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Emissions of selected monoaromatic hydrocarbons as a factor affecting the removal of single-use polymer barbecue and kitchen utensils from everyday use

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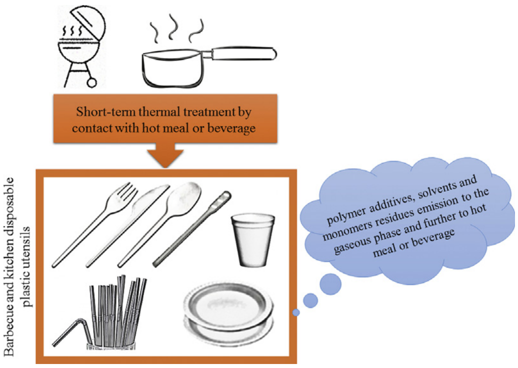
HIGHLIGHTS

- The ATR-FTIR analysis of studied samples of single-use plastic utensils was performed.
- The emission rate of selected monoaromatic hydrocarbons from single-use utensils was investigated.
- For studied samples of single-use plastic utensils the TVOCs parameter was calculated.
- Emitted solvents and monomers residues might be transported directly to the hot meal or beverage.
- The Pearson's coefficients were calculated to assess the realtion between obtained results.

GRAPHICAL ABSTRACT



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ABSTRACT

The main focus of this study is the emission of monoaromatic hydrocarbons because these are the preliminary factors of potential solvent and monomer residues present in single-use plastic barbecue and kitchen utensils comprising polystyrene, polypropylene, natural cellulose, and biodegradable polymers intended for use with hot meal or beverages. Herein, the emissions of monoaromatic hydrocarbons (styrene, benzene, toluene, ethylbenzene, and xylene compounds and the total volatile organic compounds (TVOC)) from nine types of disposable plastic utensils are reported. Seventy two samples of single-use plastic utensils were conditioned at 40 and 80 °C using a stationary emission microchamber system. The average TVOC released from the studied polystyrene, polypropylene, and natural or biodegradable utensils were (2.3 ± 1.3), (1.01 ± 0.15), and (0.48 ± 0.37) μg g^{-1}, respectively, at 40 °C and (11.1 ± 1.2), (46.1 ± 9.5), and (5.5 ± 1.1) μg g^{-1}, respectively, at 80 °C. Significant emissions of styrene (ranged from 3.5 up to 15.3 × 10^3 ng·g^{-1}), toluene (from 2.8 up to 0.53 × 10^3 ng·g^{-1}), and ethylbenzene (from 3.7 up to 5.7 × 10^3 ng·g^{-1}) from the studied samples were observed, especially at 80 °C. Thus, elevated temperatures increase the potential emission of solvent and monomer residues from plastics and could affect the quality of consumed meals or beverages, such as taste. Additionally, to determine the possible interactions between the measured chemical compounds in the plastic utensils, the Pearson's correlation coefficients were calculated.

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1. Introduction

Many consumer products are fully or partially made of synthetic materials, which are widely used as substitutes for paper, glass, and metals because of their practical and economic properties (Siracusa et al., 2008; Chang et al., 2019). Products that can come into direct contact with food or beverages are usually made of a single type of polymer, and the information about the main polymeric material is placed on the product or the packaging (Hakkarainen, 2010; Salthammer et al., 2010; Ghaffar et al., 2014). A special class of everyday polymers includes those used in food utensils, such as disposable barbecue utensils – forks, knives, cups, straws, and plates. In general, these plastic accessories are made from fossil fuels (i.e. natural gas and petroleum) or from a renewable resource such as corn, tapioca, sugarcane, or sugar beets. Various industrial plastics are used as the base substances in disposable utensils, such as styrene-based polymers (polystyrene (PS) and acrylonitrile–butadiene–styrene (ABS)), polyolefins (polyethylene (PE), polypropylene (PP)), acrylics (*homo*-polymethylmethacrylate and *co*-polymethylmethacrylate), polyamides (nylon 6), polyurethanes (polyether and polyester-based polyurethane), cellulose esters, and poly(phenylene oxides) (poly(2,6-dimethyl-1,4-phenylene oxide)). In biodegradable single-use polymer utensils, the most commonly used polymer is polylactic acid (PLA) (Mitchell et al., 2014a; van der Harst et al., 2014). For use with hot meals and hot beverages, vessels, cups, and plates are often made from paper plastic laminates (PPL) consisting of high-quality cellulose fibres with a thin internal layer of PE to prevent liquid intrusion into the absorbent paper layer (Mitchell et al., 2014b). Crucially, the final polymeric product (whether a fossil-fuel-based or biodegradable polymer) in direct contact with food is frequently a heterogeneous mixture containing the main polymer and several additives (Tuomainen et al., 2004; Järnström et al., 2007; Böhm et al., 2012). In fact, the final form of a desired polymeric material can include polymer unreacted monomers, organic solvents, cross-linkers, reaction initiators, pigments, antioxidants, plasticisers, and UV radiation filters (Poςas and Hogg, 2007; Fasano et al., 2012; Yang et al., 2017)

Nowadays, disposable plastic utensils for hot food and beverage consumption are a matter of concern because of their extensive use and, thus, the quantity produced and consumed. This problem has reached such a scale that the European Parliament and European Commission have introduced stringent restrictions on the use of disposable plastic utensils and packaging when affordable alternative solutions are available (Regulation (EC) No. 1935/, 2004; Commission Regulation (EU) No. 10/, 2011). In particular, there are problems associated with polymeric materials intended to come into direct contact with hot meals or hot beverages. During the contact of plastic materials with hot or warm foods or beverages, the transfer of compounds from the plastic utensil into the food/beverage via the gaseous phase is highly possible (Fasano et al., 2012; Rubio et al., 2019).

Even after a short period of exposure to a hot meal or beverage, disposable polymeric utensils can emit various types of chemicals, such as solvent and monomer residues, plasticisers, or UV radiation filters (Hahladakis et al., 2018). Crucially, the chemicals released from plastic materials and transported through the gaseous phase to the food product might affect flavour (Verzera et al., 2010; Van Doorn et al., 2017). The transportation of low-molecular-weight compounds, is the main problem in disposable polymer packaging for food products, especially those in contact with hot meals and beverages. The various types of plastic additives, solvents, and residual monomers are not chemically combined with the polymer structure. Therefore, during the long-term storage of a food product in a plastic container/packaging, there is a strong possibility that chemicals might migrate and dissolve in the foodstuff or beverage (Helmroth et al., 2002; Verzera et al., 2010). In addition, there are several harmful compounds, such as volatile organic compounds (VOCs) (solvents used during the preparation of polymers), phthalates (plasticisers), and benzophenone (a UV radiation filter), that can directly impact human health if they come into contact with the respiratory and digestive systems (Kawamura et al., 2003; Fasano et al., 2012; Kim and Choi, 2014; Moreta and Tena, 2015; Jeon, 2017; Hu et al., 2019). This is an interesting research subject, especially in a case of poor quality disposable polymeric products, which might lose their mechanical properties and undergo irreversible thermal deformation at elevated temperatures (Fig. 1). During deformation, the thermomechanical transformation could result in the emission of a significant amount of chemicals from the polymeric material.

