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Determination of Low-molecule-weight Aldehydes in Packed Drinking Water by High Performance Liquid Chromatography

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ABSTRACT

A high performance liquid chromatographic (HPLC) method for the determination of formaldehyde, acetaldehyde and propionaldehyde in packed drinking water was developed. Aldehydes were derivatized with 0.5 mL DNPH (2,4-dinitrophenylhydrazine) reagent and 0.1 mL 2 M perchloride at 55˚C in 60 minutes, then adsorbed in C18 glass cartridge (was performed), eluted with 5 mL acetonitrile and finally determined by HPLC. Formaldehyde, acetaldehyde and propionaldehyde-DNPH-derivatives were seperated on a Cosmosil 5C18-MS column by using acetonitrile/D.I. water (55/45, v/v) as a mobile phase, and detected with UV 360 nm. Recovery studies were performed by fortifying D.I. water with formaldehyde, acetaldehyde and propionaldehyde at various concentrations from 2 to 200 ppb. Recovery rates were in the range of 84.4 – 103.2%, 90.2 – 122.1% and 60.8 – 100.4%. The detection limits for all three aldehydes were 1 ppb.

Sixty-three packed drinking water samples collected from various markets were analyzed. These results were well below the guideline for drinking water quality by WHO (900 μg/L).

Key words: packed drinking water, formaldehyde, acetaldehyde, propionaldehyde, DNPH derivative

INTRODUCTION

As early as 1981, packed drinking water has been available in Taiwan. In recent years, along with prosperous economic development, water pollution is becoming a serious problem. The deterioration of water quality in some area has aroused public concern about pursuing nature, pure and healthy drinking water. Packed water, including mineral water, packed drinking water, distilled water and reverse osmosis water, with the ready-to-drink character, are therefore full of the market.

Other than satisfying thirst, there are various marketing strategies like gymnasium, entertainment ground, or SPA etc. Packed water could even replace traditional boiled water as the main source of drinking water at home and in families place, as well.

Due to early moldy-like and recent benzene contamination, the safety of packed water are public concerns. Water could contain low molecular weight aldehydes like formaldehyde, acetaldehyde and propionaldehyde because of natural biological transformation of microbials, non-biological gas-liquid phase transformation, or photo-thermal chemical reactions, as well as artificial industry pollution, ozone processing in wasted water, or ozone processing in drinking water to reduce methyl tricloride generated by chlorination process. The high chemical reactivity of these aldehydes has caused serious concern in the environmental protection point of view. Furthermore, packed water could also contain aldehydes due to malmanufacturing process, unsuitable container, or incomplete cleaning of the container. In the 1996 survey (Ling *et al.*[8]) of mineral water on the market, formaldehyde content were ND – 47.6 ppb, acetaldehyde content was 4.1 – 165.0 ppb, while propionaldehyde was not detected.

Low-molecular-weight aldehydes can be analyzed with spectrophotometry[9-11], gas chromatography[11-13], liquid chromatography[4,14-20] etc. However, gas or liquid chromatography is more applicable to detect low-molecular-weight aldehydes in water with higher detection sensitivity. Both US EPA[21] and ROC EPA[8] use HPLC system to detect DNPH derivatives. The reaction equation of DNPH was shown in Figure 1[8]. According to literature, detection sensitivity can be improved by using solvent extraction and solid phase absorption column[4,15-18]. However, due to matrix effect, it cannot be effectively used in samples containing low molecular weight aldehydes. The purpose of this study is to establish an analysis method to reduce back-

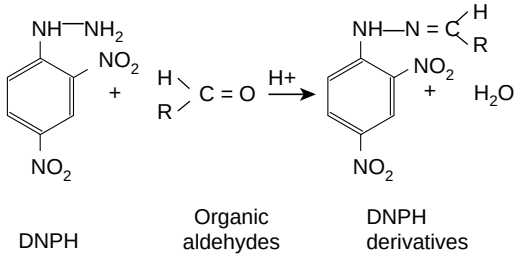

Figure 1. DNPH derivative of organic aldehydes(8)

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ground noise caused by matrix, and improve detection sensitivity. Furthermore, this method can be utilized in the investigation of packed water on the market for detection of content of formaldehyde, acetaldehyde and propionaldehyde, so that the safety of aldehyde content in packed water can be understood.

