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2 **Assessing organic chemical emissions and workers' risk of exposure**
3 **in a medical examination center using solid phase microextraction devices**

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23

24 **Abstract**

25

26 Needle trap samplers (NTS), which are environmentally friendly solid phase
27 microextraction sampling devices, were used to obtain air samples at a medical
28 examination center in a teaching hospital to determine the concentration of xylene in
29 the air. The standard active sampling method using, Method 1501, was simultaneously
30 used to evaluate the exposures exposure of worker to xylene. The concentrations of
31 xylene were much lower than the legal 100-ppm time weighted average (TWA)
32 concentration. Another organic reagent that is used in the medical examination center,
33 formaldehyde, did not exhibit co-adsorption along with the extraction of xylene by
34 NTS. The use of a fume hood satisfactorily reduced VOC emissions in the work place.
35 Additionally, a management strategy that involves chemical control banding was
36 adopted to evaluate the emission control performance of xylene and formaldehyde in
37 this work. The NTS is recommended for use as a routine micro-sampler to be worn by
38 workers to protect their health.

39

40 **Keywords:** exposure risk; sampling; solid phase microextraction; xylene;
41 formaldehyde; hospital.

42

43 1. Introduction

44

45 Indoor air quality depends on the activities of people. In particular, in some
46 indoor work places, in which central air conditioning system, such as heating,
47 ventilation, and air condition (HVAC) systems, influence indoor air quality (IAQ)
48 (Rösch *et al.*, 2014; Ayodele *et al.*, 2016). Notably, the hospital is an almost
49 completely indoor space with various medical activities continuously in progress.
50 According to Wu *et al.*'s field survey at a teaching hospital in Taiwan, more hazardous
51 chemicals and solvents are used in the medical examination center than in any other
52 work rooms (Wu, *et al.*, 1998).

53

54 Xylene and formaldehyde are the main organic compounds that are used in a
55 medical examination center (Fooyin Hospital, 2016). Human are exposed to xylene by
56 inhalation, and the resulting symptoms include excitability, mental confusion,
57 staggering, headache, dizziness, narcosis and coma. Formaldehyde mainly causes
58 irritation of the eyes and respiratory tract, pulmonary edema, inflammation and
59 pneumonia. Additionally, formaldehyde has been classified as a "probably
60 carcinogenic agent" by the International Agency for Research on Cancer (IARC)
61 (Kent, 1998). The synergistic toxicology on humans for mixtures of xylene and

62 formaldehyde has seldom been investigated. Many results of mouse tests have
63 revealed that mixtures of xylene and formaldehyde can synergistically reduce mice
64 memory, cause genetic damage in the liver, kidney and marrow, and increase the rate
65 of sperm malformation (Bai, 2012; Wu, *et al.*, 2012).

66

67 Various sampling methods have been used to measure the emissions of multiple
68 mixtures of indoor volatile organic compounds (VOCs). Can *et al.* (2015) measured
69 VOC emissions, using 200 ± 1 mg 18–35 mesh granular activated carbon as the
70 adsorbent, at a painting and printmaking department of an arts university in Turkey.
71 Liang *et al.* (2014) used Tenax adsorption tubes with an active sampling flow rate of
72 200 mL min^{-1} , maintained for 10–20 min using a pump, in the interior of a
73 construction sites of an apartment building in Beijing, China. Sax *et al.* (2006) and
74 Loh *et al.* (2007) studied the indoor organic air pollutant emissions and health risks
75 in the United States. Aromatic compounds and formaldehyde were typical of the
76 compounds that were extracted using the site samplers. However, the co-adsorption
77 of multiple compounds on the trap samplers has not been evaluated.

