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Micellar and sub-micellar liquid chromatography of terephthalic acid contaminants using a C18 column coated with Tween 20

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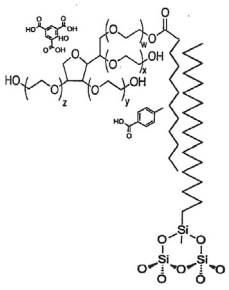
HIGHLIGHTS

- Positional aromatic isomers in industrial terephthalic acid samples separated.
- "Green" chromatography using Tween 20 surfactant coated column.
- Potential for mass spectrometry detection shown.

GRAPHICAL ABSTRACT



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ABSTRACT

The tremendous amounts of terephthalic acid (TPA) produced globally require consistent monitoring of its contaminants during the different stages of production for quality control purposes. In this paper, a simple, robust and green liquid chromatography method has been developed using an isocratic 100% aqueous mobile phase at pH 2 (dilute sulfuric acid) to separate TPA contaminants (mono-, di-, and tri-carboxylic aromatic acids) on a C18 stationary phase coated with Tween 20 (polyoxyethylene(20)sorbitan monolaurate). After optimization of all chromatographic conditions, near baseline separation of the nine carboxylic acids under investigation was achieved with a 2.5 mL/min flow rate on a 5 micron C18 silica column (100 x 4.6 mm) in under 20 min. The modified stationary phase showed an excellent capability to separate structural isomers in a reasonable time, markedly better that the bare C18 stationary phase. Plots of ln retention factor versus 1/temperature showed the expected linear relationship for the di- and tri-carboxylic aromatic acids (single retention mechanism likely) but a quadratic fit for the mono-carboxylic aromatic acids (dual retention mechanism likely). Due to the stability of the surfactant modified stationary phase, future potential mass spectrometry compatibility was shown through the alternative use of trifluoroacetic acid in the 100% H_2O (no Tween) mobile phase but still with UV detection. The developed method with 0.001% (vol/vol) Tween in the mobile phase was successfully used to analyze two different types of TPA industrial samples for all nine components plus revealing some other impurity peaks. The lowest limit of detection was 0.010 nmoles for o-phthalic acid and p-toluic acid (PTA), while the highest was 0.065 nmoles for 4-carboxybenzaldehyde (CBA). The concentrations of these important contaminants, PTA and CBA, in the mother liquor sample were 3348 mg/L and 1806 mg/L, respectively, while their respective concentrations in the purified TPA powder were 135 mg/kg and 17.7 mg/kg.

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Abbreviations

BA	benzoic acid
CBA	4-carboxybenzaldehyde
CMC	critical micelle concentration
GAC	"green" analytical chemistry
HMA	hemimellitic acid
IPA	isophthalic acid
LOD	limit of detection
LOQ	limit of quantification
MLC	micellar liquid chromatography
MP	mobile phase
MX	m-xylene
PET	polyethylene terephthalate
OPA	o-phthalic acid
OX	o-xylene
PTA	p-toluic acid
PX	p-xylene
SDS	sodium dodecyl sulfate
SP	stationary phase
TMA	trimellitic acid
TPA	terephthalic acid
TSA	trimesic acid

1. Introduction

The declaration of the pollution prevention act in 1990 by the Environmental Protection Agency in the United States persuaded analytical chemists to pursue more environmentally friendly methods to use in their research [1–3]. Twelve principles were stated by Anastas and Warner in 1998 that can work as guidelines to decrease the harmful impact of both chemical and industrial processes on the environment [4]. The green chemistry twelve principles were mainly intended for organic chemistry synthesis and the industrial processes. Not all of these principles can be applied in analytical chemistry to convert "regular" analytical chemistry into "green" analytical chemistry (GAC). Therefore, Namiesnik and co-workers suggested another twelve principles which were conveniently modified to fit the methodologies in analytical chemistry in 2013 [5]. Two of those principles, "multi-analyte methods are preferred" and "toxic reagents should be eliminated or replaced", are emphasized in this project.

One of the most ubiquitous separations and quantification techniques is liquid chromatography (LC) which can consume a significant amount of high purity organic solvents, generating costly amounts of toxic waste. If one operational high-performance liquid chromatograph generates one liter per day of mobile phase waste and knowing that some pharmaceutical companies have in hand more than 1000 LC instruments, this amount can build up to hundreds of thousands of gallons of toxic waste worldwide [6–10]. For the purpose of decreasing toxic wastes, GAC guidelines suggest that the most efficient steps to minimize the deleterious side effects of toxic wastes are reduction and/or replacement of the toxic organic solvents with more environmentally friendly solvents. The "greenest" solvent that can be used in LC is water. Micellar liquid chromatography (MLC) uses 100% aqueous solvents mixed with biodegradable surfactants; hence, it can also be considered as a "green" method of analysis [11–13].

Micellar liquid chromatography (MLC) was employed for the first time in 1979, way before the pollution prevention act and the formulation of the green chemistry principles. Because the micellar mobile phase (MP) has unique interactions with the solutes making it suitable for the separation of a wide range of analytes [14–17], the term "pseudophase liquid chromatography" was coined to distinguish it from other techniques that use surfactants but for different purposes [18,19]. The surfactants are amphiphilic compounds that have the ability to assemble in spherical configurations as micelles at or above a specific concentration called the critical micellar concentration (CMC). In aqueous solutions, the hydrophilic part of the micelle is on the surface of the micelle, while the hydrophobic region is in the core which can form a unique solvation media. Many studies have been published suggesting mathematical expressions that explain the processes and LC retention mechanisms depending on the type of surfactant used [20–24].

In general, the retention mechanism is mainly affected by three equilibrium states describing the distribution of the analyte among the mobile phase, the micelles, and the stationary phase as shown in the Supplementary Information (Fig. S1) [25]. However, another important act that must take place before the aforementioned distributions take place, is the adsorption of the surfactant onto the stationary phase which can be a very critical factor if the analysis is done below the CMC [14,20,26–28]. Due to the structural characteristics of the surfactant, the C18 stationary phase (SP) interacts with the hydrophobic part of the surfactant. The adsorption of the surfactant on the C18 SP can significantly change the following: a. Reduce markedly the pore volume and the available surface area. b. Increase polarity of the SP due to the coverage of the hydrophobic chain and putting the hydrophilic part of the surfactant on the surface of the SP. c. Alter functionality of the SP depending on the type of surfactant used, whether it was anionic, cationic, or neutral. d. Reduce the sample capacity of the column [20,26,29–33].

