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Tobacco-Specific and Volatile N-Nitrosamines in Environmental Tobacco Smoke of Offices

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Key Words

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Abstract

The tobacco-specific nitrosamines (TSNA) N-nitrosornicotine (NNN) and 4-(N-methylnitrosamino)-1-(3-pyridyl)-1-butanone (NNK) as well as the volatile N-nitrosamines N-nitrosodimethylamine (NDMA) and N-nitrosopyrrolidine (NPYR) were determined in the indoor air of a poorly ventilated office where extensive smoking took place. Under these conditions, moderate to heavy smoking resulted in indoor air concentrations of 7.6–20.0 ppm carbon monoxide, 109–290 ppb nitrogen oxides and 58.5–132 µg/m³ nicotine. The N-nitrosamine concentrations were (in nanograms/cubic meter): not detected (nd)–6.0 NNN, nd–13.5 NNK, 7.9–45.0 NDMA and 3.5–27.0 ng/m³ NPYR. Occupation of the office by a nonsmoker for 1 h would result in a maximum exposure to 0.01 µg TSNA and 0.03 µg volatile N-nitrosamines.

Introduction

Tobacco-specific nitrosamines (TSNA), such as N-nitrosornicotine (NNN), N-nitrosoanatabine, N-nitrosoanabasine and 4-(N-methylnitrosamino)-1-(3-pyridyl)-1-butanone (NNK) have been found in cured tobacco, and in mainstream smoke and sidestream smoke of various tobacco products [1]. We have recently developed an analytical method sensitive enough to determine TSNA in environmental tobacco smoke (ETS) [2], and, in the meantime, our preliminary findings have been confirmed by Brunnemann et al. [3] who have reported the presence of 8.3 and 9.3 ng/m³ NNK in the indoor air of a smoke-polluted bar.

In this paper, we report indoor air concentrations of NNN and NNK determined in a poorly ventilated office

in which extensive smoking occurred. Data are also reported for the presence of the two volatile N-nitrosamines N-nitrosodimethylamine (NDMA) and N-nitrosopyrrolidine (NPYR). As a surrogate marker for ETS, the nicotine concentration is presented together with data for carbon monoxide (CO) and nitrogen oxides (NOx) in office air.

Materials and Methods

Study Design

Air monitoring was conducted in an office (84 m³) located on the ground floor of a new laboratory in Vienna. The room was not air-conditioned and was furnished with standard office fittings. Indoor air was sampled for 2 h on 14 different occasions, during which the office was occupied by up to 5 people (smokers and nonsmokers). No

Table 1. TSNA and other tobacco smoke constituents measured in indoor air of an office with restricted ventilation

Smoking activity in each 2-hour session			Tobacco burnt ¹ g/h	Concentration in indoor air of tobacco smoke components						
cigs.	pipes	cigars		nicotine µg/m ³	NNN ng/m ³	NNK ng/m ³	NDMA ng/m ³	NPYR ng/m ³	CO ppm	NOx ppb
11	—	1	5.0	132.0	1.7	0 ²	27.2	11.6	12.5	110
18	—	—	7.2	127.0	0	1.6	13.2	4.9	13.0	157
12	1	—	8.3	117.6	3.1	3.1	13.9	5.9	12.4	182
7	1	1	6.9	101.7	2.9	4.7	23.0	8.0	11.4	133
13	1	—	8.7	100.4	4.9	2.1	7.9	5.2	11.0	140
7	1	—	6.3	93.7	6.0	4.2	14.0	27.0	9.0	194
11	—	—	4.4	89.8	1.6	4.4	22.0	13.0	7.7	109
8	1	—	6.7	87.8	2.6	8.0	25.2	10.3	20.0	290
15	—	—	6.0	84.1	3.8	10.8	9.0	4.5	14.0	192
6	1	—	5.9	72.3	2.3	4.9	36.0	11.0	8.7	110
11	—	—	4.4	69.8	3.5	13.5	8.2	3.5	7.6	125
11	—	—	4.4	64.2	1.4	2.8	12.8	6.8	10.4	120
10	1	—	7.5	61.9	3.8	4.5	NA ³	NA	13.0	172
7	1	—	6.3	58.5	1.6	3.3	45.0	18.1	10.7	175

¹ Approximate tobacco weights: Cigarette tobacco = 0.8 g; pipe tobacco = 1.2 g; cigar tobacco = 7 g.

² Limit of detection 0.1 ng/m³ for NNN and NNK.