78

79 Solid-phase microextraction (SPME), used to sample gaseous organic VOCs, is
80 environmentally friendly because it is solvent-free and can be reused (Lord, *et al.*,

81 2010). Cheng *et al.* (2011) sampled gaseous BTEXs using divinylbenzene (DVB)
82 adsorbents packed in specially made SPME devices, called needle trap samplers
83 (NTS). They successfully sampled emissions from indoor sources including
84 mosquito coils, incense coil, essential oils, air fresheners, and paint emissions in the
85 work place (Cheng, *et al.*, 2013; Cheng, *et al.*, 2014; Cheng, *et al.*, 2015; Cheng and
86 Lai, 2014; Cheng, *et al.*, 2016; Cheng, *et al.*, 2017). In the present work, NTS are
87 used to measure personal exposure to VOCs during a medical examination at a
88 teaching hospital. For comparison, VOC were actively sampled in the field
89 simultaneously, following the approach of Özden Üzmez *et al.* (2015) The
90 susceptible effect of co-adsorption for the multiple VOCs is evaluated. Finally, a
91 management recommendation, based on the concept of chemical banding, is made.

92

93 **2. Methods**

94

95 *2.1 Gaseous compound extraction using needle trap samplers*

96

97 NTS extract chemical compounds in air through needles by diffusion.

98 Accordingly, a linear gaseous concentration profile ($C(Z)$ in Fig. 1) is obtained along

99 the diffusion path (Z), and the extraction is characterized by the area (A) of opening

100 and the diffusion path length. The total amount (n) of analyte extracted in a time
101 interval (t) is estimated as the following formula (Lord, *et al.*, 2010):

102

$$103 \quad n = D_m \frac{A}{Z} \int C(t) dt \quad (1)$$

104

105 where D_m is the diffusion coefficient of a compound that is sampled by the sorbent in
106 the needle. The quantity (n) of the extracted analyte is assumed to be proportional to
107 the total sample concentration over a time interval ($C(t)$) given a constant D_m , a
108 uniform needle opening (A), and a fixed diffusion path distance (Z).

109

110 ***** *Figure 1 Here* *****

111

112 2.2 Preparation of needle trap sampler

113

114 NTS comprised of a stainless steel needle packed with DVB particles, mesh sizes
115 60–80. DVB particles were packed by aspiration until the desired length was 7 mm
116 and then when the diffusion path was 3 mm long. A very small amount of epoxy glue
117 was applied to the exposed portion of the sorbent layer to immobilize the sorbent
118 particles. To prevent blockage of the NTS epoxy-resin plug, air was drawn

119 continuously through the NTS packing phase as the epoxy cured. Finally, the DVB in
120 an NTS was conditioned by heating at the injection port of the gas chromatography
121 equipped with a flame ionization detector (GC-FID) under 260°C for 30 min.

122

123 The uniformity of the packing phase in an NTS was examined by following the
124 procedures to establish the desired sampling flow rate through an NTS (mL min^{-1}) by
125 drawing through the packing phase using an aspirating pump (Cheng, *et al.*, 2011;
126 Cheng, *et al.*, 2013; Cheng, *et al.*, 2014; Cheng, *et al.*, 2015; Cheng and Lai, 2014;
127 Cheng, *et al.*, 2016; Cheng, *et al.*, 2017). When the relative standard deviations (RSD)
128 of the sampling flow rates across three duplicate tests did not exceed 5%, the packed
129 materials inside the NTS were assumed to be uniformly immobilized. BTEX standard
130 gas samples (around 10 ppm) were prepared in a Pyrex glass bulb, in which the NTS
131 was inserted for 1–2 hrs to extract BTEX. When the RSD of the extracted mass in
132 triplicate tests were less than 5%, the adsorption capacities in the NTS were assumed
133 to be constant.

134

135 2.3 Chemicals, materials and equipment

136

137 Seven centimeter-long, 22 G stainless steel needles (OD 0.71 mm and ID 0.41

138 mm) were purchased from a local company (Herling Co. Ltd., Pingtung, Taiwan) for
139 preparing the NTS, in which DVB particles (Supelco, Bellefonte, PA, USA) were
140 used as adsorbents. Aspirating pumps used to test the NTS sampling flow rates were
141 purchased from Kitagawa (AP-20, Kawasaki, Japan), and epoxy glue was purchased
142 from Nao-Pao Applied Material Co. Ltd. (Taoyuan, Taiwan). All gases (Jing-De Gas
143 Co., Ltd., Kaohsiung, Taiwan), used in chromatographic analysis, were of ultra-high
144 purity.

