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Measurement of 16 volatile organic compounds in restaurant air contaminated with environmental tobacco smoke

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ABSTRACT

Tobacco smoke-related air pollutant levels were studied in ten Finnish restaurants. Markers of tobacco smoke were measured together with other compounds typical of tobacco smoke and indoor air. The measurements were carried out at stationary sampling points in smoking and non-smoking areas of the restaurants in 2005–2006, when at least half of the service area had to be non-smoking according to the Finnish Tobacco Act. The average concentrations (geometric mean, $\mu\text{g}/\text{m}^3$) of the 16 airborne contaminants measured in the smoking area were: nicotine 18.1; toluene 10.6; isoprene 10.2; *m,p*-xylene 5.0; limonene 4.8; benzene 3.3; furfuryl aldehyde 3.2; 1,3-butadiene 2.7; 3-ethenylpyridine (3-EP) 2.5; phenol 2.1; ethyl benzene 1.7; pyridine 1.6; *o*-xylene 1.5; 3-picoline 1.4; styrene 1.2; and naphthalene 0.45. A good correlation ($r = 0.90\text{--}0.99$, $p < 0.001$) was obtained between tobacco-specific markers (3-EP and nicotine) and 1,3-butadiene, isoprene, pyridine, furfuryl aldehyde, 3-picoline, phenol, and styrene. A poor or no correlation ($r = 0.19\text{--}0.60$) was obtained between 3-EP or nicotine and the rest of the compounds. The average concentrations of all compounds were significantly lower in the non-smoking area than in the smoking area ($p < 0.05$). In the non-smoking area, the average concentration of 3-EP was $0.35 \mu\text{g}/\text{m}^3$ and that of nicotine $1.6 \mu\text{g}/\text{m}^3$. In three restaurants, the area design and ventilation were effective: the average level of 3-EP in the non-smoking section was $< 3\%$ from that in the smoking section. In the other restaurants, tobacco smoke was spreading more freely and the corresponding value was 14–76%. A sensitive method was applied for the measurement of airborne 1,3-butadiene. The air samples were collected into Carbopack X adsorption tubes and analysed by thermal desorption/gas chromatography/mass selective detection. The precision of the method was 4.2% (at 100 ng/sample) and the limit of quantification $0.02 \mu\text{g}/\text{m}^3$.

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1. Introduction

Environmental tobacco smoke (ETS) consists mainly of side stream smoke, i.e. the smoke emitted from the smouldering cigarette. ETS contains thousands of compounds, dozens of which are carcinogens (IARC, 2004). In addition to tobacco-specific compounds, such as nicotine and 3-ethenylpyridine, it contains a number of substances that also occur in indoor air and especially in urban atmosphere (Jenkins et al., 2000; Nazaroff and Singer, 2004) as a result of combustion or direct evaporation of different materials. Of the volatile organic constituents in tobacco smoke, these include for instance benzene and 1,3-butadiene (Fowles and Dybing, 2003), both with IARC classification 1 (carcinogenic to humans). Tobacco smoke has been regarded as a notable source of 1,3-butadiene and benzene in indoor air. Both these substances occur also in vehicular exhaust gases (Kim et al., 2001). Benzene is

released also from gasoline by evaporation into ambient air, but 1,3-butadiene does not occur in gasoline (Nordlinder et al., 1996). Other tobacco smoke ingredients that, according to IARC, constitute possible carcinogenic substances (classified as 2B) are naphthalene, styrene, ethyl benzene, and isoprene. Typical compounds in tobacco smoke which have not been classified by IARC as carcinogens are pyridine, toluene, xylene, furfuryl aldehyde, picoline (= methyl pyridine), phenol, and limonene.