MATERIALS AND METHODS

I. *Materials*

Standards of DNPH derivatives of formaldehyde, acetaldehyde, and propionaldehyde, and 30% 2,4-dinitrophenylhydrazine (DNPH) in water (w/w) were purchased from Chem Service, Inc., USA. Acetonitrile (liquid chromatographic grade) and perchloric acid (reagent grade) were purchased from Merck, Germany. Other equipment used in this study included brown wide-mouth reaction bottle (100 mL, screw-capped), oven (55˚C, Shen Ren Corp Inc.), evacuation filtration device (Supelco visiprep DL 5-7044), C18 glass purification column (Chromabond C18, Macherey-Nagel, pre-treated with 10 mL acetonitrile in a flow rate of 3-5 mL per min, evacuated for additional 10 min after drainage), high-performance liquid chromatography (HPLC, Hitachi L-6200 intelligent pump, L-4250 UV-Vis detector, Shimadzu SIL-9A auto injector), chromatographic column (Cosmosil 5C18-MS Waters, 4.6 × 250 mm; mobile phase: acetonitrile/D.I. water = 55/45, v/v; flow rate: 1 mL/min; detection wavelength: 360 nm).

II. *Preparation of Reagents*

(I) *Preparation of standard sample solutions:*

Standards of DNPH derivatives of formaldehyde, acetaldehyde, and propionaldehyde were dissolved with acetonitrile to make stock standard solutions in the concentration of 1000 µg/mL. Each solution was diluted with acetonitrile to make 10 µg/mL and 100 µg/mL standard mixture solutions, then further diluted into concentrations from 0.2 to 10 µg/mL as working standard solutions.

(II) *Preparation of DNPH reagent:*

Dissolve 0.71 g of 30% DNPH with acetonitrile to make total volume of 100 mL.

(III) *Preparation of 2 M perchloride:*

Dissolve 28.71 g of 70% perchloride with D.I. water to make total volume of 100 mL.

(IV) *Preparation of sample solution:*

Water sample 50 mL was mixed with 0.5 mL of DNPH reagent and 2 drops of 2 M perchloride in a brown wide-mouth bottle, screw the cap, and reacted in 55˚C oven for 1 h. Reaction solution was poured slowly through a pre-treated purification column in a flow rate of 3-5 min/min. After evacuating for additional 10 min after drainage, the sample solution was eluted by acetonitrile in a flow rate of 1-2 mL/min. Adjusted the final volume to make 5 mL.

(V) *Preparation of standard curve:*

Each standard mixture solutions, from 0.2 to 10 µg/mL, were analyzed in triplicate using HPLC. Three standard curves were then plotted by peak area against concentration of each aldehyde.

(VI) *Identification test and quantitative test:*

Ten microliter of sample solution and standard mixture solution were auto-injected into HPLC, respectively, and the peak retention time and peak area of sample solution were compared with those of standard mixture solution. The concentration of formaldehyde, acetaldehyde and propionaldehyde in sample solution were calculated according to the equation as shown below.

$$\text{Aldehydes concentration (ppb)} = C \times \frac{V}{V_{(sample)}} \times CF \times 1000$$

C: aldehydes concentration (µg/mL) of DNPH derivatives in eluant extrapolated from the standard curve
V: eluant volume (mL)
$V_{(sample)}$: water sample volume (mL)
CF: conversion factors when converting DNPH derivatives of aldehydes to aldehydes[21]
Formaldehyde: 0.143
Acetaldehyde: 0.196
Propionaldehyde: 0.247