145
146 Gaseous samples were analyzed using a GC (6890N, Agilent, Wilmington, DE,
147 USA) that was equipped with a FID. The capillary column was HP19091Z-413 HP-1
148 PDMS (30 m × 320 μm × 0.25 μm) (Agilent Technologies, Inc., Wilmington, DE,
149 USA).

152 *2.4 Sampling and analysis*

153
154 The target sampling site was the medical examination center of a teaching
155 hospital in Donggang Township, Pingtung County, Taiwan. This general hospital was
156 established at 1971, and the building has 11 floors. The main medical departments

157 include Medicine, Surgery, Gynecology, Pediatrics, Emergency, Rehabilitation,
158 Dermatology, Ophthalmology, and non-Clinical. The medical examiners collect
159 samples from clinical patients for medical examination. Two highly hazardous
160 solvents that are used in medical examination. Xylene is used for slice staining; and
161 formalin, which contains 40% formaldehyde, is used for fixation before slicing.
162 Figures 2 (a) and (b) show the procedures that are followed by the technicians in the
163 medical examination center. During the staining and fixation procedures, VOCs are
164 emitted. Medical technicians use masks and gloves to prevent VOC exposure.
165 Ventilation hoods are used to reduce indoor VOC emissions. To measure the workers'
166 time-weighted average (TWA) exposure to VOCs, three samplers of the different
167 types were worn simultaneously by medical technicians: (a) passive NTS sampling
168 pens, developed by the authors' group; (b) detector tubes, manufactured by Gastec
169 (Kanagawa, Japan); and (c) personal sampler that comprises a charcoal tube (Method
170 1501 for xylene), or an XAD adsorbent sampler (Method 2541 for formaldehyde)
171 with a personal air pump for active sampling, which was developed by the U.S.
172 National Institute for Occupational Safety and Health (2003). The NTS sampling pen
173 (Fig. 3) is a microminiaturized sampler (with a mass of only 200 g and a length of 20
174 cm), with auxiliary functions of detecting humidity and temperature. The stainless
175 steel needle for extracting VOCs is installed inside the pen.

176

177 ***** *Figure 2 Here* *****

178

179 ***** *Figure 3 Here* *****

180

181 After VOC sampling, an NTS was inserted into the injection ports of the GC-FID,
182 to desorb VOCs for analysis. The desorption time and temperature at the injection
183 port were 30 s and 250 °C. The temperature of the GC was increased from 50 °C in
184 increments of 15 °C min⁻¹ to 180 °C and then held for 2 min. The FID detector was
185 heated to 300 °C. The flow rate of the carrier gas, nitrogen, was 1.2 mL min⁻¹, and the
186 split-off operating mode was used. The following discussion section presents the
187 calibration procedure and the way in which the BTEX concentrations and the personal
188 TWA concentrations to which workers were exposed from the mass that was extracted
189 using NTS. The concentrations of xylene were obtained using the calibration formula
190 in Table 1.

191

192 **3. Results and Discussion**

193

194 *3.1 Determining xylene concentration and confirming co-adsorption of formaldehyde*

195 using needle trap samplers

196

197 According to Eq. (1) (Lord, *et al.*, 2010), the total amount of extracted analyte (n)
198 is proportional to the interval of extraction (t) and the concentration of VOC (C),
199 given that the coefficients D_m , Z and A are fixed. Six concentrations (0, 2, 5, 10, 15
200 and 20 ppm) of xylene were prepared in a Pyrex glass bulb with a volume of around
201 500 mL. The NTS were inserted in series into the bulbs to conduct extraction tests for
202 1 hr. The extracted masses of xylene and the corresponding integrated areas of the
203 peaks that were obtained by GC-FID are used to yield calibration equations in Table 1.
204 The concentrations of xylene in the air samples in the medical examination center are
205 determined using Eq. (2) and from the areas under the GC-FID peaks. If the practical
206 sampling time with an NTS is 8 hrs (480 min), then the measured concentrations of
207 BTEXs must be multiplied by 0.125 (60 min/480 min = 0.125) because the
208 multiplying factor (f) for time weighting is calculated as $f = 60 \text{ min}/(\text{practical}$
209 $\text{sampling time})$.