In this study, the interdisciplinary research was performed to provide additional information concerning the factors affecting the removal (or reduction in the quantity) of single-use plastic barbecue and kitchen utensils in everyday use. They key aim was to determine the emissions of selected chemicals (solvent or monomer residues) to the gaseous phase from selected commonly applied single-use polymer utensils. The measured emissions (determined by thermal treatment) from plastic utensils indicate that solvent or monomer residues could be transported directly to hot meals and beverages and might significantly affect flavour and taste. Furthermore, the attempt to develop an analytical protocol to characterise the studied polymeric materials, including their chemical composition and chemicals emission potential, i.e. the emission profile of selected monoaromatic hydrocarbons that represent solvents and monomer residues was described. Moreover, from a social point of view, the study and results will help to increase societal awareness of the quality of polymeric products and to identify the polymeric materials that might have the most significant potential impact on human health.

2. Materials and methods

2.1. Characteristics of the disposable utensils

For the analysis of the emissions of monoaromatic hydrocarbons, nine different disposable plastic utensils were purchased from a local wholesaler. The utensils were collected directly from a new batch of 100 lots packed in closed PE bags. For each sample, 10 random products were selected from a batch and covered with aluminium foil before laboratory analysis. These materials were (i) white PS cups for hot beverages (KB_PS), average mass of (0.6934 ± 0.0045) g; (ii) transparent PS teaspoons (LAB_PS), average mass of (1.429 ± 0.047) g; (iii) black PS lids for takeaway coffee cup (LID_PS), average mass of (3.074 ± 0.095) g; (iv) black PP straws for hot and cold beverages (SL_BC), average mass of (1.500 ± 0.074) g; (v) white PS knifes (SZT_WH), average mass of (2.249 ± 0.081) g; (vi) transparent PS forks (SZT_BB), average mass of (4.313 ± 0.041) g; (vii) cups made from paper plastic laminates (PPL) consisting of high-quality cellulose fibre with a thin internal PE coating (KB_CEL_PE), average mass of (3.43 ± 0.13) g; (viii) black PP

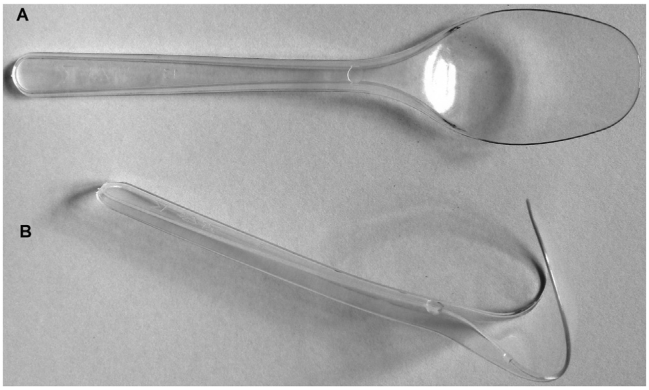

Fig. 1. Appearance of a disposable transparent teaspoon made of polystyrene (A) before and (B) after being used to mix a hot beverage.

teaspoons for ice cream (LAC_PP), average mass of (1.7321 ± 0.0077) g; and (ix) transparent biodegradable PLA straws (SL_PLA), average mass of (1.883 ± 0.021) g. All of the selected plastic utensils are commonly used and are intended to come into contact hot meals and beverages.

All the food accessories were classified into three groups on the basis of the main polymeric material: (i) PS, (ii) PP, and (iii) partially or completely made of natural or biodegradable material (cellulose or PLA, respectively). The total number of disposable plastic utensils analysed for the emissions of monoaromatic hydrocarbons was 72.

2.2. Reagents, analytical equipment, and analytical procedure

A commercially available reference standard solution containing 13 VOCs (including monoaromatic hydrocarbons) dissolved in methanol at a concentration of 2000 $\mu g \cdot mL^{-1}$ each (EPA VOC Mix 2, Supelco, USA) was used as an external standard for the thermal desorption–gas chromatography–flame ionisation detector (TD-GC-FID) calibration process. As an internal standard for the total VOCs (TVOCs) parameter calculation and as an injection and recovery standard, a commercially available reference standard solution containing deuterated toluene in methanol at a concentration of 2000 $\mu g \cdot mL^{-1}$ (d_8-toluene, Supelco, USA) was employed. Methanol for GC (MS SupraSolv®, Merck KGaA, Darmstadt, Germany) was used as a solvent during the preparation of the calibration solutions.

Stainless-steel tubes containing Tenax TA (Merck KGaA, Darmstadt, Germany) were used as the sampling devices for monoaromatic hydrocarbons emitted from the studied materials to the gaseous phase. Applied tubes with sorbent were cleaned prior to use by thermal extraction under a stream of inert gas (300 °C for 30 min under nitrogen at a flow rate of 50 $mL \cdot min^{-1}$).

The studies of the selected monoaromatic hydrocarbons (as a representative of potential solvent residues) released to the gaseous phase from commercially available disposable plastic barbecue utensils were performed using a Micro-Chamber/Thermal Extractor™ (μ-CTE™ 250, Markes International, Inc) equipped with four similar high-quality polished steel microscale chambers (inner volume: 114 cm^3 each). Detailed information about the operating parameters and the range of working conditions of the stationary analytical equipment used has been reported previously (Marć et al., 2015; Marć and Zabiegała, 2017; Marć et al., 2017). To achieve analyte liberation from the Tenax TA sorbent, a two-stage TD technique was applied (Markes Series 2 Thermal Desorption Systems; UNITY/TD-100). For analyte separation, identification, and determination, GC-FID system was used (Agilent 7820A GC, Agilent Technologies, Inc.). The selected disposable plastic utensil samples were analysed in a stationary emission chamber at 40 and 80 °C. The lower temperature was used to assess the emissions of selected monoaromatic hydrocarbons from a given plastic material in the case when the food product (meal or beverage) was lukewarm or cold (i.e. a long time after preparation or un-heated beverages), whereas the higher temperature was used to estimate the emissions of chemical compounds from the utensils when directly exposed to hot meals or beverages (i.e. shortly after thermal treatment). Detailed information about the analytical procedure used to assess the emissions of selected monoaromatic hydrocarbons from the studied disposable plastic utensils is shown in Supplementary Fig. 1.

