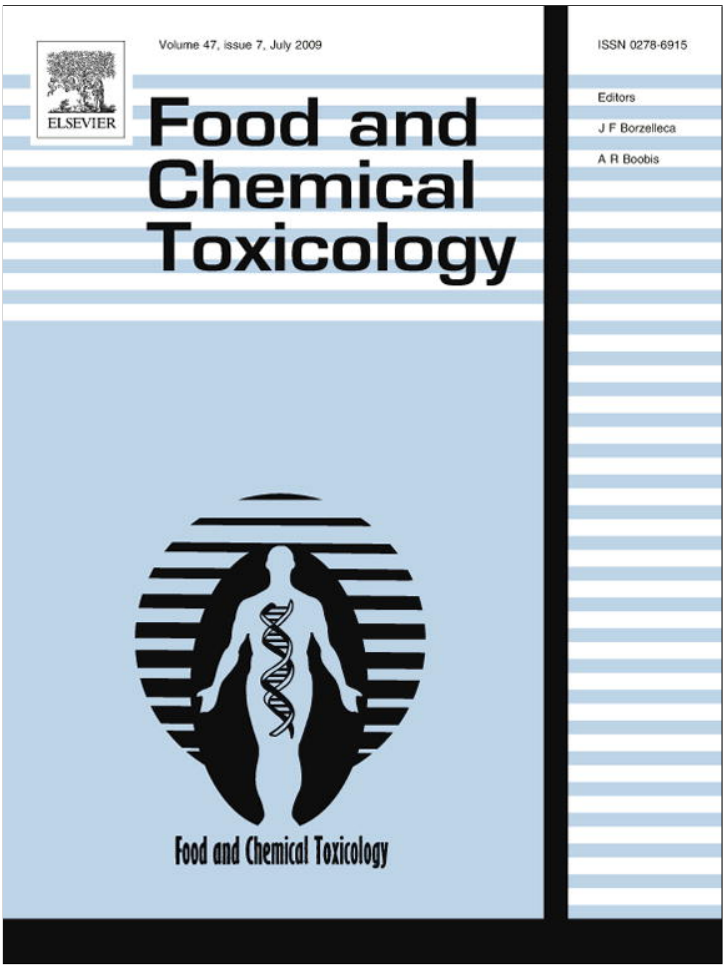

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Indoor air contamination during a waterpipe (narghile) smoking session

Hermann Fromme [a,*], Silvio Dietrich [a], Dieter Heitmann [b], Holger Dressel [c], Jürgen Diemer [b], Thomas Schulz [d], Rudolf A. Jörres [c], Knut Berlin [a], Wolfgang Völkel [a]

[a] Department of Environmental Health, Bavarian Health and Food Safety Authority, Veterinärstrasse 2, D-85764 Oberschleissheim, Germany
[b] Bavarian Environment Agency, Buergermeister-Ulrich-Strasse 160, D-86179 Augsburg, Germany
[c] Institute and Outpatient Clinic for Occupational, Social and Environmental Medicine, Ludwig-Maximilians-University, Ziemssenstrasse 1, D-80336 München, Germany
[d] Federal Institute for Risk Assessment, Thielallee 88-92, D-14195 Berlin, Germany

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ABSTRACT

The smoke of waterpipe contains numerous substances of health concern, but people mistakenly believe that this smoking method is less harmful and addictive than cigarettes.

An experiment was performed in a 57 m^3 room on two dates with no smoking on the first date and waterpipe smoking for 4 h on the second date. We measured volatile organic compounds (VOC), polycyclic aromatic hydrocarbons (PAH), metals, carbon monoxide (CO), nitrogen oxides (e.g. NO), as well as particle mass (PM), particle number concentration (PNC) and particle surface area in indoor air.

High concentrations were observed for the target analytes during the 4-h smoking event. The median (90th percentile) values of $PM_{2.5}$, PNC, CO and NO were 393 (737 μg/m^3), 289,000 (550,000 particles/cm^3), 51 (65 ppm) and 0.11 (0.13 ppm), respectively. The particle size distribution has a maximum of particles relating to a diameter of 17 nm. The seven carcinogenic PAH were found to be a factor 2.6 higher during the smoking session compared to the control day.

