46.0 mg/L and 187 mg/L, respectively. The quantities of formaldehyde and acetaldehyde in the bottled water brands investigated varied but did not surpass the WHO-recommended standards for drinking water. There were no significant differences in formaldehyde and acetaldehyde concentrations across all samples, although there were variances in their levels for a specific brand of bottled drinking water stored under varied settings. The amounts of these aldehydes were found to be lowest in refrigerated samples and greatest in those exposed to sunshine, with levels increasing with storage duration. As a result, it is thought that storage conditions and time influenced the production of these aldehydes in the sample [6,7]. [8] Noticed that Based on the data thus far, it can be concluded that there is no discernible difference in the IR spectra of the bottles under various exposure conditions, showing that the PET does not deteriorate in natural sunlight or during market storage. It could be due to oxidation and carboxylic group production accrued during bottle processing, and so the change accrues for fresh bottles before filling. The lack of antimony detection could be attributed to the employment of a novel type of initiator during the synthesis of PET polymer materials, or to the samples' low Sb levels. According to the findings, the IR spectra of the bottles do not differ under different exposure situations. The half maximum effective concentration (EC50) values were 100 mol/L, and bottled water is not harmful under different weathering storage circumstances. [9] Observed that a High-performance liquid chromatograph was used to determine the amounts of formaldehyde and acetaldehyde (HPLC). Formaldehyde and acetaldehyde levels were evident in all bottled water samples taken from Iranian supermarkets and shops. In this investigation, the minimum and maximum amounts of formaldehyde and acetaldehyde were 12-45 mg/l and 25-120 mg/l, respectively. In Iranian bottled waters, the average amounts of formaldehyde and acetaldehyde were 28.6 mg/l and 61.3 mg/l, respectively. Overall, it is possible to conclude that the bottled waters available in Iran are safe for human consumption in terms of formaldehyde and acetaldehyde levels. During the manufacture of PET bottles, acetaldehyde (AA) and formaldehyde (FA) are generated at elevated temperatures. AA concentrations in PET bottles typically vary from 1 mg/kg (mineral water bottles) to 10 mg/kg (bottles of soft drinks). The employment of scavengers results in a decreased concentration of AA in mineral water bottles. AA concentrations in PET bottled water samples on the Italian market are shown to provide a minor health risk to users. Even assuming the entire mass transfer of AA, the migration of AA from PET bottles can never surpass SML due to low concentrations of AA in the PET bottle wall. However, because the organoleptic threshold is low and ranges between 10 g/L (retro-nasal) and 25 g/L, AA migration into mineral water can readily exceed it. PET bottles do not meet the sensory criteria of Article 3 of Regulation 1935/2004 in this situation due to changes in the taste of water [10].

5. Health Effects of Formaldehyde and Acetaldehyde in Humans Through Various Mediums:

Plastic bottles contain formaldehyde and acetaldehyde. AA and FA are Present in plastic bottles because of manufacturing procedures or plastic component degradation. Migration into the contents is possible, especially at elevated temperatures or long-term storage. Formaldehyde may be present in indoor air due to off-gassing from building materials, furniture, and household goods, and Acetaldehyde may be present in ambient air due to combustion processes such as smoking and car emissions. Workers in manufacturing, healthcare, and labs may be exposed when producing or using formaldehyde-containing goods. Employees in the food sector and chemical industries can be exposed to the acetaldehyde content. Formaldehyde is used in textile finishing procedures, residual levels may be found in clothing items, especially new ones. Acetaldehyde can be used in textile dyes and finishing, sometimes leaving trace quantities in materials. Formaldehyde has been Found in various insulating materials and particle board used in building. Acetaldehyde may be found in building materials due to external contamination or industrial activities. Cigarette smoke contains formaldehyde and acetaldehyde. Cigarette smoke contains both substances, exposing smokers and others nearby to secondhand smoking. Certain cleaning products may include formaldehyde-based preservatives. Acetaldehyde Found in some cleaning solutions and may occur from the breakdown of other chemical components [18]



Flowchart 1- represents the acetaldehyde and formaldehyde Concentrations in Different applications

6. Changes in Acetaldehyde and Formaldehyde Concentration in Foods Depending On The Typical Home Cooking Methods:

Analytical techniques for evaluating hazardous compounds such as acetaldehyde and formaldehyde were verified using solid-phase microextraction-gas chromatography/mass spectrometry in four distinct matrixes. Beef, rapeseed oil, tinned pork ham, egg, and rice wine were all cooked using standard home cooking methods such as boiling, pan-frying, and stir-frying. In addition, monosaccharides, disaccharides, alanine, and glycine were cooked to produce both aldehydes. All validation metrics, including accuracy, precision, limit of detection, limit of quantification, and uncertainty, for four separate matrices fell within suggested limits, validating the present method's validity. Acetaldehyde concentrations varied from undetectable to 17.92 mg/g, whereas formaldehyde levels ranged from undetectable to 0.27 mg/g. In general, boiling reduced both aldehydes except acetaldehyde in eggs. Pan- and stir-frying significantly raised the aldehyde level in rapeseed oil, whereas pan-frying increased the acetaldehyde concentration in canned pig ham and eggs. Fructose and sucrose produced more aldehydes than maltose and glucose when heated. Depending on the kind of food, cooking had varying impacts on acetaldehyde and formaldehyde levels [13]. Figure 4 represents the various kinds of foods used for the study.