RESULTS AND DISCUSSION

I. *Selection of HPLC Analysis Conditions*

(I) *Selection of detection wave length*

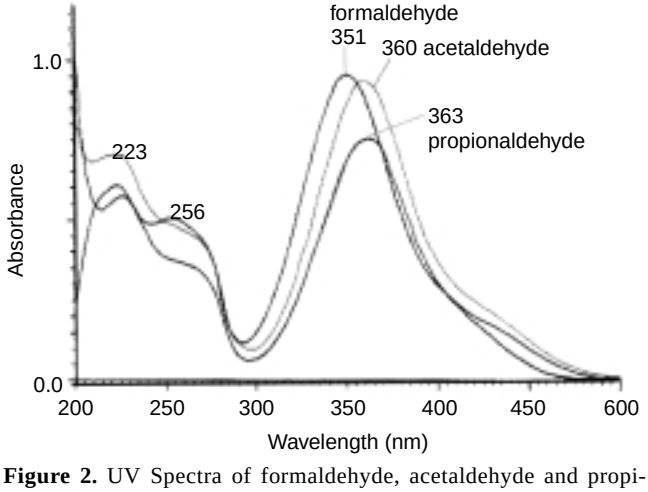

Figure 2. UV Spectra of formaldehyde, acetaldehyde and propionaldehyde DNPH-derivatives.

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10 µg/mL solution of DNPH derivatives of formaldehyde, acetaldehyde and propionaldehyde was prepared and scanned at 200-500 nm wavelength of spectrophotometer. The peak absorption of formaldehyde DNPH-derivative can be read at 351 nm, and the peak absorption of acetaldehyde and propionaldehyde DNPH-derivatives can be read at 360 nm and 363 nm, respectively. Therefore, in the spectrum shown in Figure 2, 360 nm can be the selected wavelength to detect the 3 aldehyde DNPH-derivatives. 360 nm is also selected by the US EPA method 554[21] to detect carbonyl-DNPH derivatives.

(II) *Selection of mobile phase*

Refer to the mobile phase condition in literature[15-17], different ratio of acetonitrile/D.I. water mixture was used as mobile phase. 1 µg/mL standard solution of formaldehyde, acetaldehyde, and propionaldehyde-DNPH derivatives mixture was injected to HPLC and the chromatogram was compared. The best condition can be obtained when acetonitrile/D.I. water (55/45, v/v) is used as the mobile phase (Figure 3). Also, previous chromatogram was used as a reference to determine different ration of methyl alcohol / D.I. water as the mobile phase. The best condition can be obtained when methanol/D.I. water (68/32, v/v) is used. However, after a test of derivatized reaction of DI water, the chromatogram of acetonitrile/D.I. water (55/45, v/v) was found to have less background noise than that of methanol/D.I. water (68/32, v/v). Comparing these 2 mobile phase solutions, we found there was less organic