To obtain information about the type of polymer comprising the disposable plastic utensils, Fourier transform infrared spectroscopy (FTIR) analysis was performed. The FTIR analysis of the sampled utensils was carried out with a Nicolet Spectrometer IR200 from Thermo Scientific (Waltham, MA, USA). The device had an attenuated total reflection (ATR) attachment with a diamond crystal. Measurements were performed with 1 cm^{-1} resolution in the range from 4000 to 400 cm^{-1} using 64 scans.

2.3. Calibration of the TD-GC-FID system and quality assurance and quality control (QA/QC) characteristics

The mass of monoaromatic hydrocarbons (and the emission in nanograms of chemical compound per gram of studied material) released from the studied disposable plastic utensils to the gaseous phase and collected on the Tenax TA was evaluated using the external standard calibration method (ESTD). To calibrate the TD-GC-FID system, a reference methanolic standard solution containing 13 VOCs (including benzene, toluene, ethylbenzene, and xylenes and styrene) at a concentration of 2000 $\mu g \cdot mL^{-1}$ each and a reference standard solution containing deuterated toluene in methanol (2000 $\mu g \cdot mL^{-1}$) were employed. The calibration of the TD-GC-FID system using the standard solutions was carried out using the previously published procedure (Pennequin-Cardinala et al., 2005; Zabiegała et al., 2010; Marć et al., 2014; Marć et al., 2017). Briefly, to achieve a high quality calibration, two calibration curves were prepared in two mass ranges: (i) 1–200 ng per sorbent and (i) 200–4000 ng per sorbent. This allowed the measurement of analytes at both very low and very high concentrations with good linearity of detector response. Each calibration curve contained five points associated with five freshly prepared calibration solutions in methanol. Each point on the calibration curve was repeated three times. Each time, 1 μL of freshly prepared calibration solution was injected via syringe directly into the Tenax TA and then transferred to the unit with a flow of inert gas. Thus, the analytes were transferred to the furthest parts of the sorption material and the solvent (methanol) was removed. After a defined time period, the tubes were removed from the unit, transferred to the TD-GC-FID system, and analysed using the same TD-GC-FID conditions as the real samples. The values of the correlation coefficients (R^2) of the calibration curves for analytes from 1 to 100 ng per sorbent ranged from 0.987 to 0.996 for benzene and styrene, respectively, and those from 100 to 4000 ng per sorbent were estimated to range from 0.992 to 0.995 for benzene and styrene, respectively.

To ensure the quality of the analytical procedure and results (QA/QC protocol), several standard solutions were employed. In the case of the Tenax TA, before each analysis, the tubes were conditioned for 30 min at 300 °C under a stream of inert gas. After each analysis (containing four Tenax TA tubes), a randomly selected tube was once again analysed to check the analyte desorption efficiency. In addition, a defined amount of deuterated toluene (500 ng per tube), which represents a compound that could be adsorbed on the sorption medium in the strongest way, was used to check the recovery of the desorption process. It was measured that the recovery of analytes was ±7%, which is an acceptable value in the case of TD. To consider background effects, before each analysis series, the value of a blank sample was checked. The estimated value of the blank sample takes into account the purity of the applied gases (especially nitrogen attached to the μ-CTE-250 system), the wall-memory effects of the stainless-steel emission chambers in the μ-CTE-250 system, the purity of the applied sorption medium, the purity of the microtrap in the TD unit, and the potential impurities that might be present in the chromatographic system. The blank sample analysis was performed under similar conditions as those used for the analysis of the calibration solutions and real samples and estimated for the entire measured set, thus taking into account the factors mentioned above. The calculated masses of the analytes in the monoaromatic hydrocarbons group were corrected for the blank sample value. Moreover, to consider potential changes in the detector response during the study and potential losses of analytes during the sampling and liberation of analytes, before every sampling period, a defined amount of deuterated toluene was placed on each sorbent in a stainless-steel tube (500 ng per sorbent). The limit of detection (LOD) was calculated based on the calibration curve equation for the smallest mass of analytes introduced on the sorbent, and ranged from 0.040 ng for ethylbenzene to 0.063 ng for benzene. The limit of quantitation (LOQ) was calculated as three times the LOD values.

3. Results and discussion

3.1. Identification of the main polymeric materials – ATR-FTIR analysis

Manufacturers of single-use plastic utensils are obliged to declare the polymer used in plastic utensils for food use. Nevertheless, it is desirable to confirm this information and to identify the key polymer type. The results of the ATR-FTIR analysis of the plastic samples are shown in Fig. 2. The IR spectra shown in Fig. 2 were compared to spectra in the Hummel Polymer Sample Library database, which contains the IR spectra of frequently used polymers. For all spectra, the confirmation/match factors ranged from 93% to 96%. However, a manual interpretation, as well as a comparison with literature IR spectra, was also carried out. For this reason, detailed information about assignments of various prominent IR bands (wavenumbers and the corresponding types of vibration) for the shown in Fig. 2 results of ATR-FTIR spectrum of selected single-use plastic utensils was listed in Table 1.

The ATR-FTIR analysis of the black straws for hot and cold beverages indicates that these are composed of atactic PP, and the IR spectra contain clear characteristic absorption bands in the range 2950–2848 cm^{-1}. These bands are manly associated with the symmetric and asymmetric stretching vibrations of the C—H bonds present in the –CH$_2$ groups of the aliphatic chains. A characteristic medium intensity peak at 1375 cm^{-1} corresponds to the deformation vibrations of methyl groups (methylene chains located between the methyl group and the main chain in the form of alkyl branches) was observed (Nekhoroshev et al., 2005). The medium intensity bands from 1461 to 1452 cm^{-1} might be typical of the asymmetric and symmetric bending vibrations of the –CH$_2$ and –CH$_3$ groups (Santhoskumar et al., 2010; Ebadi-Dehaghani et al., 2016). For the white plastic hot beverage cups and the transparent teaspoons, two peaks – strong at 696 and medium at 750 cm^{-1} were observed, which could be associated with the styrene copolymers and their derivatives. In addition, observed medium intensity peaks at 1492 and 1452 cm^{-1} might correspond to out-of-plane bending vibrations of the C—H groups of benzene rings with one substituent characteristic for polystyrene and styrene copolymers (Motyakin and Schlick, 2006). There was a clear medium intensity peak at 1600 cm^{-1}, which might correspond to the vinyl group (C=C stretching vibrations) characteristic for PS materials (Munteanu and Vasile, 2005; Olmos et al., 2014). The final FTIR spectrum in Fig. 3 is that of the transparent straws for hot or cold beverages, which (according to the manufacturer) are made of PLA, a type of aliphatic polyester derived from renewable resources. The intense bands from 1200 to 1050 cm^{-1} might correspond to the ester and lactone compounds. In addition, the most intense peak at 1083 cm^{-1} might be associated with the –C-O-C– symmetrical stretching vibration. The FTIR spectra between 1760 and 1740 cm^{-1}, might correspond to the stretching vibration of C=O bonds (especially the strong peak at 1747 cm^{-1}) (Mofokeng et al., 2011).