Some of the aforementioned points are considered as drawbacks especially the reduction in pore size, volume, and the capacity of the SP. However, micelles add additional selectivity and unique characteristics like the ability to solubilize different kinds of analytes including small charged molecules to the bulky hydrophobic molecules. Besides, due to the additional equilibrium between the three phases SP, MP, and micellar pseudophase, MLC has the ability to differentiate between closely related hydrophilic compounds, isomers, even enantiomers, of which RPLC is usually incapable of handling [34–37]. The successful separation of phthalic acid isomers by us provided excellent motivation to find a "greener" MLC method to separate and quantify terephthalic acid impurities in industrial samples using different surfactants [38].

Terephthalic acid (TPA) is used on a large scale to synthesize pesticides, fibers, and thermoplastics. However, the majority of the synthesized TPA is consumed in the preparation of polyethylene terephthalate (PET) polymer and other related polymers (polybutylene terephthalate and polytrimethylene terephthalate), these latter two on a smaller scale. These polymers are used in food packaging, soft drinks bottles, textiles, and many other applications. In 2019 the global production of TPA is estimated to be more than 81 million tons with an increase in global demand by 5% each year [39–43]. The starting material for TPA production is p-xylene

Table 1
List of nine carboxylic acids with corresponding abbreviations, structures, and dissociation constants.

	Acid Name	pKa1	pKa2	pKa3	Abbreviation
Mix 1	Trimellitic (1,2,4-COOH)	2.52	3.84	5.20	TMA
	Hemimellitic (1,2,3-COOH)	2.80	4.20	5.87	HMA
	Trimesic (1,3,5-COOH)	3.12	3.89	4.70	TSA
Mix 2	o-Phthalic (1,2-COOH)	2.76	5.28	–	OPA
	Isophthalic (1,3-COOH)	3.70	4.60	–	IPA
	Terephthalic (1,4-COOH)	3.51	4.46	–	TPA
Mix 3	4-Carboxybenzaldehyde (1-COOH, 4-CHO)	3.78	–	–	CBA
	Benzoic (1-COOH)	4.17	–	–	BA
	p-Toluic (1-COOH, 4-CH_3)	4.36	–	–	PTA

(PX) which is produced industrially from crude oil. However, less than 6% of the synthesized PET is made from bio-based TPA based on a plant-based PX starting material [43,44].

The purity of PX can directly affect the purity of the produced TPA. PX is converted to TPA by oxidation with air in the presence of hydrobromic acid, manganese(II) acetate, and cobalt(II) acetate in acetic acid as a solvent. This oxidation process is done via a free radical mechanism, which is known as the AMOCO process [45,46]. Many factors affect the purity of the final TPA product like the purity of the PX starting material. All of these contaminants (Table 1), can originate from the presence of o-xylene (OX), m-xylene (MX), and the trimethylated benzene impurities in PX, as shown in Fig. S2 in the Supplementary Information, and described briefly below.

According to previous thermodynamic studies, PX and the traces of OX and MX are easily oxidized during the first step of oxidation. However, once p-toluic acid (PTA) is formed, the oxidation process becomes slower due to the presence of the electron-withdrawing (COOH) group [47–53]. PTA is then oxidized to 4-carboxybenzaldehyde (CBA) which can ultimately oxidize to TPA. The traces of OX and MX can be readily oxidized to o-phthalic acid (OPA) and isophthalic acid (IPA), respectively. Due to the free radical oxidation nature and the excessive oxidizing agents used in the reaction mixture, TPA, OPA, and IPA can be oxidized to benzoic acid (BA) by losing the CO_2 group [45,48,53,54]. The traces of 1,2,3-trimethyl benzene, 1,2,4-trimethyl benzene, and 1,3,5-trimethyl benzene are oxidized to hemimellitic acid (HMA), trimellitic acid (TMA), and trimesic acid (TSA), respectively. The impurities in the TPA can drastically reduce the quality of the produced PET. The presence of CBA can decrease the polymerization reaction rate and therefore the molecular weight of the produced PET leading to the formation of undesirable yellow colored and weaker polymer. Other contaminants like PTA and BA can terminate the polymerization of TPA leading to the fluctuation in the molecular weight of the produced PET. Structural deformation can occur during the polymerization process due to the presence of OPA, IPA, TMA, HMA, and TSA. Hence, it is of great importance to minimize the concentration of these contaminants in the final product [55–58] and monitoring the concentration of the aforementioned contaminants is essential for quality control.

There is a small collection of published literature that deals with the determination of TPA contaminants in real samples using different techniques. Capillary electrophoresis was done by Wu and co-workers [59] to analyze TPA, and some of the possible contaminants. Their method focused on CBA, PTA, and other substituted benzoate species such as salicylate, as well as isomers of phthalate. The major drawbacks of this and other similar methods are the equilibration time of the capillary can be long, and at least three significant contaminants (TMA, HMA, TSA) were not considered [59,60]. Another study was done by Huang and co-workers [55] to determine TPA and eight other carboxylic acids impurities in industrial mother liquor TPA synthesis samples using micro-emulsion electrokinetic chromatography. This method has two significant drawbacks; the first is using solvents like octane and cyclohexanol and the second is the sophisticated gradient elution program require to achieve a full separation in about 20 min. RPLC has also been used to determine the nine carboxylic acids by Yuan and co-workers [61]. However, the lengthy analysis time, the use of non-environmentally friendly solvents, and the three step gradient elution program were the main disadvantages of this and other similar methods [61–63]. Previously, we have separated TMA, HMA, TSA, OPA, IPA, TPA, CBA, BA, and PTA by MLC using a SDS surfactant. The concentration of SDS in the mobile phase has to stay well above the CMC which makes it incompatible with mass spectrometry. A step flowrate gradient was used to reduce the overall analysis time under 20 min, however, compatibility with LC-MS remains an issue [12].

Tween 20 is a nonionic amphiphilic sorbitan ester of lauric acid with a total of twenty oxyethylene subunits connected to the sorbitol. It has a wide application in food industry, cosmetics, and biochemical applications as an emulsifying agent. Its amphiphilic character is amenable for forming stable oil-water emulsions. The biochemical applications include removing of proteins in cell membranes, lysing mammalian cells, and as a blocking agent in membrane based immunoassays [64,65].