Emission factors (emission/cigarette; $\mu\text{g}/\text{cig}$) have been measured for several compounds in side-stream smoke (IARC, 2004; Jenkins et al., 2000), and these can be used in the assessment of the proportion of different compounds in ETS. If the number of cigarettes smoked and the volume and air exchange rate of the room are known, it is possible to evaluate the concentration of a compound in air based on the emission factors and using a simple mass balance model (Bohanon et al., 2003; Ott, 1999). The concentration of a compound in a room ($\mu\text{g}/\text{m}^3$) is the emission rate ($\mu\text{g}/\text{h}$) divided by the air flow rate (m^3/h). However, uncertainty in the results may be caused by the fact that—excluding 3-EP and nicotine—the above substances are not

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specific to tobacco smoke, but can also occur in smoke-free indoor air. Furthermore, there is variation in the emission factors between different tests. Therefore, the actual concentrations are best gained through measurements. Several studies have been published reporting volatile organic compounds (VOC) emissions measured in test conditions, usually the emission rate per cigarette (Bi et al., 2005; Daisey et al., 1998; Hodgson et al., 1996; Löfroth et al., 1989; Martin et al., 1997; Singer et al., 2002, 2003). However, although ETS in the restaurants has been the object of several studies in recent years, few studies have reported VOC concentrations, because their focus has often been on the measurement of particular ETS markers.

The aim of this study was to determine the occurrence of tobacco smoke-related airborne contaminants in restaurants by measuring specific and non-specific tobacco smoke constituents. This study offers additional compound-specific data on micro-environmental concentrations of carcinogens or otherwise harmful components in ETS that complement our former research of markers (Kuusimäki et al., 2007). The applicability of these compounds to tobacco smoke markers was also studied. This study also provided data on whether the non-smoking areas in restaurants were actually smoke-free.

2. Materials and methods

2.1. Restaurants

The field study was carried out in ten restaurants in Helsinki between November 2005 and May 2006. Restaurants with well-separated smoking and non-smoking areas were chosen for the measurements. For comparison, indoor control samples were collected in non-smoking office rooms.

The restaurants, presented in Table 1, were chosen in co-operation with the Finnish Hotel and Restaurant Association. The selected restaurants consisted of five restaurants for dining and socializing, four pubs, and one lunch cafe. The floor areas were 90–300 m² and the number of customer seats ranged from 56 to 180. In six restaurants, the size of the smoking area covered approximately half of the entire customer area, in three about one-third, and in one about one-tenth. During the measurements, the indoor temperature varied from 19 to 24 °C and the relative humidity of air ranged from 12% to 46%.

Six of the restaurants had been established during the amended Tobacco Act and, thus, the requirements on ventilation and smoking areas had been taken into account at the design stage. However, some of these establishments were situated in old buildings with restricting building regulations.

2.2. Field measurements

The compounds studied were measured simultaneously at the smoking and non-smoking areas at stationary sampling points as close to the working area as possible. At both sections, the sampling was carried out at two sampling points.

The measurements were carried out between Tuesday and Friday of 1 week. Each sampling period lasted for 5 h.

The airborne concentrations of tobacco-specific markers, 3-ethenylpyridine and nicotine, were measured in order to detect whether the non-smoking areas were smoke-free and to establish the concentration of ETS in smoking areas. Furthermore, the levels of 14 additional compounds were measured: 1,3-butadiene, isoprene, benzene, pyridine, toluene, furfuryl aldehyde, 3-picoline, ethyl benzene, *m,p*-xylene, *o*-xylene, styrene, phenol, limonene, and naphthalene.

2.3. Sampling

The measurements were carried out using four different samplers. For the purposes of method comparison of the markers (reported elsewhere), three different samplers were used for 3-EP and two for nicotine. 3-EP was collected on 3M (OVM 3500) diffusive sampler; the sampling rate was 24 ml/min. 3-EP and nicotine were collected in charcoal tubes (SKC 226-01) with a pump at a rate of 100 ml/min (smoking section) or 200 ml/min (non-smoking section). The pumps were manufactured by SKC (222 series).