3.2. Emissions of measured monoaromatic hydrocarbons from the studied disposable plastic barbecue and kitchen utensils

3.2.1. Disposable plastic utensils made of PS

On the basis of the ATR-FTIR analysis (see spectra in Fig. 2 and Table 1), the main chemical compounds that could be emitted from the PS utensils could be (i) unreacted polymer monomers (mainly unreacted styrene, which remains in the resin if the polymerisation reaction is not complete) (Gennari et al., 2012) and (ii) volatile residual solvents used during the polymerisation process, such as toluene or ethylbenzene. Moreover, it might be expected that aromatic hydrocarbons will have a much greater impact on a total amount of compounds released into the gaseous phase from the samples than aliphatic

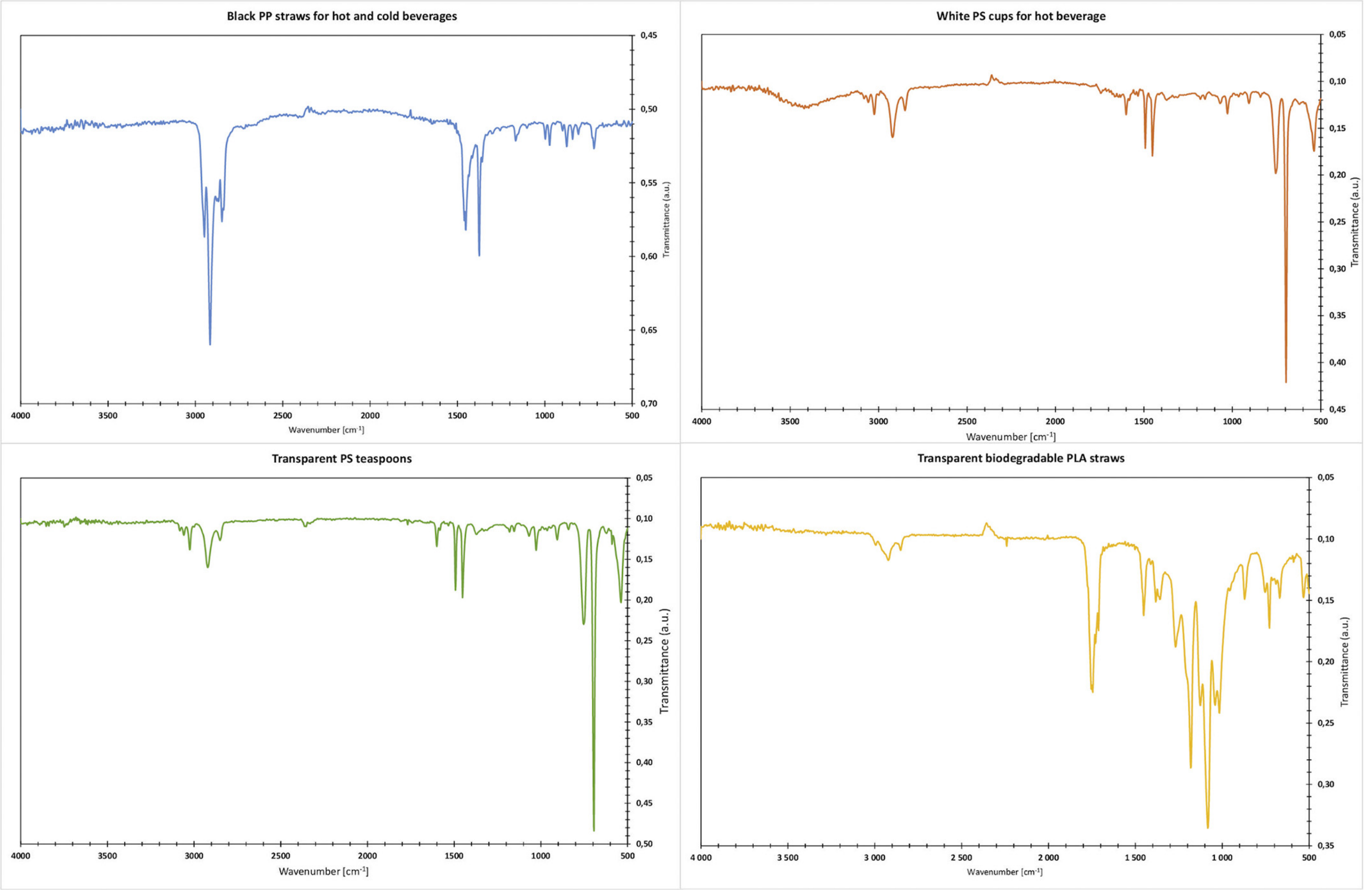

Fig. 2. ATR-FTIR spectra of selected disposable utensils made of polystyrene, polypropylene, and biodegradable polylactic acid.

Table 1
Assignments of various IR bands for selected samples of disposable plastic utensils (frequencies are recorded with an accuracy of ±1 cm^{-1}) .

Wavenumber (cm^{-1})	Vibration type and assignment	Studied sample
2951 (m) 2916 (s) 2850 (m) 1452 (m) 1375 (m) 1169 (w) 999 (w) 974 (w) 839 (w) 808 (w)	-CH_3 asymmetrical stretching -CH_2 asymmetrical stretching -CH_3 stretching -CH_3 asymmetrical bending -CH_3 symmetrical bending C – H wagging C – C stretching C – C stretching C – H rocking C – C skeletal	Black straws for hot and cold beverages made of atactic PP
2993 (w) 2922 (w) 2852 (w) 1753–1747 (m) 1452 (w) 1383–1358 (w) 1267 (w) 1180 (s) 1083 (s) 1038 (m) 872 (w)	-CH_3 asymmetrical stretching C – H asymmetrical stretching C – H symmetrical stretching C = O stretching -CH_3 asymmetrical bending -CH_3 symmetrical bending -C = O bending -C-O stretching -C-O-C– symmetrical stretching C—H deformation C – H rocking	Transparent biodegradable PLA straws
3028 (w) 2918 (m) 2854 (w) 1603 (w) 1493 (m) 1452 (m) 1026 (w) 756 (m) 694 (s) 536 (m)	C – H stretching (ring) -CH_2 asymmetrical stretching -CH_2 symmetrical stretching C = C stretching (vinyl group) C – H out-of-plane bending (ring) C = C out-of-plane bending (ring) C = C stretching (ring) C – H bending (ring) C – H bending (ring) C – H skeletal (ring)	Transparent PS teaspoons
3028 (w) 2918 (m) 2848 (w) 1600 (w) 1493 (m) 1452 (m) 1026 (w) 752 (m) 696 (s) 536 (m)	C – H stretching (ring) - CH_2 asymmetrical stretching -CH_2 symmetrical stretching C = C stretching (vinyl group) C – H out-of-plane bending (ring) C = C out-of-plane bending (ring) C = C stretching (ring) C – H bending (ring) C – H bending (ring) C – H skeletal (ring)	White PS cups for hot beverage

s – strong; m – medium; w – weak.

