Separation and Determination of Carbonyl Compounds in Indoor Air Using Two-Step Gradient Capillary Electrochromatography

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A method of two-step gradient capillary electrochromatography (CEC) was developed to measure 12 carbonyls (aldehydes and ketones) in indoor air samples. The carbonyls were derivatized with 2,4-dinitrophenylhydrazine (DNPH) and then separated by a two-step gradient CEC on a C8 column. Effects of various instrumental conditions on the separation, including buffer concentration, organic modifiers, voltage, and cassette temperature, were investigated. The method detection limits for the 12 carbonyls ranged from 0.2 µg to 1.6 µg per sample and the recoveries were generally between 90 and 120%. A subset of 30 indoor air samples containing formaldehyde and acetaldehyde from 75 randomly selected homes in the city of Ottawa, Canada were measured using the CEC method. The concentrations of formaldehyde and acetaldehyde in these indoor air samples ranged from 5.8 µg/m³ to 85 µg/m³, and from 4.4 µg/m³ to 38 µg/m³, respectively. The comparison between CEC and the traditional HPLC method shows a good agreement in measured values.

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Introduction

Carbonyls (a common term for aldehydes and ketones) are reactive volatile substances. They are of concern to the public due to their potential adverse health effects and environmental prevalence. For example, formaldehyde and acetaldehyde are potent sensory irritants and are classified as probable human carcinogens.^{1,2} They are present in indoor air with likely sources including cigarette smoking, building materials, furniture, fuel combustion, and consumer products such as wood products.³⁻⁷

Due to the reactive nature of the carbonyls, these chemicals are derivatized with a derivatization agent during or immediately after sample collection. The most widely used derivatization agent is 2,4-dinitrophenylhydrazine (DNPH), which reacts with the carbonyl group of the carbonyls to form hydrazones (DNPH-carbonyls) that can be separated using HPLC or GC.⁸⁻¹¹ Derivatization of carbonyls with *o*-(2,3,4,5,6-pentafluorobenzyl)hydroxylamine (PFBOA), followed by GC/ECD or GC/MS analysis, has been also reported.^{12,13} Dansylhydrazine (DNSH) is another derivatization agent reported for the determination of carbonyls.¹⁴

Capillary electrochromatography (CEC) is a relatively new micro-separation technique that combines the concepts of HPLC and capillary zone electrophoresis (CZE).¹⁵⁻¹⁸ Because the propelling force of the mobile phase in CEC is the electroosmotic flow (EOF), not only ionic compounds but also neutral compounds can be separated. Compounds are separated in CEC according to their partition between the mobile and

stationary phases, as well as their migration ability in the electric field. Because of the powerful separation efficiency and high sensitivity of the CEC, this technique has been increasingly used as an analytical tool in the pharmaceutical industry and in medical research.¹⁹⁻²² Recently, the CEC has been also applied to the analysis of environmental pollutants including carbonyls.^{23,24} However, Zhang *et al.*²³ only measured nine carbonyls, and important carbonyls such as formaldehyde and acrolein were not included in the study. Furthermore, some critical pieces of information, such as calibration and method detection limits were missing in the previous report,²³ their absence limited its usefulness as an analytical method. Dabek-Zlotorzynska *et al.*²⁴ reported using a regular CEC system to separate 13 DNPH-carbonyls. However, acetone-DNPH and acrolein-DNPH were not separated.

In this study, we present a CEC based analytical method for the determination of 12 carbonyls in indoor air, including important chemicals such as formaldehyde and acrolein. The comparison between the two-step gradient CEC method and the conventional HPLC method is also presented in this paper.