There are only a few papers reporting the addition of Tween 20 to the run buffer as a modifier/separation enhancer in capillary electrophoresis [66–68] or to the mobile phase for LC [69,70]. In this paper [69], the authors were investigating the influence of adding different mixtures of surfactants including Tween 20 to the aqueous/methanolic MP on the retention factor of specific analytes separated by silica gel liquid chromatography. Silica gel SP was coated with the cationic surfactant cetyltrimethylammonium (CTAB) to increase the retention of the negatively charged analytes. Moreover, Tween 20 was added to the MP which will interact with the CTAB coated SP. The presence of Tween 20 will add hydrophobic-hydrophobic interaction to the coated silica gel, which will enhance the retention of neutral molecules such as fluorenone, naphthalene anthracene, and pyrene. Tween 20 also has been reported as an additive in reversed-phase LC. The adsorption of Tween 20 by the SP (Fig. 1) will add hydrophilic character and semi permeability to the hydrophobic SP and prevent proteins and peptides from being adsorbed into the SP [70]. To our knowledge and others [37], coating the SP with only Tween 20 and using a 100% aqueous MP, with or without Tween 20, to separate and analyze isomers and structurally similar compounds in a mixture has never been reported.

In this research, we are presenting an environmentally friendly method that uses Tween 20 as a surfactant to coat the C18 HPLC column, permitting the use of a totally aqueous mobile phase. The isocratic micellar conditions permit qualitative and quantitative analysis of the TPA contaminants in two different types of industrial

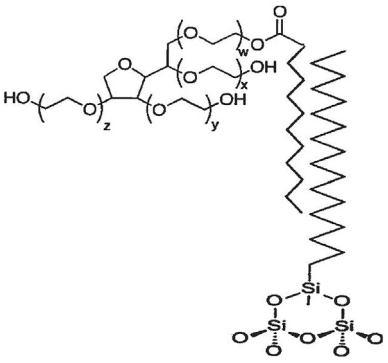

Fig. 1. The structure of Tween 20 [polyoxyethylene(20)sorbitan monolaurate, w + x + y + z = 20] and its proposed interaction with the C18 stationary phase.

samples, mother liquor of the TPA oxidation mixture, and the final purified TPA powder. Optimization of chromatographic conditions such as temperature, Tween 20 concentration, and sulfuric acid concentration was done to achieve baseline resolution in the shortest time. Quadratic trend line fits instead of linear ones of ln k' versus 1/T (van't Hoff plots) showed three of the analytes (monocarboxylic aromatic acids) were likely undergoing a mixed retention mechanism. A mass spectrometry (MS) compatibility study was done by reducing the level of Tween 20 in the MP to zero% and bringing the pH to 2.00 using trifluoroacetic acid instead of sulfuric acid. Excellent stability of Tween 20 on the modified C18 stationary phase was established ensuring that this method could be suitable for MS detection.

2. Materials and methods

2.1. Chemicals and samples

All standards were prepared from analytical grade chemicals. All solutions were made with 18.2 MΩ distilled and deionized water, treated with UV light and purified using a Milli-Q (Millipore, Bedford, MA, USA) water purification system. All sulfuric acid solutions for pH mobile phase adjustment were prepared using 98% sulfuric acid, purchased from Fisher Scientific (Pittsburgh, PA, USA). Sulfuric acid has some buffering capacity at pH 2 due to the pKa is 1.92. Benzoic acid (BA,>99%) and potassium hydrogen phthalate (OPA, 99.99%) were purchased from Fisher Scientific (Fair Lawn, NJ, USA). Isophthalic (IPA, 99%), terephthalic acid (TPA, 98%), trimellitic acid (TMA, >99%), hemimellitic acid (HMA, 98%), trimesic acid (TSA, 95%), 4-carboxybenzaldehyde (CBA, 97%), and p-toluic acid (PTA, 98%) purity were procured from Fisher Scientific (Pittsburgh, PA, USA).

TPA and eight of its most important impurities are listed in Table 1 with corresponding abbreviations, positional functional groups, and acid dissociation constants. Tween 20 solutions of different percentages were prepared from standard Tween 20 molecular biology grade liquid purchased from Sigma-Aldrich Co (St. Louis, MO, USA). The single-component acid solutions of TMA, HMA, TSA, OPA, IPA, CBA, BA, and PTA solutions were made using ten mg of the pure acid dissolved in 100 mL of 75% methanol-25% H_2O. TPA, FBA, and PTA were sonicated for 20 min to ensure their solubility. Sample simulation was done by mixing all nine carboxylic acids, 10 mg of each, in 100 mL of 75% methanol-25% H_2O; this solution was sonicated for 30 min. The mother liquor sample was provided by a Midwest TPA manufacturer, courtesy of R. E. Pauls. One mL of mother liquor solution was filtered through a 0.22 μm nylon membrane filter and then put in 100 mL volumetric flask which was brought to the mark with 75% methanol-25% H_2O. A purified TPA powder sample was kindly provided from a different Northeast company, courtesy of H. B. Sunkara. Twenty grams of this powder was placed in 500 mL of 100% HPLC grade methanol. This solution was sonicated for 30 min and then evaporated under low pressure and a 30 °C temperature to reach a volume of 50 mL. Then 10 mL of H_2O was added to bring the total volume to 60 mL which re-precipitated TPA.

2.2. Apparatus and chromatographic conditions

All chromatographic calculations as well as chromatograms, and some statistical parameters were generated using Chromeleon 7.2.9 software (Thermo Scientific, Sunnyvale, CA, USA). This same software controls the Thermo Scientific UltiMate 3000 (Thermo Scientific, Sunnyvale, CA, USA) which was equipped with a DGP 3600RS pump, online degasser, WPS 3000RS autosampler, TCC 3x00RS temperature controlled column oven, and an Ultimate multiwavelength 3000RS detector.

The optimization of the chromatographic conditions and all separations were done using a C18 Supelco Discovery column (Sigma-Aldrich, St. Louis, MO, USA) having dimensions of 100 mm × 4.0 mm, with 5 μm particle diameter. The end-capped spherical silica particles have a pore size of 180 A°, a surface area of 200 $m^2\ g^{-1}$, and carbon load of 12%. The manufacturer literature indicates the Discovery C18 column is stable from pH 2.0 to 8.0 and is recommended for resolution of geometrical isomers (example given ephedrine and pseudoephedrine) and other structurally closely related compounds. This column was coated with 1% vol/vol Tween 20 solution for 3 h at flow rate of 0.1 mL/min to ensure saturation of the stationary phase with the surfactant. The coated column was washed with a 0.002% (vol/vol) Tween 20 in pH = 2.2 solution for 15 min before starting the optimization of the chromatographic conditions. The quantification of contaminants in the two industrial samples was done using the same column. The mobile phase, composed of 0.001% (vol/vol) Tween 20 in H_2O acidified to pH = 2.2 with sulfuric acid at 15 °C, was propelled at 1.8 mL/min and single wavelength detection was set at 240 nm. Fig. S3 in the Supplementary Information shows UV spectra of the mobile phases indicating why the background UV-VIS absorbance of Tween 20 was not an issue at 240 nm or even 215 nm.