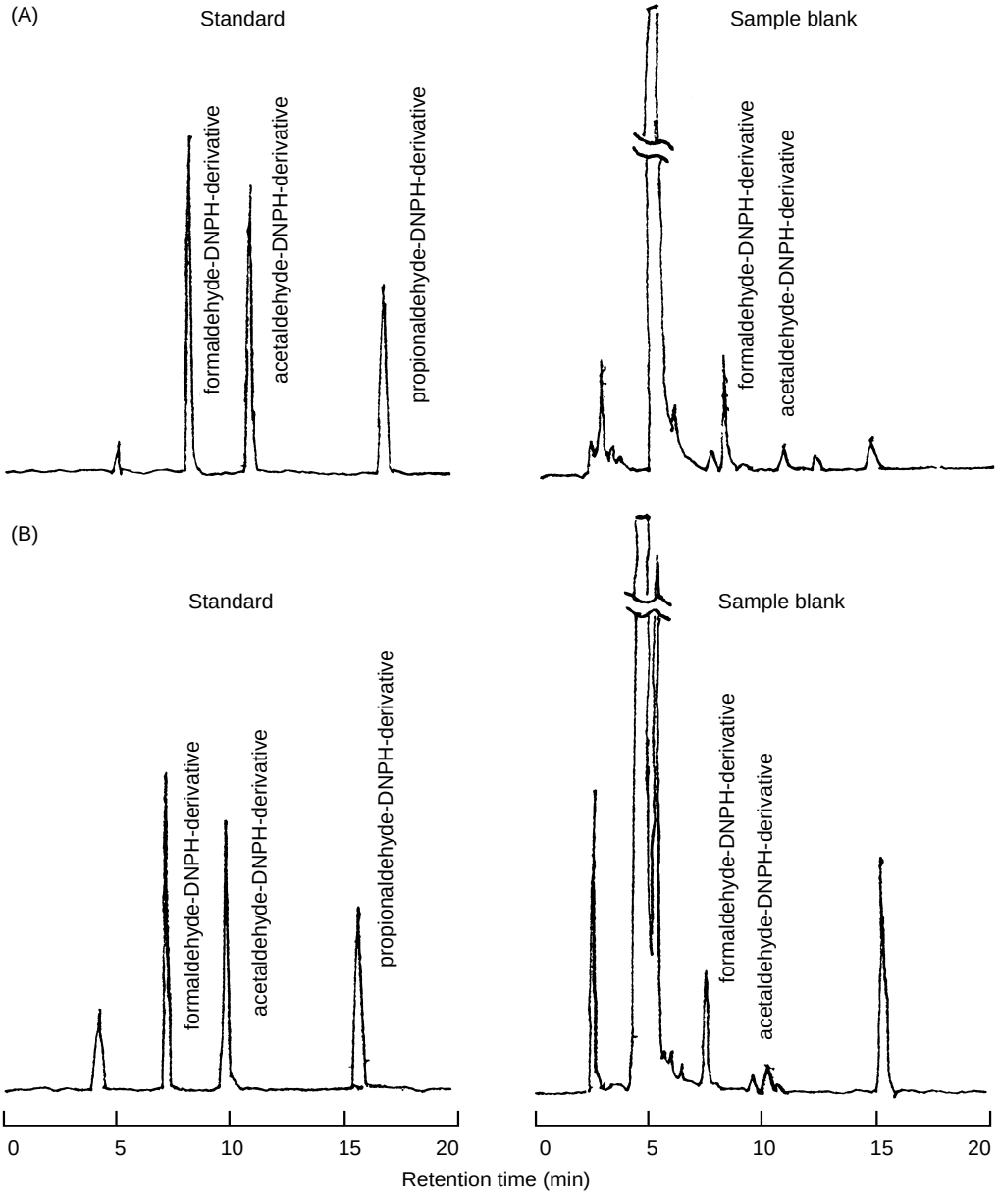

Figure 3. HPLC chromatograms of formaldehyde, acetaldehyde and propionaldehyde DNPH derivatives. Mobile phase of (A) CH_3CN/H_2O = 55 / 45 (v / v) and (B) $MeOH/H_2O$ = 68 / 32 (v / v) with flow rate at 1 mL/min and UV detection at 360 nm was used.

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solvent in acetonitrile. Therefore, acetonitrile/D.I. water (55/45, v/v) was selected to be the mobile phase.

II. *Sketching of the Standard Curve*

The standard curve shows that the absorption of formaldehyde, acetaldehyde and propionaldehyde DNPH-derivatives are in good linear relation between 0.2 – 10 µg/mL, r = 1. The linearship is very correlated.

III. *Selection of Purification Condition*

(I) *Selection of C18 purification glass column*

Sample contamination can be reduced by skipping pre-process procedures and performing analysis as soon as possible after sampling. In the literature[4, 14-17], C18 is usually used to absorb the detection subject so that sensitivity can be improved. In this study, C18 was chosen as purification packing material to absorb aldehyde DNPH-derivatives, and then eluted by acetonitrile. To save time and obtain consistent study result, commercial C18 purification column was used. Besides, aldehydes can also be released from plastic material; to avoid contamination and interfere, glass column was selected.