210

211 Earlier studies have thoroughly investigated the performance of NTS in the
212 adsorptive extraction of aromatic compounds (Cheng, *et al.*, 2011; Cheng, *et al.*, 2013;
213 Cheng, *et al.*, 2014; Cheng, *et al.*, 2015; Cheng and Lai, 2014; Cheng, *et al.*, 2016;

214 Cheng, *et al.*, 2017). Formaldehyde is also emitted in the medical examination center,
215 so an NTS extracted xylene and formaldehyde was tested. Experimental series of
216 xylene (10 ppm) and formaldehyde (2, 10, and 20 ppm) were prepared in the Pyrex
217 glass bulb, and then analyzed in duplicate using GC-FID to compare the area under
218 the peaks of xylene for each other. The mass of xylene using NTS that was extracted
219 using an NTS was not significantly different in the presence of 2–20 ppm
220 formaldehyde. In fact, mixing 10-ppm xylene with 2–20 ppm formaldehyde changed
221 the extracted mass by less than 0.5%. Several works have discussed the individual
222 effects of humidity, temperature, BTEX matrices and atmospheric wind velocity on
223 the SPME extraction of BTEX samplings, and these factors have been found to have
224 no effects on the adsorption of BTEX on common commercial adsorbents such as
225 polymethylsiloxane (PDMS), DVB and carboxen (CAR) (Pawliszyn, 1997; Tsai and
226 Kao, 2006; Schupfer and Huynh, 2008). However, this is the first to discuss the
227 co-adsorption of formaldehyde and xylene by DVB adsorbent.

228

229 3.2 Xylene emissions in the medical examination center

230

231 Three methods were used to sample and examine the TWA exposure of workers
232 to xylene. A detector tube, a passive dosi-tube (No. 122DL, Gastec) that was

233 manufactured by Gastec Corp. was used to examine BTEX TWA concentrations. The
234 detection limit for xylene was 2.13 ppm with a sampling time of 8 hrs. Five tests at
235 the medical examination center revealed no color change so the workers were exposed
236 to xylene TWA concentrations of less than 2.13 ppm.

237

238 Figure 4 shows the TWA concentrations that were detected using passive NTS
239 sampling pens and active charcoal tubes samplers (Method 1501). The differences
240 between all xylene concentrations that were measured using the NTS and Method
241 1501 were in the range 4.2–6.0%, indicating that the use of an NTS is an effective
242 alternative to Method 1501 for the occupational monitoring of indoor gaseous
243 aromatic compounds. However, the passive detector tube is not sufficiently sensitive
244 for examining the exposure of workers to xylene.

245

246 Technicians were exposed to xylene concentrations from 0.11 to 1.2 ppm, which
247 are far below the permissible exposure limit (PEL) 100 ppm (OSHA, 1992). The fume
248 hood (Fig. 2 (b)) satisfactorily protects the technician against emissions of VOCs
249 while he or she fixes slices. The low crack height of the transparent sliding door of the
250 fume hood is essential to keeping the capture speed high enough to reduce the
251 exposure of workers to VOCs.

252

253 *3.3 Formaldehyde emissions at the medical examination center*

254

255 Formalin is used for fixation before slicing in medical examinations. Formalin
256 emits gaseous formaldehyde, and a front side exhaust hood without sliding door (Fig.
257 2(a)) was used to protect technicians from those emissions. Two personal
258 formaldehyde sampling methods, the use of a passive dosi-tube (No. 91D, Gastec)
259 and active sampling Method 2541 (NIOSH, 2003), were simultaneously implemented.
260 No color change occurred in the formaldehyde dosi-tubes, indicating that the 8-hr
261 TWA exposure concentration was less than the detecting limit 0.0625 ppm.
262 Examination by GC-FID, followed by implementation of active sampling Method
263 2541, also yielded an 8-hr TWA formaldehyde exposure concentration of less than the
264 detecting limit 0.05 ppm. In summary, these field-analyzed concentrations of
265 formaldehyde, 0.0625 or 0.05 ppm, are both lower than the PEL of 0.75 ppm (OSHA,
266 1992).