3. Results and discussion

Initially, because these aromatic acids are relatively hydrophilic, a 100% aqueous mobile phase using the Discovery C18 column uncoated with surfactant was tried. Chromatograms with the mobile phase adjusted to both pH 2.2 and 6.3 are shown in Fig. 2. At the higher pH at which all the aromatic acids are ionized, all the compounds except the most hydrophobic (BA and PTA) were unretained. At the lower pH of 2.2 at which the acids were primarily neutral in charge, separation of TMA, TSA, HMA, and IPA was good. Significant peak overlap was noted for compound pairs TPA, OPA and CBA, BA. However, the analysis time of about 80 min was excessive and the peak corresponding to PTA could not be positively identified. Reduction of the hydrophobic retention plus some potential for hydrogen bonding should be possible with a Tween coated C18 column.

3.1. Optimization of Tween 20 percentage

The effect of changing Tween 20 percentage in the mobile phase (MP) on the retention factor (k') was thoroughly investigated (Fig. 3). The Tween 20 concentration was changed from 0.008% (vol/vol) which is above the CMC (60 mg/L or considering the specific

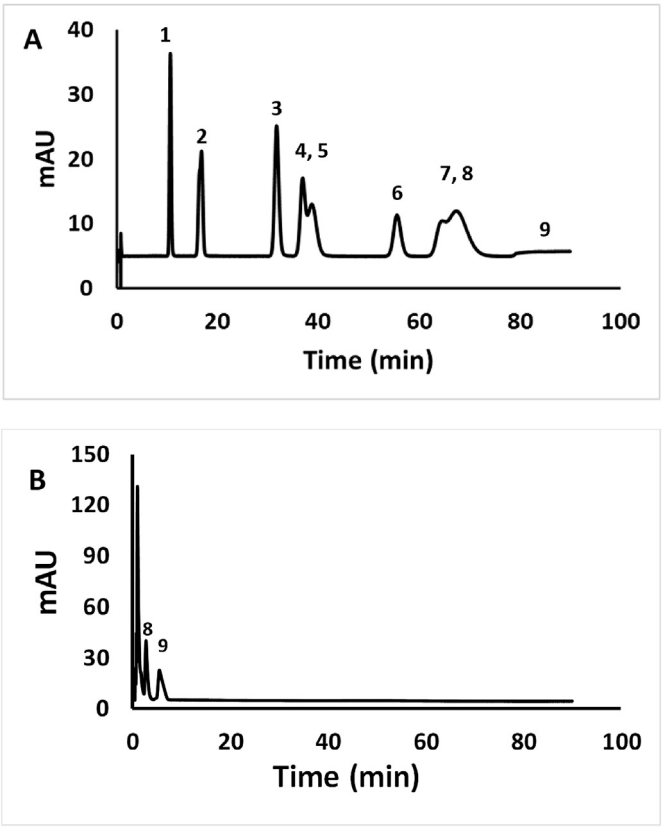

Fig. 2. A) Chromatogram of a mixture of nine acids standard using uncoated column with 100% aqueous MP at pH = 2.2, **B)** Chromatogram of the mixture of nine acids standard using uncoated column with 100% aqueous MP at pH = 6.3. Peak assignments are as follows: 1-TMA, 2-TSA, 3- HMA, 4-TPA, 5-OPA, 6-IPA, 7-CBA, 8-BA, 9-PTA. The flow rate was 1.5 mL/min; the temperature was 20 °C.

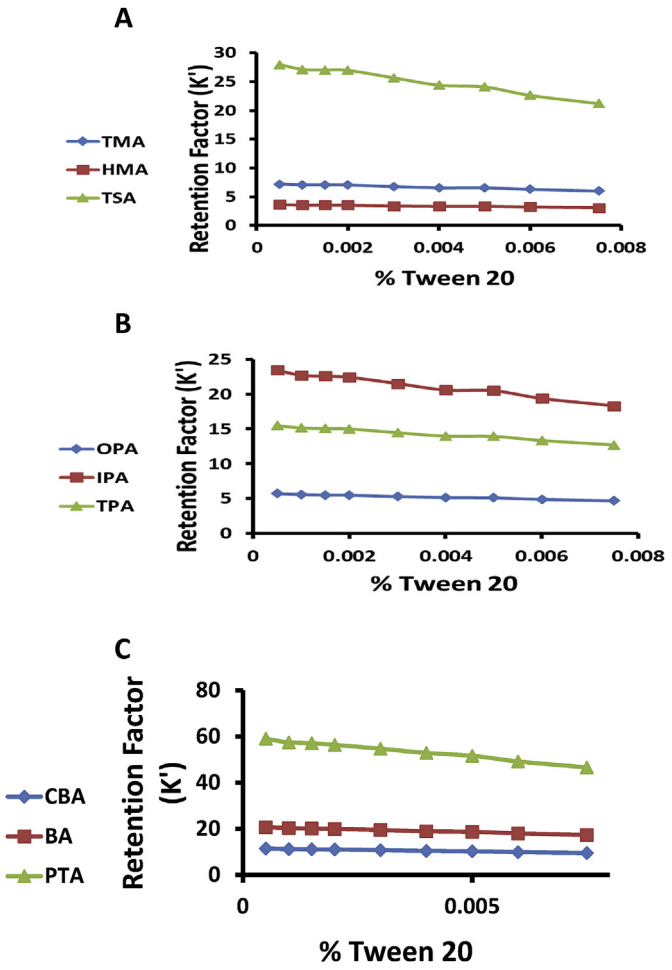

Fig. 3. The effect of changing Tween 20 percentage in the mobile phase on the retention factor: **A)** Mixture 1; **B)** Mixture 2; **C)** Mixture 3.