(II) *Elution efficacy of C18 glass purification column*

50 mL of 2 µg standard formaldehyde, acetaldehyde and propionaldehyde DNPH-derivatives mixture D.I. water was put through C18 glass purification column at 3 – 5 mL/min flow rate. After 10 min suction, acetonitrile was installed to elute the samples. One mL of sample was collected from each tube. As a result in the second mL, formaldehyde can be recovered up to 73%, acetaldehyde can be recovered up to 77%, while propionaldehyde can be recovered up to 75%. In the third mL the elution is 100% recovered. To ensure complete elution, the total volume was 5 mL.

(III) *Absorption efficacy of C18 glass purification column*

Absorption capacity of the column was tested by injection of 50 mL of D.I. water containing 2, 4, 6 and 8 µg standard formaldehyde, acetaldehyde and propionaldehyde DNPH-derivatives mixture. As a result, the recovery rate of lower concentration sample is better than higher ones. This suggests the possibility of overloading when using high concentration sample. However, the recovery rate at 85% – 96% of 8 µg (or less) derivatives standard solution shows the absorption capacity is still good.

IV. *Selection of Reaction Condition of Derivatization*

(I) *The derivatization method adopted by both ROC EPA and US EPA Method 554 is DNPH-derivatization.*

The difference is, NaCl was not utilized in US EPA Method 554. In the research of Wu, 1995[16], the pH value of derivatize reaction was 5. Therefore, in the study we did not add NaCl nor adjust pH value in order to understand the feasibility of omitting the above 2 steps. As a result, 50 mL of 0.2, 0.5, 1, 2, 3 and 4 µg formaldehyde standard D.I. water used in the test got recovery rate more than 80%. This proves the skipped steps of adding NaCl and pH value adjustment would not affect the efficiency.

(II) *The effect of adding perchloride*

In the above mentioned study, the pH value was 5. Leaving pH value intact has not led to a worse result. When adding 0.05 mL 2 M perchloride solution, the pH value of sample solution became 5. In the research of Kieber, 1990[4], it was found that a large amount of perchloride would induce the chemical reaction of propionaldehyde turning into formaldehyde, raising its level. We adjusted the amount of perchloride to 0.05, 0.1 and 0.15 mL and observed the influence. The aldehyde content, when adding perchloride at 0.05 or 0.1 mL, did not change significantly, while adding perchloride at 0.15 mL would result in a higher content of aldehydes in the high concentration (3 µg / 50 mL) sample (Figure 4), and cause peak of propionaldehyde DNPH-derivative split to 2. Therefore, adding percholride should be within 0.1 mL.

(III) *The effect of purity and amount of DNPH*

It was mentioned in the literature[8], that when DNPH is not in high purity form, it should be extracted with hexane/methylene chloride 70/30 (v/v), crystalized by acetoni-

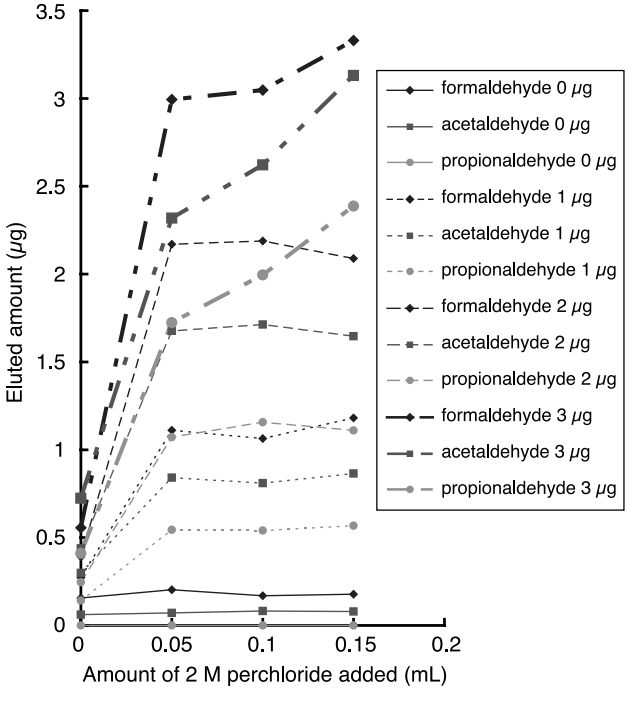

Figure 4. Effect of perchloride on the elution of DNPH derivatization of formaldehyde, acetaldehyde and propionaldehyde.