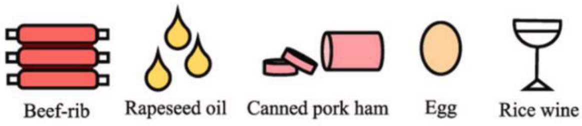


Figure 4 – various kind of foods used for the studies

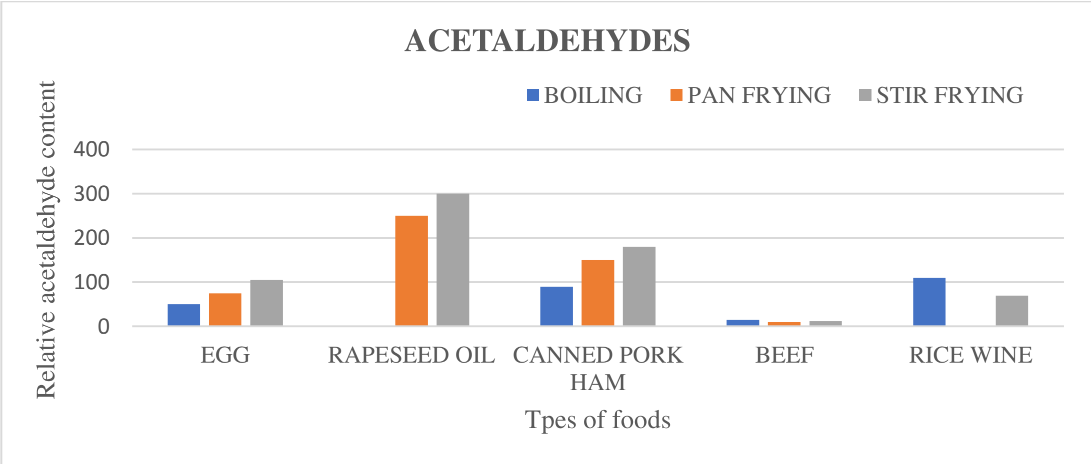


Figure 2- Represents the acetaldehyde content in various cooking methods of foods

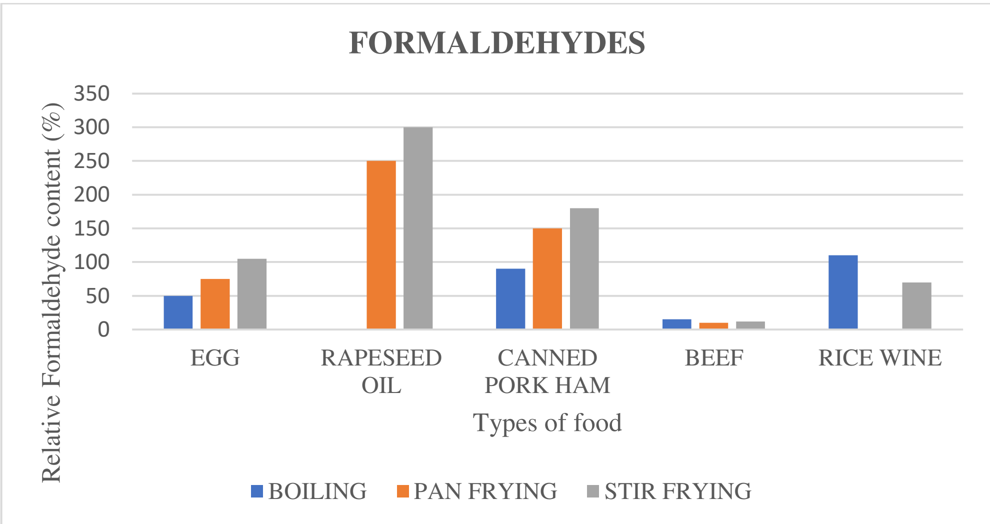


Figure 3: Release of formaldehyde concentration from various cooking conditions

The **Figure 2** and **Figure 3** represent monitoring Acetaldehyde and Formaldehyde contents according to home type cooking methods in foods. The values over and below indicate that cooking treatment increases or decreases the content of Aldehydes. Depending on the food type, different effects of the cooking process were observed in acetaldehyde and formaldehyde contents. It is recommended to reduce the level of aldehyde contents in foods by adjusting cooking methods or conditions [15].

7. Conclusion:

A review of the literature on acetaldehyde (AA) and formaldehyde (FA) determination was carried out. The migration of acetaldehyde and formaldehyde from a PET bottle to bottled drinking water has been observed to increase over time. The migration of acetaldehyde from a PET bottle to bottled drinking water dominates over the migration of formaldehyde. In all the cases the concentrations of acetaldehyde and formaldehyde are always considerably lower than the permitted level of migration from PET packaging (according to the Directive 2002/72/EEC). Based on current IARC evaluations, the presence of this chemical in our food, and the widespread use of PET bottled water, we believe that acetaldehyde residues in water should adhere to the ALARA principle (as low as achievable). The category associations for this product should advocate good practices for a better product, such as polymer quality control and transportation distance limitation.

8. Acknowledgments:

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9. References:

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