gravity of 1.1, 0.00545% (vol/vol)) to 0.0005% (vol/vol) which is 10 times below the CMC. The first impression of the k' trends for mixture 1 (Fig. 3A), mixture 2 (Fig. 3B), and mixture 3 (Fig. 3C) is as the percentage of the Tween 20 increases, the retention factor decreases. However, the change in Tween 20 percentage in the MP does not drastically change the retention factor, even at percentages above the CMC, corresponding to the three right most points. Apparently the Tween 20 coated column is the major factor for the reduction of the analyte retention as compared to Fig. 2. For the tricarboxylic acids, the retention order was TMA < TSA < HMA on the uncoated C18 column (Fig. 2) while that for the surfactant coated column was HMA < TMA < TSA (Fig. 3). These trends can be explained by the carboxylic acid positioning on the aromatic ring; HMA being the 1,2,3 isomer presents a larger hydrophobic surface to the C18 stationary phase. HMA is the 1,2,3-tricarboxyl aromatic acid and at pH = 2.2 the neutral form of this acid is dominant. It is still the least retained compound due to the fact that the structure shows three carboxylic acid groups on one side which reduces the overall area of the molecule that will participate in hydrogen bonding. Another important factor is the intramolecular hydrogen bonding within HMA itself (Fig. S4 in the Supplementary Information) which greatly decreases the hydrogen bonding between HMA and the ethylene oxide units in Tween 20. TSA is 1,3,5-tricarboxyl aromatic acid with the three carboxylic groups evenly distributed around the benzene ring and with no intramolecular hydrogen bonding, which makes hydrogen bonding interaction between TSA and ethylene oxide units of Tween 20 more effective. Moreover, the TSA molecule has higher symmetry elements than HMA and TMA (Fig. S2); higher symmetry elements can cause a reduction in the dipole moment [71].

A similar argument is not as strong for the dicarboxylic acids (mixture 2) where the retention order on the uncoated column is TPA < OPA < IPA (Fig. 2); OPA would be expected to be retained the longest. For the surfactant coated column (Fig. 3B), the ortho-carboxylic substituted OPA elutes first, followed by TPA, with IPA the most retained compound; TPA would be expected to be retained the longest.

Mixture 3 does not include structural isomers; however, it includes CBA and PTA which are two of the most important TPA contaminants. BA is especially important due to its presence in very high concentration in industrial mother liquor samples. Fig. 3C shows that CBA has the lowest retention factor among the three compounds, followed by BA. PTA has the highest retention factor among all of the nine components under investigation due to its high hydrophobicity due to the methyl group. The retention order is directly proportional to the hydrophobicity of these molecules; it is the same on both surfactant coated and uncoated C18 column (Fig. 2). However, as will be seen later, through the van't Hoff plots (ln k' vs. 1/Temperature), a mixed retention mechanism is likely for these three analytes.

3.2. Optimization of temperature

The temperature was varied from 15C to 40C and the Tween 20

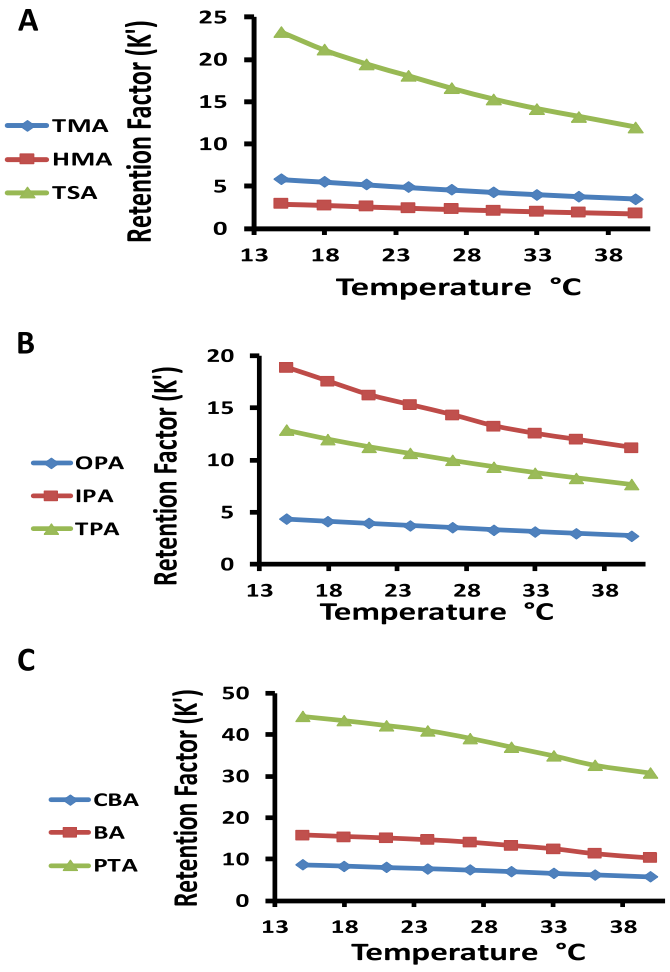

Fig. 4. The effect of changing temperature on the retention factor (k') for mixtures 1, 2, and 3 (top to bottom).

percentage was kept constant at 0.001% (vol/vol) at pH = 2.2 to ensure a reproducible coated C18 column. The retention factor of TSA in mixture 1 was significantly affected by increasing the temperature (Fig. 4A). This can be attributed to the fact that increasing the temperature can destabilize intermolecular hydrogen bonding responsible for TSA retention on the Tween 20 modified SP. HMA and TMA retention factors decreased with increasing temperature. However, this decrement was insignificant compared to the TSA retention factor decrement. The baseline separation of HMA, TMA, and TSA was achievable at all temperatures. The relationship between the retention factor of mixture 2 and changing the temperature is shown in Fig. 4B. The retention factor was inversely proportional to the temperature increment. The resolution and baseline separation among OPA, TPA, and IPA was almost unchanged throughout the entire range of temperatures under investigation. Fig. 4C shows the relationship between the retention factor and the change in temperature for mixture 3. The most significant retention factor change was in PTA; CBA and BA are much less affected by increasing temperature.

The linear relationship of ln retention factor (k') versus 1/Temperature (T) where the intercept "a" is a measure of the entropy and the slope "b" is a measure of the enthalpy is considered valid for an analyte that is undergoing retention through one mechanism [72]. Based on the trends in Fig. 3, data for the intercept and slope are tabulated in Table 2 for the di- and tri-carboxylic aromatic acids. Correlation coefficients of 0.997 or better were found for all six analytes. The slope values as expected increased with retention which was likely dominated by one mechanism, hydrogen bonding with the Tween coated stationary phase. The enthalpy value for TSA was considerably larger than that for the other analytes in Table 2 while that for OPA was the smallest. In contrast, such plots for CBA, BA, and PTA were nonlinear and a quadratic fit of the data in the form of the equation $\ln k' = a + b(1/T) + c(1/T)^2$ was required as shown in Fig. 5. Because the pH of the mobile phase was 2.2, all three of these mono-carboxylic aromatic acids would be in their one unionized form and therefore a combination of an ionized and neutral analyte causing mixed retention cannot explain the nonlinearity of ln k' with 1/T. Assuming the phase ratio remains constant [73], such plots are an indication of a mixed retention mode by the neutral analyte, in this study reversed phase with the C18 groups and hydrogen bonding with Tween. The possibility of such a mixed retention mechanism was also postulated in an early study [74] involving the separation of indole derivatives on a C18 column using a Tween 20 mobile phase. By taking the derivative of the quadratic equation and setting it to zero, the temperature at the extreme (maximum in this case) value of k' can be determined as T = -2c/b. These corresponding T values when multiplied by 1000 for CBA, BA, PTA are 274, 286, and 281 K, respectively. All our experimental temperatures were above these cold temperatures indicating the retention process was enthalpy driven [72].