In conclusion, the observed indoor air contamination of different harmful substances during a WP session is high, and exposure may pose a health risk for smokers but in particular for non-smokers who are exposed to ETS.

1. Introduction

It is well documented over the last decades that tobacco smoking is related to diverse major health threats resulting in ~440,000 deaths each year in the US alone, cost of ~$157 billion in annual health-related economic losses and results in more than 5.6 million years of potential life lost each year (US-DHHS, 2004).

Environmental tobacco smoke (ETS), partly similar to the mainstream smoke inhaled by the smoker, is a complex mixture containing thousands of chemicals, many of which are known to be carcinogens. There is convincing evidence from numerous experimental and epidemiological studies that secondhand smoke causes elevations in lung cancer, cardiovascular diseases and is responsible for other severe health effects such as bronchial asthma. Consequently, ETS was also classified as a known human carcinogen (IARC, 2004; US-DHHS, 2006).

An alternative to cigarette smoking that has been used for at least four centuries especially in Africa and Asia was waterpipe smoking, also known as narghile, argileh, hubble-bubble, shisha, goza or hooka (WHO, 2005). Generally, charcoal heats the very moist and often flavoured tobacco which produces the smoke containing tobacco and charcoal combustion products inhaled by the user once it has passed through a bowl filled with water. During a waterpipe smoking session, smokers are exposed to more smoke over a longer period of time than for cigarette smoking, because of the longer duration of such a session and the higher respiratory volume inhaled.

Traditionally, waterpipe smoking is common in Mediterranean and Arabian countries, reaching prevalences of between 19% and 30% with an increasing tendency in recent years (Chaaya et al., 2004; Asfar et al., 2005; Labib et al., 2007; El-Roueiheb et al., 2008; Maziak et al., 2008a; Almerie et al., 2008). Waterpipe smoking has a higher social and cultural acceptance than cigarette smoking. Therefore, many WP smokers mistakenly thought that this smoking habit is a social entertaining practice, done in a group, leading to a more social behaviour and relaxation (Chaaya et al., 2004; El-Roueiheb et al., 2008; Smith-Simone et al., 2008a). With increased smoking intensity, the social element will be replaced by a more individual use of WP leading to increased tobacco dependency (Maziak et al., 2004). A very high proportion of the users (~50–75%) in these countries believed that the waterpipe smoking is less harmful or safe and less addictive than cigarette smoking (Maziak et al., 2004; Asfar et al., 2005; Labib et al.,

* Corresponding author. Tel.: +49 8931560231; fax: +49 8931560835.
E-mail address: hermann.fromme@lgl.bayern.de (H. Fromme).

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2007; Jawaid et al., 2008). The same conviction was observed in surveys from western countries (e.g. Ward et al., 2007; Smith-Simone et al., 2008b).

In a study of users of a waterpipe café and an internet forum in the US, 19% and 41% reported a daily and weekly waterpipe use, respectively (Smith-Simone et al., 2008a). These results were confirmed in another US cross-sectional internet-based survey of university students (Eissenberg et al., 2008). In a first report of the prevalence of waterpipe smoking in an Arabic-speaking Australian population, 11.4% of the participants used WP (Carroll et al., 2008).

Up to now, the situation in Europe is not well documented but there are indications that waterpipes became more common in the last years. A cross-sectional survey of 937 British students showed that 38% had tried waterpipes, and cigarette smoking was the major risk factor for being a regular waterpipe smoker (Jackson and Aveyard, 2008). First results from a German representative survey of 3602 persons (12–19 years of age) in 2007 concluded that 31% of the participants used a waterpipe in the last year, and 14% in the last month (BzgA, 2007).