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trile, and dried by nitrogen. In this study, when D.I. water alone was used in blank test, there were peak at formaldehyde retention time, so we performed DNPH purity test. DNPH reagent was diluted by acetonitrile to the relative concentration of sample (DNPH reagent 0.5 mL in acetonitrile 5 mL) and then injected into HPLC. The result shows DNPH reagent has no peak at the relative retention time of formaldehyde, acetaldehyde and propionaldehyde, therefore DNPH needed no further purification. Regarding the effect of amount of DNPH to the test result, it was found no increase of recovery under 0.5, 0.7 and 1 mL DNPH reagent, so 0.5 mL was just enough for derivatize reaction.

(IV) *Effect of reaction temperature and time*

In the literature[8,15-17,21], the temperatures required by derivatization includes 4, 24 and 55˚C; different reaction time is required when the temperature is different. A cross-over design was proposed at temperature 4˚C, 24˚C (room temperature) and 55˚C, while reaction time was 20, 40, 60, 90 and 120 mins. As a result, both formaldehyde and acetaldehyde can get maximum reaction rate at 4˚C, after 60 mins reaction time. When at 55˚C, 20 mins reaction time can achieve high reaction rates for formaldehyde, acetaldehyde and propionaldehyde, and the reaction rate did not increase with reaction time (Figure 5). In summary, the condition of derivatizing reaction was set to be 55˚C at 60 mins to ensure a complete reaction for the test.

V. *Recovery Test*

0.1, 1, 5 and 10 μg/mL formaldehyde, acetaldehyde and propionaldehyde mixture standard solution, 1 mL of each, was added to 50 mL D.I. water. The recovery test was performed using the method established above, and blank test was used as control. Each test was triplicated and the retention time and peak of sample solution were compared with standard solution to identify and quantify, and calculate the recovery rate. As a result, the average recovery rate was 81.4 – 103.2%, 90.2 – 122.1%, and 60.8 – 100.4%; CV = 0.2 – 16.6% (Table 1) (Figure 6). The recovery rate of formaldehyde and acetaldehyde was good, while propionaldehyde at concentration above 20 ppb had a recovery rate of only 60%, but 100.4% when the concentration was 2 ppb. In the literature[8], the content of propionaldehyde was undetectable in drinking water, suggesting very low content of propionaldehyde. Therefore, this method should fulfill the examination objective of low molecular weight aldehydes in packed water.

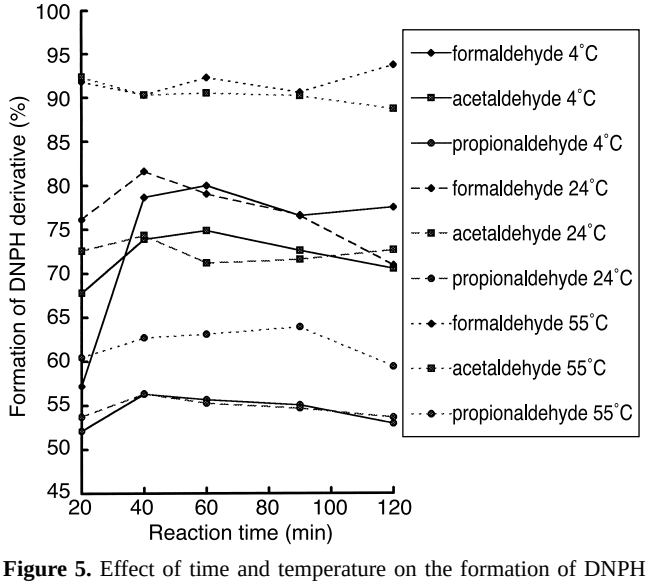

Figure 5. Effect of time and temperature on the formation of DNPH derivatization of formaldehyde, acetaldehyde and propionaldehyde in water.