hydrocarbons. The estimated emissions of monoaromatic hydrocarbons released at 40 and 80 °C from the studied PS utensils are listed in Table 2. As shown by the listed values, the emissions are temperature dependent, and an increase in temperature causes a significant increase in the emission of monoaromatic hydrocarbons to the gaseous phase. However, the dynamics of this process is affected by the quality and composition of the base polymer, as well as the additives used during its manufacture. For example, the white PS cups for hot beverages showed significant differences in emissions at the two different temperatures. The smallest difference in the monoaromatic hydrocarbon emissions at different temperatures were observed for black PS lids for coffee-to-go cups, possibly because of the use of pigments, such as carbon black. This inorganic pigment is very stable in polymer matrices, bonding and forming interactions with the base polymer (Bolgar et al., 2008). As for the pigment/colorant samples, the transparent PS products showed characteristically smaller standard deviations in the obtained emissions. Thus, it might be assumed that the transparent materials are greater homogeneous than those containing pigments. After analysing the information listed in Table 2, styrene and ethylbenzene were found to be emitted in the greatest quantities of all measured monoaromatic hydrocarbons in both the PS samples and at both temperatures. In addition, the mentioned representative monoaromatic hydrocarbons significantly affected the TVOC value. In the case of ethylbenzene emitted from PS samples at 40 °C, the contribution of this compound to the final TVOC values was 2.00% (for white PS cups for hot beverage and white PS knifes) and 13.75% (for black PS lids for coffee-to-go cups). At 80 °C, the contribution of ethylbenzene to the final TVOC value ranged from 1.52% to 14.01% for white PS knifes and black PS lids for coffee-to-go cups, respectively. Styrene, the main monomer used to produce PS, was emitted in the greatest quantity from the studied utensils. Furthermore, its effect on the final TVOC values was the most significant at both temperatures, ranging from 15.72% to 71.29% at 40 °C and 37.81% to 73.20% at 80 °C for the previously mentioned samples. These results are consistent with the results of published previously papers that considered PS materials such as coffee-to-go lids, packaging materials, stereo cassette players, cassette tapes, strips, mugs, and expanded PS. In published research investigating PS items, the main chemical compounds emitted from the studied materials were styrene, ethylbenzene, and, in several cases, toluene (Verzera et al., 2010; Mitchell et al., 2014a; Marć and Zabiegała, 2017). According to Verzera et al. (2010) the transport of styrene monomers and ethylbenzene, which might affect the quality of consumed food products, varies with the physical and chemical characteristics of the polymer and the foodstuff for foods contained in the polymer packaging.

3.2.2. Disposable plastic utensils made of PP

In the ATR-FTIR data in Fig. 2 and Table 1 for disposable PP utensils, there is a lack of characteristic aromatic ring vibrations. Thus, it is possible that the emissions of monoaromatic hydrocarbons from the PP samples could be lower than those of the PS samples. The estimated emissions of the main monoaromatic hydrocarbons at two different temperatures from the surfaces of the PP utensils are listed in Table 3. On the basis of these data, the PP utensils could be a significant source of emissions of toluene and ethylbenzene to the gaseous phase and, thus, the food product. Moreover, as for PS, the temperature significantly affected the emissions of the mentioned compounds. In the case of toluene, the emissions increased on increasing the temperature from 40 to 80 °C by 1.4 and 2.2 orders of magnitude for black PP straws for hot and cold beverages and black PP teaspoons for ice creams, respectively. For the same samples, for ethylbenzene, the emissions were increased by 1.5 and 1.9 orders of magnitude, respectively. Additionally, benzene, styrene, and *p*- and *m*-xylene were emitted at 40 °C at very low levels (even below the LOD values). However, in the case of benzene, the temperature did not significantly affect its emissions. Thus, disposable PP utensils and other everyday materials made of PP cannot be considered an important source of benzene emissions. The low emissions of the determined monoaromatic hydrocarbons at 40 °C might also be due to the use of pigments, such as carbon black. As mentioned before, this pigment is a sorbent, and its addition to the polymer might prevent the release of chemicals from the polymeric matrix into the gaseous phase at room or slightly higher temperatures.

As shown by the TVOC values listed in Table 3, for both types of PP samples, the TVOCs measured at 80 °C were about ten times higher

Table 2
General parameters of measured monoaromatic hydrocarbons emitted from the studied samples of disposable polystyrene barbecue and kitchen utensils.

Parameter	Benzene 40 °C [ng·g^{-1}]	Benzene 80 °C [ng·g^{-1}]	Toluene 40 °C [ng·g^{-1}]	Toluene 80 °C [ng·g^{-1}]	Etylbenzene 40 °C [ng·g^{-1}]	Etylbenzene 80 °C [ng·g^{-1}]	p,m-Xylene 40 °C [ng·g^{-1}]	p,m-Xylene 80 °C [ng·g^{-1}]	Styrene 40 °C [ng·g^{-1}]	Styrene 80 °C [ng·g^{-1}]	TVOC 40 °C [μg·g^{-1}]	TVOC 80 °C [μg·g^{-1}]
White PS cups for hot beverage (KB_PS)												
Mean	2.69	23.3	25.2	137	660	5669	15.5	183	1070	15,295	6.8	40.5
Stan. dev.	0.98	2.5	6.3	18	169	701	4.6	42	239	1726	6.1	4.0
Median	2.40	23.6	26.0	139	662	5367	15.8	190	1064	14,677	4.9	3.9
Min	1.81	20.4	15.2	107	382	4855	8.8	93	639	13,706	3.4	36.9
Max	5.12	27.0	34.6	160	889	6876	21.7	224	1355	18,549	23.9	47.4
Transparent PS teaspoons (LAB_PS)												
Mean	1.16	3.6	34.6	151	19.6	114	2.1	20.7	698	3993	0.98	5.5
Stan. dev.	0.31	1.4	7.7	18	3.3	13	3.8	3.2	109	723	0.23	1.0
Median	1.22	3.4	33.5	155	19.8	115	0.50	21.7	725	4046	0.93	5.8
Min	0.66	1.5	23.9	115	13.8	87	0.13	13.1	514	2437	0.68	3.3
Max	1.53	6.3	49.4	180	23.7	134	10.7	23.7	842	4650	1.48	6.2
Black PS lids for coffee-to-go cups (LID_PS)												
Mean	1.04	1.53	8.0	14.3	214	328	51.5	99	901	2610	1.56	4.40
Stan. dev.	0.35	0.98	1.7	4.2	35	55	8.0	15	135	319	0.25	0.45
Median	1.07	1.14	8.0	13.2	209	318	50.3	98	878	2648	1.57	4.41
Min	0.40	0.95	5.8	11.4	175	261	42.5	77	727	2152	1.16	3.87
Max	1.50	3.83	11.3	24.4	272	409	63.9	121	1135	3068	1.92	5.09
White PS knifes (SZT_WH)												
Mean	4.10	8.2	32.0	55	35.8	62	4.05	22.5	1166	2396	1.81	4.06
Stan. dev.	0.76	1.6	2.8	10	2.4	13	0.30	4.6	103	523	0.068	0.48
Median	3.92	8.1	32.2	51	35.8	58	4.09	23.5	1174	2593	1.789	4.13
Min	3.31	4.7	28.2	44	33.2	45	3.65	15.1	1053	1590	1.729	3.09
Max	5.24	10.2	36.2	71	38.8	82	4.56	27.3	1334	2916	1.923	4.56
Transparent PS forks (SZT_BB)												
Mean	0.56	1.39	1.75	6.9	21.5	92.58	<LOD	0.68	132	644	0.211	1.14
Stan. dev.	0.28	0.58	0.70	2.2	4.7	8.8	–	0.32	23	104	0.022	0.21
Median	0.48	1.43	1.83	7.3	21.6	95.09	<LOD	0.59	140	674	0.220	1.13
Min	0.25	0.56	0.58	3.8	14.2	77.70	–	0.44	90	488	0.163	0.84
Max	0.97	2.10	2.66	9.4	30.8	101.06	–	1.40	153	760	0.224	1.48