The health consequences of waterpipe use are not well evaluated at present, but some studies showed "short-term effects" on the cardiovascular system (Al-Kubati et al., 2006) and on pulmonary functions (Al-Fayez et al., 1998; Kiter et al., 2000) mainly due to the high carbon monoxide content. Furthermore, there is preliminary evidence that waterpipe smoking is related to an increased cancer risk [summarized in Maziak et al. (2004), Knishkowky and Amitai (2005)] and harm the fetus when done in pregnancy (Nuwayhid et al., 1998). Compared with cigarette smokers, waterpipe users receive equal or higher doses of nicotine, which causes the same or higher risk of tobacco dependency as cigarettes do (Maziak et al., 2004).

Therefore, the objective of this pilot study was to characterize the exposure situation of waterpipe and secondhand smokers by measuring (1) the indoor air concentrations of substances known as typical tobacco smoke ingredients, and (2) substances that are harmful to human health.

The target analytes were: volatile organic compounds (VOC), polycyclic aromatic hydrocarbons (PAH), elements and metals, carbon monoxide (CO), nitrogen oxides (NO_x), as well as different particle characteristics such as particle mass, particle number concentration and particle surface area. Overall, this pilot study was performed to give first insights into substances which may be relevant to characterize the ETS exposure occurring in waterpipe smoking sessions. The data will be relevant to develop a systematic strategy to characterize the exposure and body burden of smokers and secondhand smokers. The data should be used to design a more appropriate and large-scaled study.

2. Materials and methods

2.1. Study description and sampling site

The study was carried out in a room in an office building in a small town near Munich, Germany. The size of the room was 20 m^2 and the volume was 57 m^3. The measurements were taken on two days in August 2008 over the same time period. On the first day (control day), the air was monitored without any smoking activities in the room and, on the day after, with a waterpipe smoking session. Before the measurements, the room was thoroughly ventilated, and the windows were kept closed during the measurement periods. During the experiment four persons smoked one waterpipe from 10 am to 2 pm. In detail, the charcoal in the waterpipe was first ignited at 10 am and four consecutive sessions were carried out, each took ~40 min (see also Fig. 1). During the short period between the four consecutive sessions new charcoal was ignited outside the room and placed in the waterpipe together with new tobacco.

We used a common waterpipe which was ~5 years old and 8 g of a regular used tobacco sold from a retail shop. Moreover, the tobacco was finally moisturized just before the smoking session with glycerol (propane-1,2,3-triol).

The equipment for sampling and monitoring was placed on a table in the middle of the room about 1 m above floor level and ~1–2 m away from the waterpipe.

2.2. Particle mass, particle number concentration

Continuous measurements of particle mass (e.g. PM_{10}, $PM_{2.5}$) were made using an optical laser aerosol spectrometer (LAS) (Dust monitor 1.108, Grimm Technologies Inc., Ainring, Germany). This spectrometer works by constantly drawing the air sample via a volume controlled pump (1.2 L/min.) through a flat beam of laser light. All scattered signals generating while the particles cross this beam are detected with a high speed photo diode, analysed by an integrated pulse height analyzer and counted. The LAS measures particle concentrations in 16 nominal size bins from about 0.1 to 2.5 μm. For our purposes the continuous measurements were stored minute-by-minute on a data logger.