³ NA = Not analyzed.

attempt was made to influence the smoking habits and working activity of the occupants, who were free to leave and enter the office at any time. However, it was not permitted to open the windows during the smoking sessions which would have resulted in improved air quality. The number of people present, as well as the times when cigarettes, cigars and pipes were lighted, were recorded. The sampling apparatus was located close to the center of the office and installed at the breathing height of a seated person.

Analysis of ETS

All measurements were performed continuously over each 2-hour session during which smoking occurred. Nicotine was sampled on Extrelut columns (E. Merck; Darmstadt, BRD) acidified with 0.1 M HCl using a flow rate of 2.4 l/min. After sampling, the column was made alkaline by drawing through gaseous ammonia and nicotine eluted with ethyl acetate. Nicotine was determined by gas chromatography using a nitrogen-specific detector. Volatile N-nitrosamines and TSNA were sampled by drawing air through a series of 2 wash bottles containing aqueous phosphate/citrate buffer (pH 3.5) and ascorbic acid to inhibit artifactual formation of N-nitrosamines. After sampling, the buffer was extracted with methylene chloride and the extract separated by chromatography (basic aluminum oxide; Woelm II-III, Eschwege, BRD) into two fractions containing volatile nitrosamines and TSNA. Nitrosamine analysis was performed by capillary gas chromatography with detection using a thermal energy analyzer (TEA Model 502; Thermo Electron Corp., Waltham, Mass., USA) according to a published method [2]. The presence of N-nitrosamines was confirmed by rechromatography after photolysis of the sample at 365 nm [4]. Direct on-line measurement was performed for CO (Carbon Monoxide Analyzer Model 8310; Monitor Labs Inc., USA) and NOx (Nitrogen Oxide Analyzer Model 8840; Monitor Labs Inc., USA).

Results

The time-intergraded office air concentration of tobacco smoke constituents together with the amount of tobacco burned are shown in table 1. On average, 6.3 g/h of tobacco (range 4.4–8.7 g/h) was smoked during the 14 sessions. As a surrogate marker of ETS, the mean nicotine concentration was 90.1 µg/m³ (range 58.5–132 µg/m³). Mean concentrations of CO and NOx were 11.5 ppm (7.6–20.0) and 158 ppb (109–290), respectively. The mean nitrosamine concentrations were (in nanograms/cubic meter): 2.8 [not detected (nd)–6.0] NNN, 4.9 (nd–13.5) NNK, 19.8 (7.9–45.0) NDMA and 10.0 (3.5–27.0) NPYR. No correlation was observed between individual components of ETS and the amount of tobacco burnt. This may be due to differences in the types of tobacco products smoked and also to the different ventilation conditions on the various occasions, indicating that none of the components measured acts as a suitable marker of total exposure to ETS.

Discussion

The data for TSNA are slightly lower than those of Brunneemann et al. [3] who reported concentrations of 8.3 ng/m³ NNN and 9.3 ng/m³ NNK in ETS of a 'moder-

ately smoke-polluted' bar. However, comparison of the results is impeded by the lack of information on the actual ETS concentration (or an appropriate surrogate for it) in the bar. The presence of NNN and NNK in ETS is solely due to tobacco smoke. However, volatile nitrosamines occur ubiquitously in the environment [5], and concentrations of 10–90 ng/m³ have been reported in ambient air of industrial areas [6]. As a result, the presence of NDMA and NPYR in indoor air cannot be considered to be specifically due to ETS.

Subjects (both smokers and nonsmokers) present in the office during air sampling complained about the bad air conditions. This allows the conclusion that, normally, the air quality would have been improved by opening the windows. In fact, the nicotine concentration in the indoor air ranged between 58.5 and 132 µg/m³ (mean 90.1), which is considerably higher than the reported concentrations of < 50 µg/m³, most frequently < 20 µg/m³, in offices with unrestricted ventilation [7–10]. As a result, the NNN and NNK concentrations measured in our study exceed the concentrations found under realistic conditions, and present a 'worst-case' situation.

From the present data, occupation of the office by a nonsmoker for 1 h would result in a maximum exposure of 0.01 µg/h TSNA (NNN and NNK) and 0.03 µg/h volatile N-nitrosamines (NDMA and NPYR). These exposure

levels are based on the highest data values shown in table 1, they assume a respiratory volume of 0.5 m³/h with total retention of all N-nitrosamines, and are quite different from those reported by Hoffmann et al. [11] in a previous paper, who estimated a total intake of up to 0.3 µg/h NNN and NNK on exposure to ETS, based on calculations of TSNA concentrations in sidestream cigarette smoke [12]. If the exposure from actual ETS concentrations is calculated for real-life conditions [7–10], the uptake amounts to only 0.004 µg TSNA and 0.015 µg volatile N-nitrosamines.