Experimental

Chemicals and solvents

Standard DNPH-carbonyl mix (30 µg/mL per carbonyl) and LpDNPH H10 cartridges were purchased from Supelco (Bellefonte, PA, USA). The standard contained the following 12 carbonyls in DNPH derivative form: formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, hexaldehyde, butyraldehyde, crotonaldehyde, benzaldehyde, valeraldehyde, *m*-tolualdehyde, and methacrolein. Individual carbonyl compounds (99+%), tetrahydrofuran (99.9+%), methanol

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(99.9%), tris(hydroxymethyl)aminomethane (99.9+%) and hydrochloric acid (high purity grade) were obtained from Aldrich (Oakville, Ontario, Canada). Acetonitrile (99.99%) was purchased from Omni Solv, EM Science (MERCK, Darmstadt, Germany).

Each individual DNPH-carbonyl stock solution (100 µg/mL) was prepared by spiking 500 µg of its corresponding carbonyl compound onto the LpDNPH H10 cartridge, followed by elution with 5 ml of acetonitrile. The tris(hydroxymethyl)aminomethane (Tris) buffer stock solution (500 mM) was prepared by dissolving the appropriate amount of Tris in water and adjusting the pH to the desired value with 1 M or 30% aqueous HCl. The buffer solutions used for CEC analysis were prepared by diluting the Tris stock solution and mixing it with various amounts of organic solvents.

Apparatus and operating conditions

An Agilent Capillary Electrophoresis (3D-CE system) with a built-in diode-array detector (DAD) was used for CEC analysis (Agilent Technologies Inc., Waldbronn, Germany). The Agilent CE ChemStation was used for the instrument control, data acquisition, and data processing. The CEC peaks were monitored at the wavelength of 358 nm. An Agilent HPLC pump (Agilent 1100 series, G1310A, IsoPump) was used to flush the CEC column. The CEC column (CEC-Hypersil C8, 100 µm i.d. and 40 cm long, filled with 3 µm-sized silica-based particles coated with chemically bonded octylsilane (C8)) was purchased from Agilent Technologies. For column conditioning, the column was flushed with a buffer consisting of acetonitrile and 25 mM Tris (adjusted to pH = 8.5 with hydrochloric acid (HCl)) in a volume ratio of 4 to 1. The column conditioning was carried out by stepping up the voltage from 5 to 25 kV in 5-kV intervals with 10 min per step while applying a pressure of 12 bar to the column inlet. After flushing, both inlet and outlet of the column were pressurized at 12 bars and the voltage was maintained at 25 kV for another 30 min. The buffer changing during experiments was accomplished electroosmotically with pressurization of the inlet and outlet at 12 bars. Each sample was introduced by electrostatic injection at a voltage of 5 kV for 40 s.

HPLC analysis was performed using a Hewlett Packard 1090 Win HPLC system, equipped with a DAD and an autosampler, controlled by HP ChemStation software (Hewlett-Packard GmbH, Waldbronn, Germany). The peak was monitored at 358 nm. DNPH-carbonyls were separated on an LC-18 column (25 cm × 4.6 mm × 5 µm particle size), purchased from Supelco. The mobile phase consisted of acetonitrile (A), water (B), and tetrahydrofuran (C). The gradient elution was started at 20% A, 60% B, and 20% C and held here for 3 min, then the elution was increased to 80% A and 20% B over 17 min and held for another 3 min at this composition, at a flow rate of 1.5 mL/min. The gradient mixture was changed back to the initial condition of 20% A, 60% B, and 20% C over 0.5 min and was kept there for another 2.5 min before the next injection. Each sample injection volume was 20 µL.

Sample collection and preparation

A BGI 400S pump (BGI Inc., Waltham, MA, USA) coupled with a Matheson 8270 flow controller (Matheson Tri-Gas, Montgomeryville, PA, USA) was used for collecting air samples. The indoor air was drawn through the LpDNPH H10 cartridge for 100 min at a flow rate of 1.8 L/min. The pump was calibrated with Gilian Gilibrator™ 2 (Sensidyne Inc., Clearwater, FL, USA). The cartridges were decapped just before sampling, and recapped immediately after the sampling.