3.3. Optimization of sulfuric acid concentration

Sulfuric acid was used to suppress the formation of the conjugate analyte base to ensure reversed phase retention. Fig. 6A, B, and 6C represent the relation between the retention factor versus sulfuric acid concentration for respectively mixtures 1,2, and 3 using 0.001% (vol/vol) Tween 20 in the MP at pH = 2.2 and 15 °C. The sulfuric acid concentration was changed from 1 mM to 7.5 mM. For the three mixtures; the change in the concentration of sulfuric acid was not significantly affecting the retention factor. However, using a MP in which sulfuric acid concentration is less than 1 mM will lead to total loss in resolution and peak splitting due to the presence of the acid and the conjugate base, especially for mixture 1 and mixture 2. Increasing the concentration of sulfuric acid leads to a slight increment in the retention factor. In addition to the ion suppression mechanism, this can also be attributed the increment of the ionic strength in the MP which pushes the neutral analytes to interact more with the neutral SP.

Typically, C18 stationary phase chains collapse under 100% aqueous MPs. However, the C18 SP still strongly retains the hydrophobic analytes like CBA and PTA (Fig. 2). Coating the C18 SP with Tween 20 surfactant will protect the C18 chains from collapsing and drastically decrease its hydrophobicity as previously mentioned in the text and shown in Fig. 3. Therefore, the retention of the hydrophobic analytes like CBA and PTA will be reasonably short. Applying the optimum separation conditions, Fig. 7A shows a chromatogram of the nine carboxylic acids using Tween 20 coated

Table 2
Coefficients a and b with correlation coefficient (R^2) for plots based on the van't Hoff equation: $\ln k' = a + b(1/T)$.

Compound	a	b	R^2
HMA	−5.417	1871	0.9993
TPA	−3.954	1876	0.9998
TSA	−5.136	2384	0.9997
OPA	−4.336	1677	0.9970
TPA	−3.954	1876	0.9998
IPA	−3.722	1917	0.9971

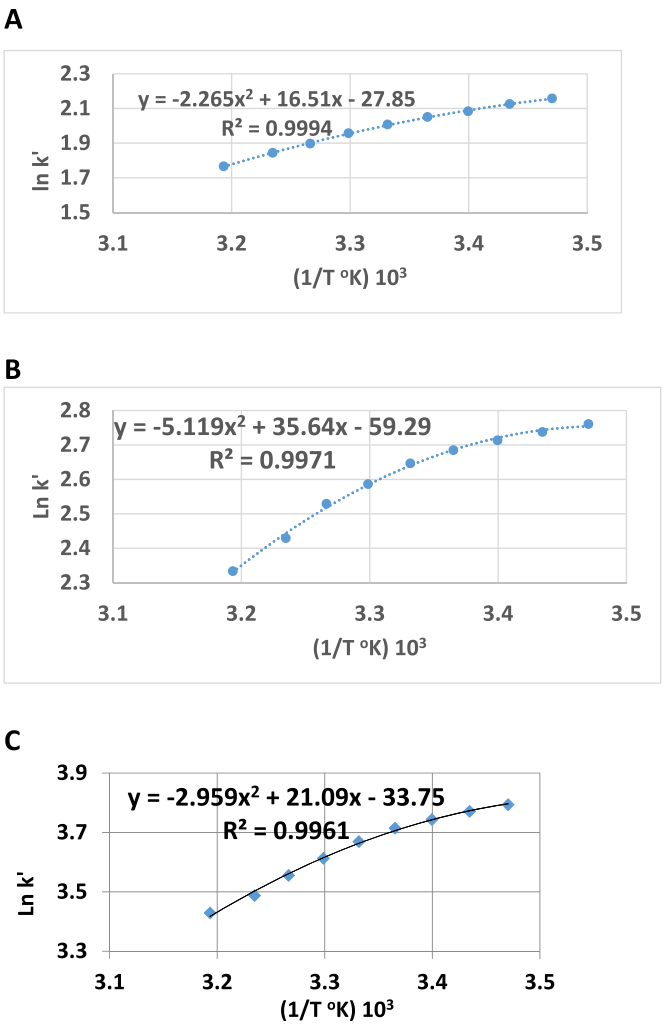

Fig. 5. Plots of ln retention factor (k') versus (1/T °K) 10^3 for **A)** CBA; **B)** BA; **C)** PTA.

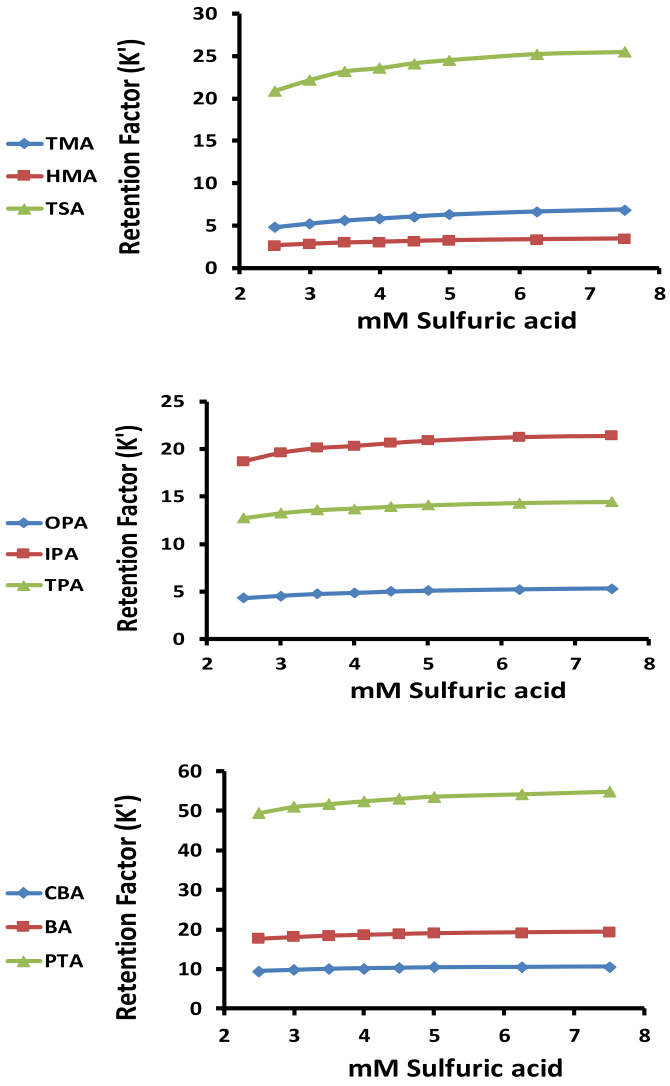

Fig. 6. The effect of changing the concentration of sulfuric acid on the retention factor (k') for mixtures 1, 2, and 3 (top to bottom).