The measurements show that ETS contains 5–15 ng/m³ NNN and NNK, 2 nitrosamines specific to tobacco smoke. Assuming an ETS exposure time of 3 h/day [13], the daily exposure to N-nitrosamines from ETS amounts to < 0.1 µg combined TSNA and volatile N-nitrosamines. This exposure to N-nitrosamines is considerably smaller than the measured average daily intake of 0.2–0.3 µg of volatile N-nitrosamines (NDMA, NPYR and N-nitrosopiperidine) [14], and an estimated total daily dietary intake of 10–100 µg for all dietary N-nitrosamines [15]. ETS exposure provides only a small contribution to the total daily N-nitrosamine exposure, which has to be taken into consideration when calculating any carcinogenic risk due to ETS exposure.

References

- Hoffmann D, Adams JD, Brunnemann KD, Hecht SS: Assessment of tobacco-specific N-nitrosamines in tobacco products. *Cancer Res* 1979;39:2505–2509.
- Begutter H, Klus H, Ultsch L: Kapillargaschromatographische Bestimmung flüchtiger und tabakspezifischer N-Nitrosamine mittels des Thermo Energy Analyzers. *J Chromatogr* 1985; 321:475–479.
- Brunnemann KD, Cox JE, Hoffmann D: Methods of analysis for tobacco-specific-N-nitrosamines in indoor air (poster presentation). *Nitroso Compounds: Biological Mechanisms, Exposures and Cancer Etiology*, Kailua Kona, Nov 1991.
- Fiddler W, Doerr RC, Piotrowski EG: Observations on the use of thermal energy analyzer as a specific detector for nitrosamines; in Walker EA, Castegnaro M, Gričič L, Lyle RE (eds): *Environmental Aspects of N-Nitroso Compounds*. IARC Scientific Publications No 19. Lyon, International Agency for Research on Cancer, 1978, pp 33–39.
- Tricker AR, Spiegelhalter B, Preussmann R: Environmental exposure to preformed nitroso compounds. *Cancer Surv* 1989;8:251–272.
- Akkan Z, Preussmann R, Spiegelhalter B: N-Nitrosamines in ambient air of industrial areas in Germany. *J Cancer Res Clin Oncol* 1991; 117:S14.
- Muramatsu M, Umemura S, Okada T, Tomita H: Estimation of personal exposure to tobacco smoke with a newly developed nicotine personal monitor. *Environ Res* 1984;35:218–227.
- Hammond SK, Leaderer BP, Roche AC, Schenker M: Collection and analysis of nicotine as a marker for environmental tobacco smoke. *Atmos Environ* 1987;21:457–462.
- Proctor CJ, Warren ND, Bevan MAJ: Measurements of environmental tobacco smoke in an air-conditioned office building. *Environ Technol Lett* 1989;10:1003–1018.
- Coultas DB, Samet JM, McCarthy JF, Spengler JD: A personal monitoring study to assess workplace exposure to environmental tobacco smoke. *Am J Public Health* 1990;80:988–990.
- Hoffmann D, Adams JD, Brunnemann KD: A critical look at N-nitrosamines in environmental tobacco smoke. *Toxicol Lett* 1987;35:1–8.
- Hoffmann D, Brunnemann KD, Adams JD, Hecht SS: Formation and analysis of N-nitrosamines in tobacco products and their endogenous formation in consumers; in O'Neill IK, van Borstel PC, Miller CT, Long J (eds): *N-Nitroso Compounds: Occurrence, Biological Effects and Relevance to Human Cancer*. IARC Scientific Publications No 57. Lyon, International Agency for Research on Cancer, 1984, pp 743–762.
- Letzel HW, Johnson LC: The extent of passive smoking in the Federal Republic of Germany. *Prev Med* 1984;13:717–729.
- Tricker AR, Pfundstein B, Theobald E, Preussmann R, Spiegelhalter B: Mean daily intake of volatile N-nitrosamines from foods and beverages in West Germany in 1989–1990. *Food Chem Toxicol* 1991;11:729–732.
- Tricker AR, Preussmann R: Carcinogenic N-nitrosamines in the diet: Occurrence, formation, mechanisms and carcinogenic potential. *Mutat Res* 1991;259:277–289.