Each cartridge was wrapped with aluminium foil during and after the sampling to avoid exposure to the light. Each cartridge was placed in a cooler during transportation and kept in a refrigerator in the laboratory until sample extraction.

To each cartridge, 5 ml of acetonitrile was added to elute the DNPH-carbonyls; the eluate was collected in a 5-mL volumetric flask over a period of about 10 min. Each eluate was adjusted to the 5-mL mark afterwards with acetonitrile. After thorough mixing, aliquots of the eluate were transferred to two 1.5-mL HPLC vials for CEC and HPLC analyses, respectively. One cartridge blank (cartridge without spiking) and one laboratory control (cartridge spiked with 3 µg of each DNPH-carbonyls) were processed together with each batch of samples.

Results and Discussion

Effects of buffer concentration

The ionic strength of the mobile phase is regulated by the concentration of the tris(hydroxymethyl)aminomethane/hydrochloric acid (Tris-HCl) buffer, which impacts the electroosmotic flow (EOF) in CEC. Once the optimal pH value of 8.5 was determined, the effect of buffer concentrations on the separation was evaluated. Five to 100 mM Tris-HCl buffer stock solution was mixed with 70% organic modifiers and 10% water, all on volume basis, to form the final mobile phases that contained Tris-HCl concentration between 1 and 20 mM. When the Tris-HCl concentration in the mobile phase increased from 1 mM to 20 mM, the electric current increased from 1.8 to 3.5 µA at the fixed voltage of 30 kV due to increased EOF. However, the increasing of electric current in the packed column promoted the generation of Joule heat, which could cause the formation of bubbles in the packed column. Meanwhile, with increasing of the Tris-HCl buffer concentration, a decreased CEC separation efficiency and broadening of the peak band were also observed. Under our experimental condition 10 mM Tris-HCl concentration in the mobile phase generated enough EOF in the CEC system without creating over-heating problems inside the column, when the column cassette temperature was maintained at 20°C and a pressure of 12 bars was applied on the both ends of the column.

Effect of organic modifiers

Methanol and acetonitrile were used as organic modifiers of the mobile phase in the CEC system for the separation of DNPH-carbonyls. The ratio of permittivity (mobility) to viscosity for methanol is only about half of that for acetonitrile,²⁴ which means that acetonitrile has more mobility than methanol. Experimentally, it was observed that, with the same concentration of organic solvent in the mobile phase, the retention times of the analytes in the acetonitrile-Tris-HCl solution were much shorter than those in the methanol-Tris-HCl solution. On the other hand, methanol provided better separation efficiency at the expense of longer elution times.

Therefore, a mixture of methanol and acetonitrile had been used as organic modifier. Following the leads of HPLC separation conditions,¹¹ different amounts of methanol and acetonitrile were mixed with Tris-HCl buffer to form various mobile phases. Results showed that neither methanol nor acetonitrile as organic modifier alone would give satisfactory separation of all 12 target carbonyls. An organic modifier consisting of methanol as the major component (from 50 to 70%) and a small portion of acetonitrile (about 10%) was found to be the most suitable condition in mobile phase for the separation of the targets.

Table 1 CEC method performance and indoor air concentrations ($\mu\text{g m}^{-3}$) of airborne carbonyls