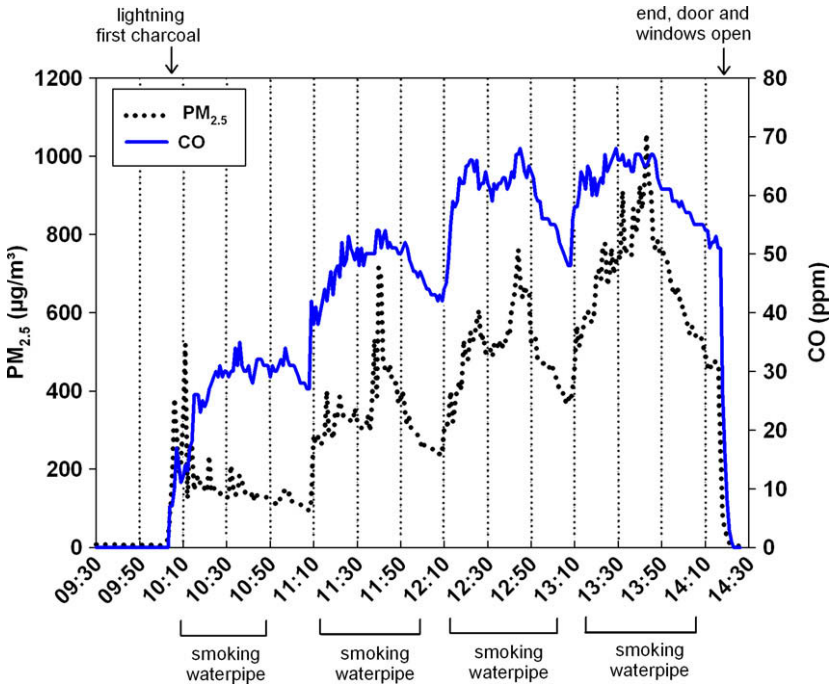

Fig. 1. Results of continuously measured $PM_{2.5}$ and CO concentrations during the four waterpipe smoking sessions.

Particle number concentrations (PNC) were measured using a Wide Range Aerosol Spectrometer (WRAS, Grimm Technologies Inc., Ainring, Germany). The WRAS consists of two particle counters and sizers: The GRIMM 5.400 SMPS + C and the GRIMM 1.108 LAS (described before), both combined via software to one unit. The GRIMM 5.400 consists out of a high resolution particle counter (CPC) attached to a "Vienna Type" electrostatic classifier (M-DMA). The complete WRAS covers the range from 0.005 to 20 μm.

2.3. Particle surface area

Airborne particle surface was measured continuously by Nanosurface Area Monitor (NSAM) model 3550 (SMPS, TSI Inc., Shoreview, MN, USA). The NSAM provides a method for monitoring the human lung-deposited area of particles (expressed as $\mu m^2/cm^3$) corresponding to alveolar regions of the lung. The NSAM is based on diffusion charging of sampled particles in the size range from 0.01 to 1.0 μm, followed by detection of charged aerosol using an electrometer.

2.4. Volatile organic compounds

Indoor air samples were collected for 4 h during the waterpipe smoking period with a constant flow of 0.08 L/min with Tenax GR as adsorbent in the first tube and Chromosorb 106 in the second tube and analysed using a thermodesorption unit (Gerstel, Muelheim, Germany) coupled to a gas chromatograph/mass spectrometer (GC/MS; gas chromatograph 6890A coupled to MSD 5973 N, Agilent, Waldbronn, Germany). The limit of detection (LOD) for a single compound was 0.05 $\mu g/m^3$ using a sample volume of 20 L. The total concentration of volatile organic compounds (TVOC) was determined as toluene equivalents according to the international guideline ISO 16000-6. The total peak area of the total ion chromatogram in the retention time window between *n*-hexane and *n*-hexadecane was quantified using the response factor of toluene obtained by external calibration. TVOC values as toluene equivalents are semiquantitative since single compounds in the sample may have response factors that may widely deviate from that of toluene.

2.5. Polycyclic aromatic hydrocarbons

Gaseous and "particle-bound PAH" were determined by collecting indoor air with a medium volume sampler equipped with a sampling unit consisting of a $PM_{2.5}$-inlet, a quartz fibre filter and a polyurethane foam. The sampling period was 4 h concurrent with the waterpipe sessions. Filter and PU foam was extracted with toluene after addition of deuterated PAH standards, purified on a silica column and analysed by GC/MS. Naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(*a*)anthracene, chrysene, benzo(*b*)-fluoranthene, benzo(*k*)-fluoranthene, benzo(*a*)pyrene, dibenzo(*a*,*h*)anthracene, indeno(1,2,3-*cd*)pyrene, and benzo(*g*,*h*,*i*)perylene were determined as single PAH compounds. The LOD was 0.1 ng/m^3 using a sample volume of 10 m^3.

2.6. Metals/elements

Samples were collected on 47 mm quartz fibre filters (Pieper, Bad Zwischenahn, Germany) using a medium volume sampler equipped with a $PM_{2.5}$ sampler as sample inlet operated at a constant flow of 2.3 m^3/h for a 4-h sampling period concurrent with the waterpipe smoking period. Before using, the filters from the same production lot were analysed for their heavy metal blank values. After a closed-vessel microwave decomposition of the filter samples using nitric acid and hydrogen peroxide as oxidising agents, the target analytes were measured using inductively coupled plasma-mass spectrometry (ICP-MS).