267

268 According to a field examination, the draught air speed 10-cm outside the
269 exhaust hood was 2.2–3.0 m s⁻¹ during the fixation procedures, meeting the

270 operational standard (draught air speed of at least 2.0 m s^{-1}) that was set by the
271 Environmental and Occupational Health Center of the hospital.

272

273 *3.4 Assessing the workers' health risk in the medical examination center*

274

275 In Taiwan, the management of the health risks of workers who are exposed to
276 hazardous chemicals is legislated using the concept of chemical control banding
277 (Department of Labor, Taiwan, 2014). Typically, personal detection concentrations
278 (PDCs) at work and the PELs of significant chemical compounds are used to calculate
279 an Exposure Hazard Ranking (EHR). Table 2 demonstrates the EHRs for xylene and
280 formaldehyde are Rank 1, meaning that the procedures for controlling chemical
281 emissions at the medical examination center were highly successful, because the local
282 ventilation systems are especially effective.

283

284 **4. Conclusions**

285

286 The packed NTS are recommended as personal passive samplers in the medical
287 examination center in this investigation because it has almost the same sampling
288 efficiency as Method 1501 of the NIOSH (2003). In Taiwan, the Ministry of Labor

289 requires that harmful organic solvents and chemicals in a working environment must
290 be air-sampled and analyzed and an official declaration then submitted to the
291 government at least once every six months (Department of Labor, Taiwan, 2016). A
292 multiple functional sampling pen with an NTS should be promoted to be a sampler of
293 organic compounds for routine monitoring purpose. Method 1501 of the NIOSH [21]
294 requires workers to wear a sorbent tube that is connected to a pump to assess
295 exposures to organic chemicals. Workers commonly complain about the
296 inconvenience of carrying this equipment. This work evaluated the use of a mini
297 sampler for the application on occupational health.

298
299 SPME sampling is environmentally friendly. Agnieszka Gałuszka, proposed that
300 the objectives of green analysis technology are the reduction of the use of hazardous
301 substances (Gałuszka *et al.*, 2012). The sampling and analysis of pollutants using
302 NTS require only small amounts of analytical reagents; however, the extraction of
303 VOCs from activated carbon requires large amounts of organic solvents. Notably, the
304 NTS can be reused many times for sampling. The reduplications of sampling and
305 analysis of chemicals using NTS in the work place promote the effective control of
306 risk associated with harmful chemicals as part of a management system that is based
307 on chemical banding.

308

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310

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314

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393 marrow, *J Nanjing Xiaozhuang University* 6: 73–76.

394 **Table 1.** The calibration equation of extracted mass of xylene and the corresponding
395 integral analysis area by GC-FID

396

VOCs	Calibration equations ^a
xylene	$y = 2.6007x - 3.3366, R^2 = 0.9996$ (2)

397 Note:

398 a. Abbreviations in Eq. (2):

399 y = the xylene concentration in ppm; x = the integral area analyzed by GC-FID; and

400 R^2 = the first order linear coefficient of regression.

401

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403 **Table 2.** The EHR of hazardous chemicals assessed in this study.

404

Exposure Hazard Ranking ^{a,b} (EHR)	Chemicals	PDC in sites (ppm)	PEL (ppm)	PDC/PEL ^a (%)
1	Xylene	0.11–1.2	100	0.11–1.2
1	Formaldehyde	0.05–0.0625	0.75	6.7–8.7

405 Note:

406 a. PDC is “personal detection concentrations” and PEL is “permissible exposure
407 limit”. Rank 1: $PDC/PEL < 50\%$; Rank 2: $PDC/PEL \geq 50\%$; and Rank 3: PDC/PEL
408 $\geq 100\%$.