Chemical	Calibration	Retention time	MDL study ($n = 7$) (spiking at $1.0 \mu\text{g}$ per compound per sample)		Recovery study ($n = 5$) (spiking at $3.0 \mu\text{g}$ per compound per sample)		Indoor air	
	r^2	Mean $\pm$ s.d./min	s.d. ($\mu\text{g}/\text{sample}$)	MDL ^a ($\mu\text{g}/\text{sample}$)	RSD, %	Recovery, %	Mean ^b / $\mu\text{g m}^{-3}$	Range/ $\mu\text{g m}^{-3}$
Formaldehyde	0.995	20.10 ± 0.15	0.12	0.4	6.5	108	28	5.8 – 85 ($n = 30$)
Acetaldehyde	0.998	25.51 ± 0.22	0.31	1.0	7.6	104	18	4.4 – 38 ($n = 30$)
Acrolein	0.999	30.07 ± 0.21	0.10	0.3	7.0	106	ND	ND
Acetone	0.993	31.48 ± 0.27	0.49	1.5	11.3	120	93	12 – 1300 ($n = 30$)
Propionaldehyde	0.991	32.41 ± 0.31	0.21	0.6	15.9	68	3.1	5.2 – 15 ($n = 6$)
Crotonaldehyde	0.989	35.97 ± 0.20	0.46	1.4	15.3	95	ND	ND
Methacrolein	0.998	38.40 ± 0.25	0.15	0.5	7.9	97	3.0	2.8 – 13 ($n = 11$)
Butyraldehyde	0.999	40.05 ± 0.23	0.07	0.2	5.2	90	2.2	1.4 – 11 ($n = 18$)
Benzaldehyde	0.994	43.42 ± 0.26	0.11	0.3	4.1	105	2.7	1.6 – 54 ($n = 4$)
Valeraldehyde	0.992	51.89 ± 0.33	0.51	1.6	6.3	105	ND	ND
<i>m</i> -Tolualdehyde	0.996	55.31 ± 0.25	0.33	1.0	15.5	124	9.6	4.6 – 99 ($n = 12$)
Hexaldehyde	0.989	58.07 ± 0.19	0.33	1.0	17.8	148	3.5	12 – 14 ($n = 2$)

a. MDL = $3.143 \times \text{s.d.}$, targets were not detected in the lab blanks. b. When calculating the mean, $1/2$ MDL value was used for non-detected values.

Separation of 12 carbonyls using mobile phases with different organic modifiers is presented in Figs. 1a, 1b and 1c. All 12 targets were completely separated with 50% of methanol and 10% of acetonitrile in the mobile phase, but the analytical run time was very long (130 min) (Fig. 1a). When the concentration of organic modifier was increased to 70% of methanol and 10% of acetonitrile in mobile phase, the analytical run time was shortened to about 22 min, but only the last two peaks were separated (Fig. 1b). Using 60% of methanol and 7.5% of acetonitrile in mobile phase, a reasonable analytical run time was achieved with the separation of all peaks except peaks 4 and 5 (Fig. 1c). It was interesting to note that, for the third mobile phase, it was necessary to reduce the volume of acetonitrile from 10% to 7.5% in order to get satisfactory separation for the middle four peaks (crotonaldehyde, methacrolein, butyraldehyde and benzaldehyde).

Influence of cassette temperature and applied voltage

Higher column temperature in CEC leads to lower viscosity of the mobile phase, which resulted in increased EOF for a given voltage.²⁵ As in HPLC, van't Hoff plots of $\ln \kappa$ (here κ is the retention factor) versus $1/T$ (T is the temperature) in CEC has been demonstrated to be linear, but the relation has different slopes for different compounds.²⁵ Although the temperature inside the column (column temperature) differs from the cassette temperature because of Joule heating, our experiment showed that the retention times of the targets shortened when the cassette temperature increased, at the expense of decreased separation efficiency. Lower temperature resulted in improved separation efficiency at longer analytical run time. A column temperature of 20°C was found to be a good balance between separation efficiency and analytical time.

EOF is directly proportional to the electric field (applied voltage) of the column and the Joule heating also depends on the magnitude of the applied voltage. Therefore, higher electric field accelerates the movement of the analytes in the cassette, which helps reduce the analytical run time. The experiments showed that, with the increase of applied voltage, the risk of bubble formation in the column increased because of Joule heating. It was found that 25 kV was the maximum voltage that could be applied without suffering the bubble formation problem when both ends of the column were pressurized at 12 bars.