VI. *Test of Limit of Quantification*

0.05 μg/mL formaldehyde, acetaldehyde and propionaldehyde mixture standard solution, 1 mL of each, was added to 50 mL D.I. water. The recovery test was performed using the method established above, and blank test was used as comparison. Each test was triplicated and the retention time and peak of sample solution were compared with standard solution to identify and quantify, and calculate recovery rate and limit of quantification. As a result, the average recovery rate of triplicate test was 157.6%, 145.4% and 116.7%, CV = 2.8-30.5% (Table 1). Because there was very low content of formaldehyde and acetaldehyde in D.I. water, when performing analysis at very low quantity, the recovery rate of formaldehyde and acetaldehyde was substantially influenced. For the recovery rate of propionaldehye, the CV was found to be very low, indicating run-on a good recovery rate at 1 ppb using this method. The ration of signal/noise (S/N) is above 3, indicating 1 ppb can be the limit of quantification of this method (Figure 6).

VII. *Stability Test of Sample Solution*

Due to relatively poor stability of derivatives, the sample solution was put to stability test after the reaction. The sample solution of recovery test was preserved 7 days at 4˚C; HPLC assay was done every other day. As a result, sample solution concentration between 20-200 ppb showed no difference within 7 days after the recovery test. Sample solution concentration between 1-2 ppb showed difference within 10% on the third day. Over the seventh days, the

Table 1. Recovery[a] of formaldehyde, acetaldehyde and propionaldehyde in distilled water.

	Recovery (%)		
Spiked level (ppb)	formaldehyde	acetaldehyde	propionaldehyde
1	157.6 (23.4)[b]	145.4 (30.5)	116.7 (2.8)
2	106.9 (16.6)	122.1 (15.9)	100.4 (11.9)
20	103.2 (5.2)	105.6 (6.1)	66.4 (2.0)
100	84.4 (0.2)	90.2 (0.9)	60.8 (0.4)
200	83.9 (1.8)	93.0 (1.3)	62.1 (0.8)

a: Average of three determinations. b: CV%.