Benzene, pyridine, toluene, furfuryl aldehyde, 3-picoline, ethyl benzene, *m,p*-xylene, *o*-xylene, styrene, phenol, limonene, naphthalene plus 3-EP, and nicotine were collected in Tenax steel tubes (stainless steel, Perkin-Elmer or Supelco, Tenax TA 60/80 mesh, Chrompack; 150 mg/tube) with a pump at 60–70 ml/min.

1,3-Butadiene and isoprene were collected in Carboxpack X steel tubes (stainless steel, Perkin-Elmer or Supelco, Carboxpack X 60/80 mesh, Supelco; 300 mg/tube) with a pump at 50 ml/min. The sampling of 1,3-butadiene was based on a recent method (Martin et al., 2005), and the performance of Carboxpack X tubes (sampling volume, breakthrough and the storage stability) was tested prior to the field measurements.

Performance of the sampling of 1,3-BD was tested by collecting parallel air samples from a parking garage knowing that exhaust gas of the vehicles contains low concentrations of 1,3-BD. The sampling capacity of Carboxpack X tubes was tested for two loads of the adsorbent (200 or 300 mg/tube) and by using two consecutive tubes. Air was sucked by pumps (SKC 222 series) at 50–60 ml/min for 5–6 h.

2.4. Analysis of 3M and charcoal tube samples

3-EP was desorbed from 3M samplers and charcoal tubes using pyridine/toluene solution (15%) and analysed by a gas chromatograph/mass spectrometer (GC/MSD) (Vainiotalo et al., 2001; Kuusimäki et al., 2006). For the diffusive method, the limit of quantification (LOQ) of 3-EP was 0.5–1 ng/sample corresponding to a concentration of 0.07–0.14 µg/m³ for an air sample of 7.2 l. For the charcoal tube method, the LOQ values were 1 ng/sample (3-EP) and 8 ng/sample (nicotine) corresponding to air concentrations of 0.02 and 0.1 µg/m³ for a sample of 60 l.

2.5. Analysis of Tenax and Carboxpack X samples

The VOCs and 1,3-BD plus isoprene were analysed using thermal desorption sample injection (TurboMatrix 650, Perkin-Elmer) connected to a gas chromatograph (6890N, Agilent Technologies) and a mass spectrometer (5973 inert MSD, Agilent Technologies). The conditions during the analysis are indicated in Table 2. For the above compounds, a thermal desorption method using mass selective detection was chosen due to its sensitivity and selectivity. The calibration samples for the VOC analysis were prepared by injecting 1 µl of the calibration mixture

Table 1
Information on restaurants and measurements

Time of measurement		Restaurant code	Restaurant type	Business started (year)	Renovations (year)	Opening hours	Customer seats (n)	Floor area (m ²)	Serving staff (n)	Staff/shift (n)	Smoking area ^a	T (°C)	RH (%)
Day	Time												
3.11.05	13.30–18.30/th	1	DS	2002	2002–2005	10.30–02	80	150	10	2	1/3	22	46
22.11.05	9–14/tu	2	LC	2003	–	7–15	56	100	2	2	1/3	19	23
14.12.05	15–20/we	3	DS	1913	2006	11–22 (24)	80	300	6	2–3	1/10	22	24
2.2.06	15–20/th	4	DS	1991	2004	11–22	70	150	8	1–4	1/2	22	19
8.2.06	11–16/we	5	DS	2004	–	11–23	180	250	10	2–7	1/3	21	12
5.5.06	11–16/fr	6	DS	2002	–	11–23	150	250	7	2–4	1/2	23	31
2.2.06	15–20/th	7	pub	1991	–	11–24	80	90	5	1–2	1/2	24	18
22.3.06	11–17.30/th	8	pub	1974	–	9–24	108	150	5	1–2	1/2	21	17
30.3.06	15–20/th	9	pub	2005	2005	14–02	120	200	6	1–4	1/2	22	30
10.5.06	15–21/we	10	pub	2003	2003	15–01	88	200	3	1–2	1/2	23	29

DS = dining and socializing, LC = lunch cafe, T = air temperature, RH = relative humidity of air, n = number of subjects/parameters.

^a Section of the entire customer area.

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