Two-step gradient CEC system

In HPLC systems, it is common to use a gradient mobile phase which is programmed by the solvent delivery control system to improve separation efficiency of the analytes. In contrast, most of the CEC separations were performed without gradient.²⁵ Like HPLC, application of a gradient buffer system in CEC, such as a two-step or multi-step gradient, could lead to improved separation efficiency. Because commercial CE instruments now have auto-switch systems for buffer changing, it is possible to achieve two-step or multi-step gradient buffer system within a CEC system.

In this study, switching of mobile phase was controlled by the instrument operating system and was automated for the sequence run. During the switch, both inlet and outlet voltages were reduced to zero to halt the flow. The voltage was raised to its original value once the switch is completed. The optimal two-step gradient CEC buffer system was developed based on the separation results of these three individual mobile solutions. As discussed earlier, single mobile phase could not provide satisfactory results to meet the separation efficiency within a reasonable analytical run time. The multi-step gradient system incorporates the advantage of each single mobile phase. In our case, all mobile phases contained 20% of 50 mM Tris-HCl buffer (to a final concentration of 10 mM in the mobile phase). In addition to Tris-HCl buffer, the initial mobile phase contained 50% of methanol and 10% of acetonitrile as organic modifier, and 20% of water, the same values as those used in Fig. 1a, to allow good separation of the first five peaks. It was not necessary to wait for the complete elution of the first five peaks before switching to the next mobile phase. At 18 min, the mobile phase was changed to 60% of methanol and 7.5% of acetonitrile as organic modifier, and 12.5% of water, the same values as the one in Fig. 1c, for the separation of the next four peaks. The third mobile phase (70% of methanol and 10% of acetonitrile as organic modifier, same condition as in Fig. 1b) was introduced at 45 min for the elution of the last three peaks. At 60 min, after the completion of eluting all 12 targets, the system was switched to the initial mobile phase in preparation for the next run. It was found that a good reproducibility of retention time for DNPH-carbonyls (Table 1) can be achieved in this condition with, however, a worse RSD compared to those for the HPLC and GC method.

Fig. 1 Effect of organic modifier: sample 1.5 ppm DNPHs in a mixture of acetonitrile and 50 mM Tris, adjusted to pH 8.5 (ratio 4:1). Column: CEC capillary C8, 3 μ m, 100 μ m/40 cm; voltage, 25 kV; cassette temperature, 20°C; injection, 5 kV/40 s. Mobile phase composition (methanol:acetonitrile:50 mM Tris:water, volume based) was (a) 50:10:20:30, (b) 70:10:20:0, (c) 60:7.5:20:12.5, (d) 0–18 min, 50:10:20:30, 18–45 min, 60:7.5:20:12.5, 45–60 min, 70:10:20:0. Peak identification: 1, formaldehyde-DNPH; 2, acetaldehyde-DNPH; 3, acrolein-DNPH; 4, acetone-DNPH, 5, propionaldehyde-DNPH; 6, crotonaldehyde-DNPH; 7, methacrolein-DNPH; 8, butyraldehyde-DNPH; 9, benzaldehyde-DNPH; 10, valeraldehyde-DNPH; 11, *m*-tolualdehyde-DNPH; 12, hexaldehyde-DNPH.

Instrument performance

Linear calibration curves were observed with coefficient of determination (r^2) at or greater than 0.99 for all target analytes (Table 1). For example, the r^2 values for formaldehyde, acetaldehyde, *m*-tolualdehyde and hexaldehyde were 0.995, 0.998, 0.993, and 0.989, respectively. The dynamic range was from 0.06 to 1.5 μ g/mL for all the DNPH-carbonyls, compared to the dynamic range of HPLC method (0.5 to 20 μ g/mL)²⁸ and GC method (0.07 to 13 μ g/mL).¹¹ However, the calibration curve of the GC method was not linear, rather, it follows a second-order polynomial equation.¹¹

Fig. 2 Sample-to-sample comparison between HPLC and CEC methods for (a) formaldehyde, (b) acetaldehyde, and (c) *m*-tolualdehyde. Conditions same as in Fig. 1d.