column with 80 ppm of the surfactant in the mobile phase. Fig. 7B shows the baseline separation all analytes using the coated column but only the acidified mobile phase with no Tween at a faster flowrate. Resolution of peaks 6 and 7 is improved.

3.4. Mass spectrometry compatibility

One of the many objectives of this project is to be able to run a 100% aqueous MP on the modified SP to convert the UV-VIS detection compatible sub-micellar method into a mass spectrometry compatible method. Fig. 8A shows a chromatogram of the baseline separation of the nine carboxylic acids using trifluoroacetic acid instead of sulfuric acid in the 100% water mobile phase. As expected, it is very similar to the chromatogram in Fig. 7B. To investigate the stability of the coated SP and reproducibility, we injected the simulated sample that contains all the nine contaminants under investigation ten times separated by a time period of 1 h in between injections using the chromatographic conditions in Fig. 8A. Fig. 8B shows that the retention factor profiles versus the number of injections of all analytes present in the mixture over a time period of 10 h. The constant retention factor trend indicates the stability of the Tween 20 on the C18 SP (as also indicated in [74]) and that there is future potential that this method can be made MS compatible, similar to that previously reported [75].

3.5. Quantitative analysis of industrial samples

Quantitative analysis was done using standard solutions of the nine carboxylic acids to generate six point calibration curves to be used to measure these contaminants in two industrial samples. All measurements were done in triplicate and the R^2 correlation coefficients were 0.9999. Fig. 9A shows the standard chromatogram of the nine compounds under investigation. Table 3 lists all calibration curves regression parameters including the limit of detection (LOD) and the limit of quantification (LOQ). LOD values ranged from about 0.01 to 0.08 nmol (1–10 ppm). The chromatogram for the mother liquor from oxidative TPA synthesis and the chromatogram for the final purified TPA powder are shown in Fig. 9B and C. Retention reproducibility for all 9 peaks in the two sample chromatograms generally ranged from 0.1 to 2% RSD (n = 3) with an average value of 0.67%. The benzoic acid peak dominates the mother liquor chromatogram and TPA dominates the purified powder chromatogram, as expected. Both sample chromatograms indicate trace amounts of unknown contaminants that were not identified. Despite differences in relative peak intensities in the different samples, all nine acids were detected in quantifiable amounts except for OPA and TSA in the purified TPA powder sample (Table 3).

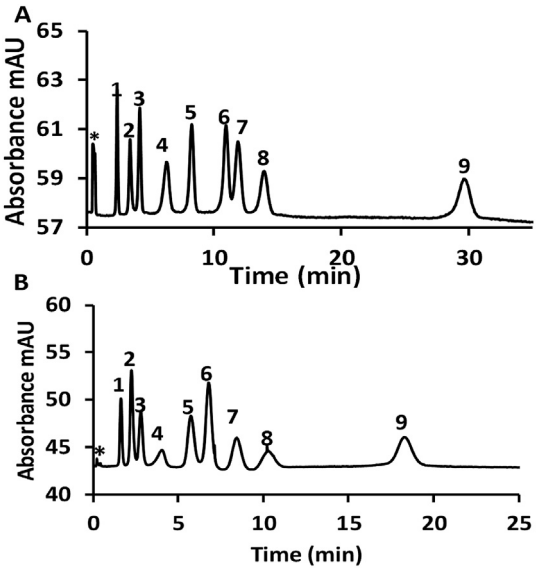

Fig. 7. Chromatograms of the nine carboxylic acids on the Tween coated column: **A)** Mobile phase was 0.008% Tween 20 (above CMC) acidified to pH = 2.2 using sulfuric acid at 15 °C at a flow rate of 1.5 mL/min. **B)** Mobile phase was 100% water acidified to pH = 2.2 using sulfuric acid at 15 °C at a flow rate of 2.5 mL/min. Peak assignments for both chromatograms are as follows: 1-HMA, 2-OPA, 3-TMA, 4-CBA, 5-TPA, 6-BA, 7-IPA, 8-TSA, 9-PTA, *unretained components.

Benzoic acid in the mother liquor sample has the highest concentration among the contaminants, attributed to the excessive oxidation of PX (and other contaminants) to benzoic acid after decarboxylation takes place. The TPA has a low concentration (291.8 ppm) because of its low solubility. The most important contaminant concentrations for assay of TPA quality were 3348 ppm for PTA and 1806 ppm for CBA. TSA has the lowest concentration among the carboxylic acids under investigation. The

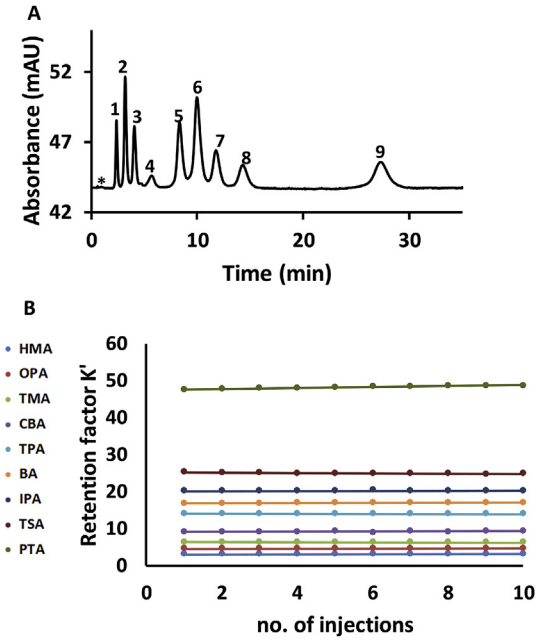

Fig. 8. A) Baseline separation of all nine carboxylic acids (peak assignment same as in Fig. 7B) using MS compatible conditions of 100% H_2O acidified with trifluoroacetic acid to pH 2.2 with a flow rate was 1.5 mL/min and column temperature of 15 °C. **B)** The retention factor versus the number of injections using the 100% aqueous MP acidified with trifluoroacetic acid.