those measured at 40 °C. Interestingly, the monoaromatic hydrocarbon emissions from the studied disposable PP utensils make a very small contribution to the final TVOC values. In the case of ethylbenzene emissions from the studied PP samples at 40 °C, the final TVOC values ranged from 1.71% (black PP straws for hot and cold beverages) to 10.34% (for black PP teaspoons for ice creams). At 80 °C, the contribution of ethylbenzene to the final TVOC value ranged from 2.58% to 8.06% for black PP straws for hot and cold beverages and black PP teaspoons for ice creams, respectively. The results can be explained by combining the ATR-FTIR analysis and emission measurements. PP does not contain aromatic rings, so monoaromatic hydrocarbon emissions should make little contribution to the TVOC parameter. Considering the chemical structure of PP and analysing the GC-FID results, the main factor affecting the TVOC parameter could be linear or branched aliphatic hydrocarbons, such as 2,2,4,6,6-pentamethyl heptane, decane, undecane, octadecane, dodecane, and tridecane. These chemical compounds might be generated by the thermal degradation of PP and have a more significant impact on the TVOC parameter than monoaromatic hydrocarbons. This phenomenon was reported by Mitchell et al. (2014a). On analysing the PP samples, the authors noted that a large quantity of branched alkanes was emitted, and the presence of these emissions can be used as tentative identification of PP-based objects (Mitchell et al., 2014a). Thus, the presence and emission of monoaromatic hydrocarbons from disposable PP utensils might be due to the additives, pigments, and solvents used during their manufacture but not the PP itself.

Table 3
General parameters of measured monoaromatic hydrocarbons emitted from the studied samples of disposable polypropylene barbecue and kitchen utensils.

Parameter	Benzene 40 °C [ng·g^{-1}]	Benzene 80 °C [ng·g^{-1}]	Toluene 40 °C [ng·g^{-1}]	Toluene 80 °C [ng·g^{-1}]	Etylbenzene 40 °C [ng·g^{-1}]	Etylbenzene 80 °C [ng·g^{-1}]	p,m-Xylene 40 °C [ng·g^{-1}]	p,m-Xylene 80 °C [ng·g^{-1}]	Styrene 40 °C [ng·g^{-1}]	Styrene 80 °C [ng·g^{-1}]	TVOC 40 °C [μg·g^{-1}]	TVOC 80 °C [μg·g^{-1}]
Black PP straws for hot and cold beverages (SL_BC)												
Mean	1.8	1.83	20.5	526	22.3	769	2.73	30.4	2.6	16.3	1.30	29.8
Stan. dev.	1.2	0.75	7.5	176	5.0	78	0.82	4.2	2.7	8.9	0.26	4.1
Median	1.4	1.87	19.3	528	20.5	773	2.84	30.5	1.9	15.4	1.29	29.6
Min	1.2	0.76	12.2	315	16.4	675	1.32	24.5	0.29	5.7	0.86	22.0
Max	4.6	2.64	32.0	758	32.0	891	3.74	36.5	6.3	33.0	1.76	35.7
Black PP teaspoons for ice-creams (LAC_PP)												
Mean	0.64	<LOD	0.57	96.2	71.5	5029	<LOD	77	<LOD	22.3	0.691	62
Stan. dev.	0.21	–	0.18	7.4	4.1	759	–	11	–	5.5	0.046	15
Median	0.64	–	0.50	95.7	71.7	4787	–	79	–	22.0	0.689	59
Min	0.39	–	0.43	88.2	67.3	4441	–	62	–	17.3	0.639	49
Max	0.90	–	0.84	105.1	75.3	6099	–	88	–	27.9	0.748	83

3.2.3. Disposable plastic utensils made of natural or biodegradable materials

As shown in the ATR-FTIR data in Fig. 2 and Table 1 for the biodegradable disposable utensils made of PLA, there is a lack of strong bands corresponding to aromatic rings. However, there is a medium intensity peak at 729 cm^{-1}, which could be related to aromatic ring out-of-plane deformation vibrations, for example, benzene, toluene or their derivatives. Thus, several monoaromatic hydrocarbons (at low or very low concentrations) might be present in studied product made of PLA and be emitted from it to the gaseous phase. Table 4 lists the emissions of the monoaromatic hydrocarbons at two different temperatures from the non-standard polymers (natural cellulose fibre and PLA). For the biodegradable utensils (PLA straws), interestingly, the emissions of ethylbenzene and styrene decreased significantly as the treatment temperature increased. Thus, disposable PLA utensils are potential sources of ethylbenzene and styrene only at the beginning of their use, and the emissions of those compounds is intense even at slightly elevated temperature. After treatment at 40 °C, the emissions of ethylbenzene and styrene decreased to very low levels (residual emission at 80 °C in the range from 2.1 up to 6.8 ng·g^{-1} and 2.0 up to 8.0 ng·g^{-1} for ethylbenzene and styrene, respectively). Generally, it is possible that the detected ethylbenzene and styrene in PLA utensils at seasoning temp. 80 °C might originate from ambient air that became enclose in the sample chamber during material sampling. Additionally, mentioned compounds might be adsorbed on the surface of studied PLA utensils during their storage process (lack of information from the manufacturer about specific storage time and conditions) and then during their seasoning in the chamber at 40 °C and under inert gas flow rate might be rapidly released from the surface of studied materials to the gaseous phase. This might lead to the conclusion, that ethylbenzene and styrene are not strongly combined to the surface of studied PLA samples and during their seasoning process, they might be flushed and the emission of mentioned compound decreases. Thus, considering this and the values in Table 4, the studied disposable PLA utensils might not be significant sources of ethylbenzene and styrene emissions. Davis et al. (2019) also observed that styrene and ethylbenzene are emitted from PLA materials (PLA-based 3D printer filament) in detection frequencies of 56% and 33%, respectively (from 0.1 to 0.2 μg·g^{-1}). Moreover, Tábi et al. (2010) noticed that the crystalline structure of PLA might undergo some changes at 80 °C, this phenomenon might affect the diffusion and potential emission of selected VOCs to the gaseous phase.