The method detection limit is also given in Table 1. The method detection limits for the 12 carbonyls ranged from 0.2 μ g to 1.6 μ g per sample, or 0.04 to 0.32 ng/ μ L of the final injection solution. Compared to the instrument detection limit reported by Zhu *et al.*,¹¹ the detection limits of our CEC method were 2 to 5 times higher than the HPLC (0.012 to 0.032 ng/ μ L) or GC method (0.01 to 0.2 ng/ μ L). The recoveries of target analytes in this method were generally between 90% and 120%, except for hexaldehyde (148%), with a typical relative standard deviation (RSD) at around 10%.

Measurement of indoor air

Thirty indoor air samples that contained amounts of formaldehyde and acetaldehyde detectable by the HPLC method were chosen for the CEC analysis. These samples were a subset of samples from 75 randomly selected homes in the city of Ottawa. The summary results are given in Table 1. It was found that the concentration ranges of formaldehyde and acetaldehyde in indoor air samples were 5.8 μ g/ m^3 to 85 μ g/ m^3 (mean: 28 μ g/ m^3), and 4.4 μ g/ m^3 to 38 μ g/ m^3 (mean: 18 μ g/ m^3), respectively. The other major carbonyl found in indoor air was acetone, a solvent commonly used in many consumer products. Three carbonyls, acrolein, crotonaldehyde, and valeraldehyde, were not detected in the samples, while the other six carbonyls were detected in low frequency (Table 1).

Concentrations of airborne carbonyls, particularly

formaldehyde and acetaldehyde, in residential indoor air have been reported in the range of 20 $\mu\text{g}/\text{m}^3$ to 60 $\mu\text{g}/\text{m}^3$, with occasionally high values in the 100 $\mu\text{g}/\text{m}^3$ marker, in the United States, Germany, and other countries.²⁶ Gonzalez-Flesca *et al.* recently reported an indoor air concentration range of 16.0 $\mu\text{g}/\text{m}^3$ to 44.6 $\mu\text{g}/\text{m}^3$ for formaldehyde with a mean value of 25.0 $\mu\text{g}/\text{m}^3$, and a range of 7.4 $\mu\text{g}/\text{m}^3$ to 50.6 $\mu\text{g}/\text{m}^3$ for acetaldehyde with a mean value of 24.3 $\mu\text{g}/\text{m}^3$, respectively, measured in 10 homes.²⁷ The levels detected in this study were similar to the results of these earlier studies.

Comparison of values determined by CEC and HPLC method

The eluates had also been subjected to HPLC analysis using established standard HPLC methods.^{28,29} A sample-by-sample comparison of formaldehyde, acetaldehyde and *m*-tolualdehyde is graphically illustrated in Fig. 2. For formaldehyde, the slope was 1.13, meaning that the values measured using HPLC method were slightly higher than those of CEC. On the other hand, the slope was 0.95 for acetaldehyde, indicating that the CEC values were slightly higher than those of HPLC. In general, the differences were within 20% of each other for each chemical. However, for tolualdehyde, the slope was 1.44, indicating that the values from HPLC was about 50% higher than those from CEC method.

Conclusion

Compared to the published studies, our two-step gradient CEC method is the first report that details the separation of all 12 commonly monitored carbonyls in their DNPH derivative form. Our study not only includes the measurements of such important carbonyls as formaldehyde, but also provides information on method performance for its application. Although the 12 DNPH-carbonyls can be separated and measured under GC¹¹ or HPLC^{28,29} conditions, the separation of acrolein-DNPH is often a challenge under HPLC condition while DNPH-carbonyls do not respond to the GC instrument in a linear manner. Our CEC method separates the peak of acrolein-DNPH well from that of both acetone-DNPH and propionaldehyde-DNPH and our method also has reasonable linear calibration range for all targets, thereby providing an alternative to measure these chemicals in environments such as indoor air.

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