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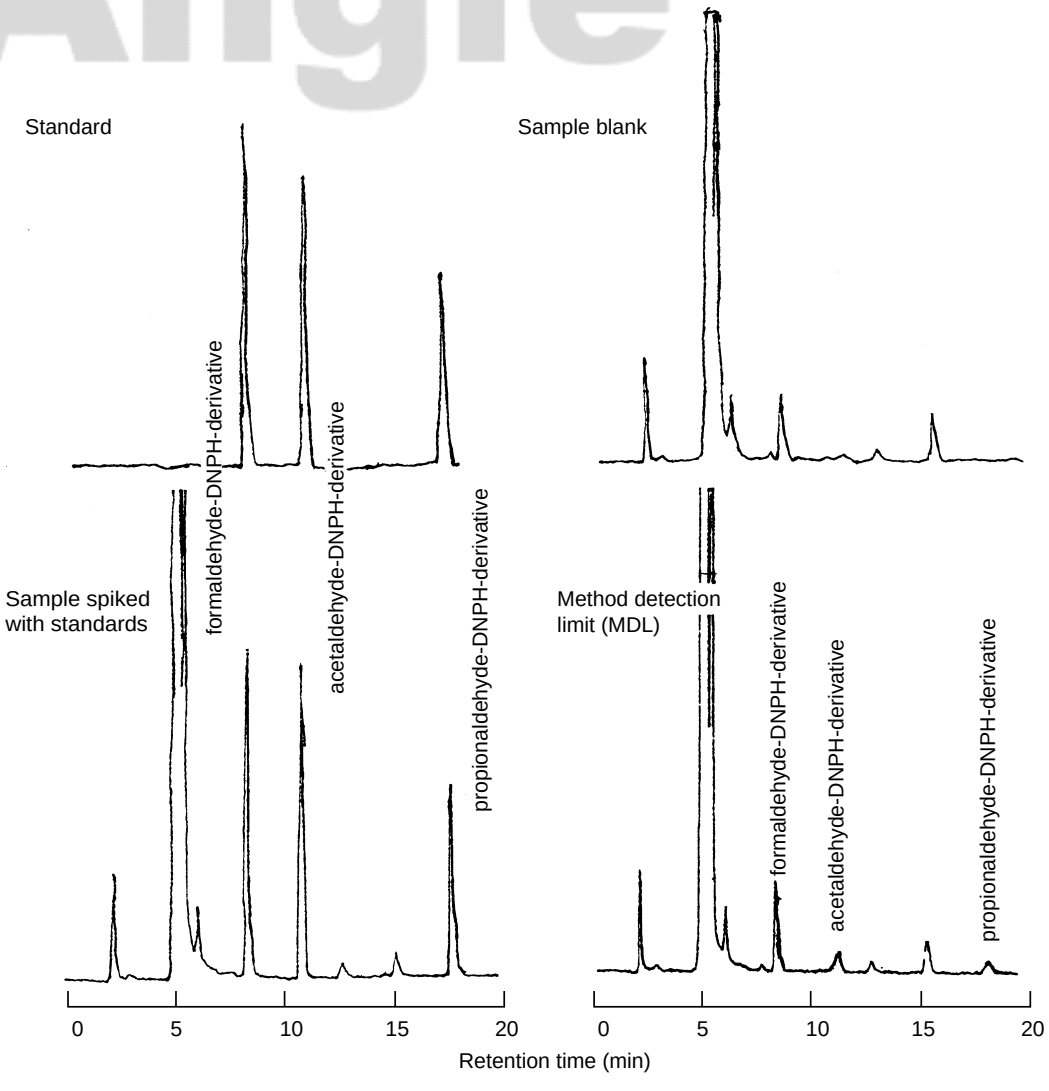

Figure 6. HPLC chromatograms of formaldehyde, acetaldehyde and propionaldehyde DNPH derivatives in D.I. water. A mobile phase of CH_3CN/H_2O = 55 / 45 (v / v) with flow rate at 1 mL/min and UV detection at 360nm was used. Concentration of standard solution was 1 ppm and ample spiked with 100 ppb standard and 1 ppb MDL.

largest difference of formaldehyde was about 20%, while in acetaldehyde and propionaldehyde the difference was within 10%. This means, sample solution should be tested within 3 days of derivatization (figure 2). In the Kieber 1990[4] research, sea water sample was derivatized and underwent Sep-Pak purification process at different time point. The result showed no difference within 14 days and enough stability was proved.

VIII. *Comparison of Packed Water and Automat Barreled Water with WHO Guidelines for Drinking-Water Quality*[22]

In the Guideline for quality of drinking water established by WHO; the limit of formaldehyde content is 900 µg/L. In this study, 63 brands of packed water and 13 brands of automat barreled water were tested for formaldehyde content. All detected quantity were within 129 ppb, far below the WHO limit,.

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Table 2. Stability of DNPH-derivatives of formaldehyde, acetaldehyde and propionaldehyde in sample solution.

Aldehyde	Spiked level (ppb)	Recovery (%) 1 day	3 days	5 days	7 days
formaldehyde	200	83.87	90.72	93.35	91.18
	100	81.37	87.59	89.73	88.6
	20	103.23	98.22	100.57	99.51
	2	106.9	121.14	120.55	131.19
	0.1	157.6	151.98	154.23	174.19
acetaldehyde	200	93	95.48	96.18	94.68
	100	90.2	92.42	92.89	92.15
	20	105.57	99.02	104.3	103.98
	2	122.07	129.08	133.86	128.33
	0.1	145.43	149.02	164.58	145.47
propionaldehyde	200	62.07	62.02	62.95	62.31
	100	60.8	61.12	61.66	61.41
	20	66.4	67.62	69.2	68.23
	2	100.37	96.47	106.32	106.2
	0.1	116.73	107.97	121.12	108.39

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