As for the other monoaromatic hydrocarbons emitted from the studied PLA samples, there is a clear relationship between the emission quantity and the temperature. Increasing the temperature in the environmental chamber significantly increased the emissions of benzene, toluene, and *p*, *m*-xylene to the gaseous phase. One of the general reasons for this situation is the fact that applied solvents (mainly toluene and *p*, *m*-xylene) used during the manufacturing process of a final PLA single-use utensil might be contaminated with benzene. The emission of these compounds, especially toluene and *p*, *m*-xylene, was also confirmed during studies on PLA-based materials performed by Kim et al. (2015). The increasing emissions of organic compounds with increasing temperature was also associated with an increased TVOC parameter. The TVOC parameter increased by approx.. 12 times and it might be also related to the relatively linear polymer structure. The direct effect of the temperature increase on the TVOC parameter (the increase in emissions of a broad range of VOCs) in the case of materials made of PLA was also reported by Steinle (2016). In addition, Davis et al. (2019) and Azimi et al. (2016) reported the general effects on TVOC values (excluding the mentioned monoaromatic hydrocarbons) in the cases of PLA materials containing caprolactam, acetophenone, benzaldehyde, nonanal, decanal, and 1-butanol. However, at this point it should be highlighted that the manufacturer of studied single-use PLA utensils do not put the information about the type of renewable resource (corn, sugar beet pulp, etc.) from which the monomer was formed. It is possible to conclude that several contaminants present in employed renewable resource might be released form the final plastic product.

Considering the data listed in Table 4 for the high-quality cellulose fibre with a thin internal PE coating, increasing the treatment temperature caused a reduction in the emissions of all of the measured monoaromatic hydrocarbons. However, the TVOC value at 80 °C was approximately three times higher than at 40 °C. Thus, the disposable utensils are likely not significant sources of monoaromatic hydrocarbons, and their presence in the studied samples might be caused by adsorption of monoaromatic hydrocarbons from the air (gaseous phase). As reported by Ion et al. (2015), cellulose can be used for the removal and recovery of VOC species from polluted air streams. In addition, several monoaromatic hydrocarbons might be present in the thin PE film (internal surface of the coffee-to-go cups) and be emitted to the gaseous phase. However, Mitchell et al. (2014a) reported that cellulose fibres are a source of low toluene emissions, even at 23 °C. This is consistent with the data in Table 4 regarding the mean and median toluene emissions (the main monoaromatic hydrocarbon emitted from studied coffee-to-go cups). Moreover, the higher TVOC value at 80 °C than that at 40 °C might be caused by emissions from the PE coating, such as aliphatic hydrocarbons including dodecane, tridecane, tetradecane, and pentadecane (Mitchell et al., 2014a; Mitchell et al., 2014b).

To determine the possible interactions between the analysed chemical compounds in the plastic utensils, the Pearson correlation coefficients (PCCs) were determined. The PCCs between the measured emissions of the selected monoaromatic hydrocarbons and the TVOC values measured at two different temperatures are listed in Table 5. The PCC values calculated for the PS utensils indicate a strong or a very strong correlation between the emission of styrene and the other measured monoaromatic hydrocarbons, as well as the TVOC parameter,

Table 4
General parameters of measured monoaromatic hydrocarbons emitted from the studied samples of disposable plastic utensils not fully made of regular polymer material (natural or biodegradable material).

Parameter	Benzene 40 °C [ng·g^{-1}]	Benzene 80 °C [ng·g^{-1}]	Toluene 40 °C [ng·g^{-1}]	Toluene 80 °C [ng·g^{-1}]	Etylbenzene 40 °C [ng·g^{-1}]	Etylbenzene 80 °C [ng·g^{-1}]	p,m-Xylene 40 °C [ng·g^{-1}]	p,m-Xylene 80 °C [ng·g^{-1}]	Styrene 40 °C [ng·g^{-1}]	Styrene 80 °C [ng·g^{-1}]	TVOC 40 °C [μg·g^{-1}]	TVOC 80 °C [μg·g^{-1}]
Cups made from paper plastic laminates consist of high quality cellulose fibre with a thin internal PE coating (KB_CEL_PE)												
Mean	2.5	1.2	4.0	2.8	0.65	<LOD	1.9	0.99	4.8	3.7	0.134	0.43
Stan. dev.	2.7	1.4	2.0	3.0	0.53	–	2.9	0.81	3.6	2.6	0.028	0.36
Median	1.6	0.62	4.4	2.2	0.65	–	0.37	0.53	3.3	2.7	0.131	0.24
Min	0.23	0.26	0.45	0.14	0.11	–	0.11	0.33	1.8	1.1	0.096	0.12
Max	7.5	3.3	6.5	7.5	1.18	–	8.2	2.43	12.2	7.8	0.175	0.97
Transparent biodegradable polylactic acid straws (SL_PLA)												
Mean	0.66	10.1	5.4	229	41	3.7	<LOD	7.6	196	3.5	0.83	10.6
Stan. dev.	0.33	1.3	8.2	50	52	1.9	–	1.4	222	2.3	0.71	1.8
Median	0.65	9.9	1.7	204	11	3.1	–	7.3	122	2.6	0.42	10.2
Min	0.18	8.6	0.44	192	11	2.1	–	6.3	20	2.0	0.37	8.8
Max	1.04	12.4	17.6	313	101	6.8	–	10.4	446	8.0	2.07	13.0

Table 5
Pearson's correlation coefficients (PPCs) between emission rates of monoaromatic hydrocarbons in two different temperature levels for studied samples of disposable plastic barbecue and kitchen utensils (at a significance level of $p < 0.05$)*.