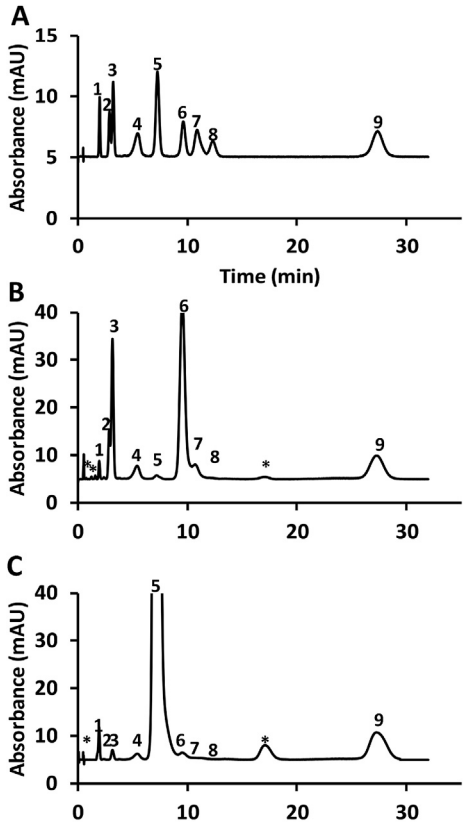

Fig. 9. A) Three chromatograms of a mixture of nine acids standard, **B)** Mother liquor unknown, and **C)** An extract of purified TPA powder. Peak assignments are as follows: 1-HMA, 2-OPA, 3-TMA, 4-CBA, 5-TPA, 6-BA, 7-IPA, 8-TSA, 9-PTA, * unknown. See end of section 2.2 for chromatographic conditions.

analysis of the purified TPA powder showed that the highest contaminant concentration was PTA; this can be explained by the last step of TPA synthesis, which was the reduction of CBA to PTA. However, the final TPA product still has many impurities, including

Table 3
Analytical figures of merits and regression parameters of the nine calibration curves and the concentration of the measurable contaminants in the mother liquor of TPA synthesis and the purified TPA powder.

Compound	A[a]	B[b]	LOD[c]	LOQ[d]	mg/L[e]	mg/Kg[f]
HMA	0.0097	2.126	0.0112 (2.35)	0.0373 (7.835)	1009	60.7
OPA	−0.005	1.979	0.01 (1.66)	0.0333 (5.533)	3248	Trace
TMA	−0.075	4.48	0.0279 (5.859)	0.0929 (19.53)	6243	16.8
CBA	−0.097	3.157	0.0647 (9.709)	0.2156 (32.36)	1806	17.7
TPA	−0.187	7.754	0.0233 (3.873)	0.0777 (12.91)	291.8	3472
BA	−0.114	2.928	0.0265 (3.234)	0.0883 (10.78)	18030	32.0
IPA	−0.147	4.504	0.0188 (3.127)	0.0627 (10.42)	1618	7.86
TSA	−0.083	3.136	0.0483 (10.16)	0.1611 (33.85)	191	Trace
PTA	−0.198	6.038	0.0102 (1.388)	0.034 (4.626)	3348	134.5

[a] y-Intercept (nanomole).
[b] Slope from the linear regression equation based on nanomoles.
[c] Limit of detection values are given in nanomoles (and in ppm based on 1 μL injection volume). Their values were calculated using the following equation: LOD= (3*σ)/B where σ is the standard deviation of the y-intercept.
[d] Limit of quantitation values are given in nanomoles (and in ppm based on 1 μL injection volume). Their values were calculated using the following equation: LOD= (10*σ)/B where σ is the standard deviation of the y-intercept.
[e] The concentration of the contaminants in the mother liquor sample. Percent relative standard deviation (%RSD) (n = 3) ranged from 0.3 to 6% with an average value of 1.7%.
[f] The concentration of the contaminants in the purified TPA powder. %RSD (n = 3) ranged from 0.1 to 6% (average 3%) with the exception of BA which was 16%.

CBA, but their concentrations are low enough to be acceptable for industrial use. An estimate of accuracy can be learned from a ratio comparison of these mother liquor data to those reported previously in our previous study [12]. In that study, the sample was from a different lot but ratios on the order of 1–1.8 were found for TMA, BA, PTA, OPA, and TSA. Ratios for HMA and CBA were greater than 3 and that for TPA was 0.3. Recently, we have found that these aromatic acids can be separated using a gradient by reversed phase HPLC. A ratio comparison of the Tween study to this reversed phase one using samples taken from the same lots showed mother liquor mg/L ratios ranging from 0.8 to 1.2 for IPA, PTA, OPA, TPA, TMA, and TSA; the other three acid ratios were outside this range. Because the ppm values of the aromatic acid impurities were less in the purified powder sample and some were considered trace, the ratio comparison between Tween and reversed phase for those measureable impurities is between 0.9 and 1.5 for PTA, TMA, CBA, IPA, and BA. In general, it seems that future attention needs to be addressed to more reproducible sampling.

4. Conclusions

An environmentally friendly separation method has been developed using Tween 20 as C18 stationary phase modifier that requires no toxic solvents in the mobile phase. Optimization for chromatographic conditions has been done to ensure a baseline separation of all TPA contaminants under 20 min using isocratic conditions and single wavelength detection at 240 nm. Linear relationships of ln k' versus 1/T (van't Hoff plots) were found for the di- and tri-carboxylic aromatic acids however such plots for the mono-carboxylic aromatic acids (CBA, BA, and PTA) showed quadratic trendlines indicating a likely dual retention mechanism. Excellent stability of the modified stationary phase have been shown with a 100% aqueous MP acidified with trifluoroacetic acid showing potential future compatible application to mass spectrometry. Nine calibration curves have been generated and the regression parameters including the limits of detections (LOD) and the limits of quantifications (LOQ) have been determined. The optimized method was used to determine the concentration of expected TPA impurities in two different types of industrial samples and even a few unknown contaminants were noted in the purified powder sample.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Abd al-karim F. Ali: Methodology, Validation, Formal analysis, Investigation, Writing - original draft, Visualization. **Neil D. Danielson:** Conceptualization, Formal analysis, Resources, Writing - review & editing, Supervision.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aca.2020.01.036.

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