409 b. The strategy of control for chemicals must be preserved as the currently existing
410 approach for EHR Rank 1; and new strategies of control must be considered for
411 EHR Rank 2 and 3 to improve the workers’ exposure of chemicals.

412

413 **Figure Captions**

414

Fig. 1 Schematic needle trap sampler.

Fig. 2 Solvent use during the medical examination: (a) slice staining; and (b) fixation of slice.

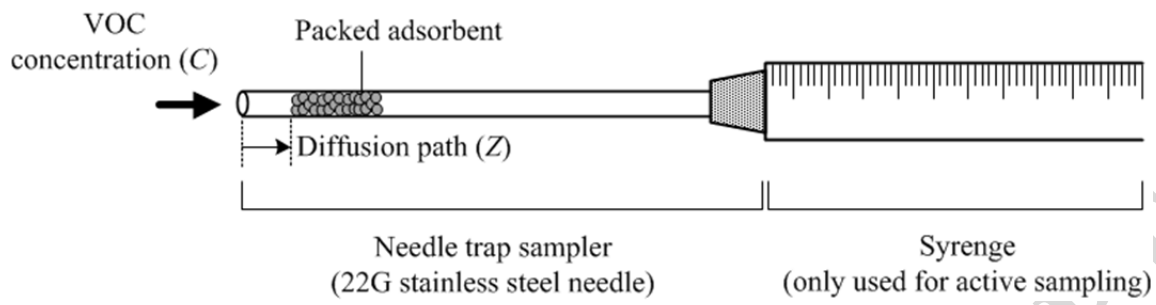
Fig. 3 The multiple functional NTS pens. (a) The appearance and its auxiliary functions, detectors of humidity and temperature. (b) The needle for extracting VOCs is inside the pen and its length is 7 cm. The gas compounds diffuse into the side hole of sampling pen and then are extracted by the NTS.

Fig. 4 The variations of emitted concentrations of xylene at different sampling sequences via personal sampling taken by passive needle trap samplers and active charcoal tubes.

415

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418

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420

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Fig. 1

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423

(a)



424

425

(b)



426

427

428

429

Fig. 2

430



431

432

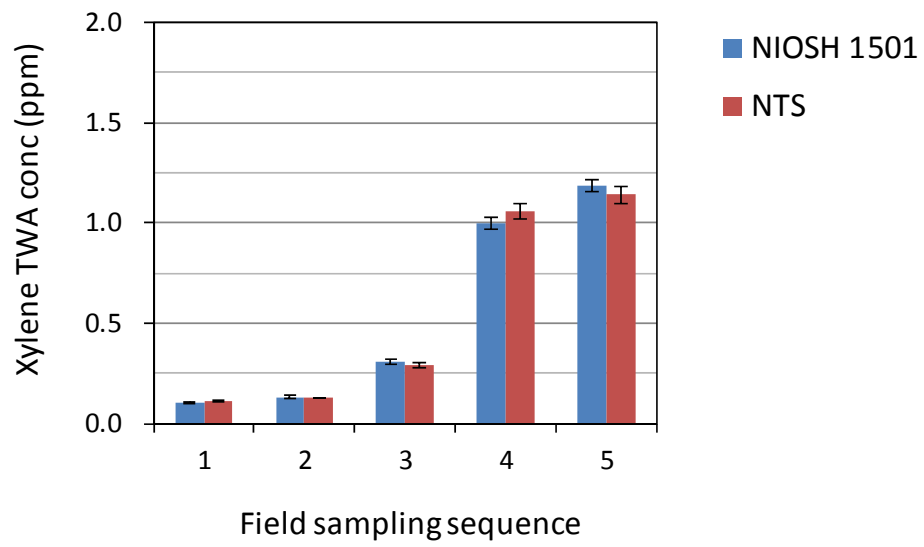
433

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Fig. 3

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437
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Fig. 4

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