Disposable plastic utensils made of polystyrene

	Benzene	Toluene	Ethylbenzene	p,m-Xylene	Styrene	TVOC
Parametr [for 40 °C]						
Benzene	1	**0.59**	0.27	−0.45	**0.73**	0.48
Toluene		1	0.12	**−0.85**	**0.64**	0.29
Ethylbenzene			1	0.20	0.51	**0.74**
p,m-Xylene				1	0.014	0.017
Styrene					1	0.48
TVOC						1
Parametr [for 80 °C]						
Benzene	1	0.57	**0.93**	**0.73**	**0.94**	**0.95**
Toluene		1	0.50	0.29	**0.65**	0.56
Ethylbenzene			1	**0.83**	**0.97**	**0.99**
p,m-Xylene				1	**0.81**	**0.82**
Styrene					1	**0.99**
TVOC						1

Disposable plastic utensils made of polypropylene

	Benzene	Toluene	Ethylbenzene	p,m-Xylene	Styrene	TVOC
Parametr [for 40 °C]						
Benzene	1	0.44	**−0.61**	0.11	−0.45	0.11
Toluene		1	**−0.78**	0.16	**0.82**	**0.85**
Ethylbenzene			1	0.081	**0.91**	**−0.74**
p,m-Xylene				1	−0.051	0.22
Styrene					1	0.51
TVOC						1
Parametr [for 80 °C]						
Benzene	1	0.44	−0.056	**0.61**	−0.50	0.37
Toluene		1	**−0.81**	**−0.73**	−0.53	**−0.65**
Ethylbenzene			1	**0.94**	0.34	**0.88**
p,m-Xylene				1	0.29	**0.79**
Styrene					1	0.40
TVOC						1

Disposable plastic utensils not fully made of regular polymer material (natural or biodegradable polymer)

	Benzene	Toluene	Ethylbenzene	p,m-Xylene	Styrene	TVOC
Parametr [for 40 °C]						
Benzene	1	0.11	**−0.61**	0.033	−0.27	−0.26
Toluene		1	−0.22	0.48	−0.23	−0.47
Ethylbenzene			1	0.11	**0.70**	**0.98**
p,m-Xylene				1	0.52	0.23
Styrene					1	**0.76**
TVOC						1
Parametr [for 80 °C]						
Benzene	1	**0.94**	0.43	**0.93**	0.18	**0.92**
Toluene		1	**0.90**	**0.97**	−0.28	**0.96**
Ethylbenzene			1	**0.79**	−0.54	**0.68**
p,m-Xylene				1	−0.15	**0.96**
Styrene					1	0.071
TVOC						1

* only statistically significant correlation coefficients are marked by bold (> 0.60); Very weak correlation: 0.0 ÷ 0.2; weak correlation: 0.2 ÷ 0.4; medium correlation: 0.4 ÷ 0.6; strong correlation: 0.6 ÷ 0.8; very strong correlation: 0.8 ÷ 1.0.

at 80 °C. The strongest correlation was calculated between styrene, ethylbenzene, and the TVOC. This phenomenon is consistent with previously published studies that report that elevated temperatures cause significant styrene and ethylbenzene emissions, mainly as a monomer and solvent residues. Moreover, in the case of PS, the monoaromatic hydrocarbons have a significant impact on the TVOC and constitute a significant amount of the total organic content released into the gaseous phase from the PS materials. Furthermore, there was a strong correlation (0.64 and 0.65) between the emission of styrene and toluene from the studied PS disposable utensils at both 40 and 80 °C. This relationship might be associated with the fact that toluene is used as a solvent during the manufacturing and processing of polymers and, thus, could be present in and emitted from the studied samples. For all samples, the TVOC values were more strongly correlated with the monoaromatic hydrocarbons at 80 than 40 °C. The calculated PCC values for the disposable PP utensils confirm the previous findings that the monoaromatic hydrocarbon are not the main compounds affecting the final TVOC value. A similar relationship was observed for utensils made from natural or biodegradable material at 40 °C. For the PP samples, at both temperatures, increased emissions of ethylbenzene are associated with decreased emissions of toluene (inverse correlation). An inverse correlation between the toluene emissions and those of other monoaromatic hydrocarbons, as well as the TVOC values, was observed at 80 °C. Thus, toluene could be the main solvent used during manufacturing and processing instead of ethylbenzene or other organic solvents (aliphatic or aromatic compounds). For the samples comprising natural and biodegradable polymers, there was a strong or very strong correlation between the emissions of *p*, *m*-xylene and the emissions of benzene, toluene, and ethylbenzene at 80 °C. Moreover, at 80 °C, excluding the emissions of styrene, the emissions of monoaromatic hydrocarbons had a significant impact on the final TVOC value. Consequently, the emissions of monoaromatic hydrocarbons from natural and biodegradable utensils might be caused by the impurities in the final product, as well as the formation of monoaromatic hydrocarbons during the thermal treatment of the samples. Additionally, monoaromatic hydrocarbons could be contained within the natural and biodegradable polymer matrices, and only elevated temperature might cause their emission to the gaseous phases by weakening the bonds between the polymer matrix and the additives or residues.

4. Conclusions

In this study, the occurrence of key monoaromatic hydrocarbons, i.e. benzene, toluene, ethylbenzene, styrene, and *p*, *m*-xylene, was measured in nine types of single-use plastic barbecue and kitchen utensils made of regular and biodegradable polymers. Significant emissions of styrene, toluene, and ethylbenzene from the plastic samples were observed, especially at 80 °C. Thus, elevated temperatures (caused by e.g. hot water or hot meals) increase the emission potential of solvent residues from plastics and could affect the quality of consumed meals or beverages. In addition, as shown in Fig. 1, elevated temperatures might affect the mechanical structure of the used disposable plastic utensils. This mechanical effect might affect the emissions of solvent residues to the consumed foodstuff or beverage. The disposable utensils made of natural or biodegradable polymers were also sources of monoaromatic hydrocarbons, and the temperature of the meal or beverage could be a main factor affecting the studied biopolymer degradation processes. Cellulose (covered with a layer of PE) and PLA are considered to be more environmentally friendly materials for disposable utensils, but performed preliminary studies indicate that more detailed studies on the effects of these materials on food quality and taste are required. Especially, that the manufacturer of single-use PLA utensils do not put the information about the type of renewable resource from which the monomer was formed. It is possible to conclude that several contaminants present in employed renewable resource might be released form the final plastic product and affect the food flavour.

On the basis of research results, monoaromatic hydrocarbons in the polymer packaging could be transported directly to foods and beverages during preparation or consumption at elevated temperatures. These results contribute to knowledge about the migration of harmful chemicals that can affect the quality of food products and human health. This will increase societal awareness about the quality of these products and aid the estimation of which kind of single-use polymer utensils should be removed for everyday use in the near future, not only because of their environmental impact but also their possibly negative health effects when used to consume hot meals or beverages.

Concerning the most harmful xenobiotic compound emitted from the studied single-used barbecue and kitchen plastic utensils, benzene, this information should be considered as a preliminary data to stimulate further, more advanced investigations. It is crucial to define the main emission sources of this harmful compound properly; for example, whether it is the preparation process, which is mainly associated with the type and quality of applied reagents and other additives, or the polymer itself.

Declaration of competing interest

The author declares that there are no conflicts of interest.

The following is the supplementary data related to this article.Supplementary Fig. 1General scheme of the applied analytical procedure for the determination of monoaromatic hydrocarbon emissions from disposable plastic utensils.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.137485.

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