2.7. Nitrogen oxide/carbon monoxide

Nitrogen oxides were measured using a CLD 700 AL analyzer (ECO PHYSICS AG, Munich, Germany). Sample gas is drawn into the analyzer, mixed with internally-produced ozone, divided in two equal streams and given into separate reaction chambers. NO and NO_x were detected photoelectrically by chemiluminescence. In order to assure reliability of its results the analyzer has a calibration module for the zero level and the NO reference gas. The analyzer monitored sequential NO and NO_x while NO_2 was calculated from the difference. Every 30 s data with a lag time of less than a second and a minimal detectable concentration 0.05 ppb were produced.

Carbon monoxide was measured using a Testo analyzer (TESTO AG, Lenzkirch, Germany).

2.8. Parameters of indoor climate

Indoor carbon dioxide was measured using a continuously monitoring infrared sensor (Testo 445, Germany). The instrument was programmed for a 1-min data logging interval and was averaged over the 4-h period during the waterpipe smoking experiment. Calibration of the CO_2 sensors included linearity checks at four concentrations (0, 350, 1000, 2000 ppm) during the weeks prior to sampling. Indoor humidity and temperature were measured simultaneously with a separate sensor connected to the Testo instrument.

3. Results

The results of particle measurements, inorganic gases and the basic climate parameters during a 4-h smoking session and on the control day are presented in Table 1. In Fig. 1 the results of $PM_{2.5}$ and CO readings during the smoking experiment are given. Overall, all parameters were higher on the smoking day than on the day without smoking. Very high mass-related concentrations were observable with mean (90th percentile) values of 393 $\mu g/m^3$ (737 $\mu g/m^3$) for $PM_{2.5}$. Furthermore, the particle number concentration (PNC) and the particle surface area also reached high median values of 97,000 particles/cm^3 and 1517,000 $\mu m^2/cm^3$, respectively.

Fig. 2 depicts the results of the particle number concentrations for different size classes during the first of the four consecutive waterpipe smoking sessions. When the charcoal of the waterpipe was first lit at 10:04 am, we observed a very high PNC concentration for some minutes probably caused by lighting the charcoal. At this time, the maximum of the particle diameter was nearly 100 nm. This is followed by a period in which the waterpipe was smoked. It could be seen from the graph that during this time period smaller particles with a diameter of 17 nm dominated the particle number concentrations.

With regard to the inorganic gases, we observed measurements on the second day that were much higher than on the first (control) day, reaching maximum values of 0.07 and 0.14 ppm for NO and NO_2 and up to 68 ppm for CO (Table 1). While the levels of NO_2

Table 1
Distribution parameters of particulate matter, inorganic gases and climate parameters in indoor air during a 4-h smoking session (values in brackets are in the same room with no smoking activities).

	10th Percentile	Median	90th Percentile	Mean
Particles				
$PM_{1.0}$ ($\mu g/m^3$)	121 (2)	384 (2)	721 (2)	396 (2)
$PM_{2.5}$ ($\mu g/m^3$)	125 (3)	393 (4)	737 (5)	406 (4)
PM_{10} ($\mu g/m^3$)	150 (6)	422 (12)	760 (19)	432 (13)
Particle number concentration (N/cm^3)	138,000 (4600)	289,000 (6200)	550,000 (7100)	319,000 (6000)
Particle surface area ($\mu m^2/cm^3$)	24,349,000 (344,000)	96,813,000 (1517,000)	188,096,000 (2587,000)	101,916,474 (1487,000)
Anorganic gases				
NO (ppm)	0.05 (<0.05)	0.11 (<0.05)	0.13 (<0.05)	0.10 (<0.05)
NO_2 (ppm)	0.05 (<0.05)	0.06 (<0.05)	0.07 (<0.05)	0.06 (<0.05)
CO (ppm)	27 (<1)	51 (<1)	65 (<1)	48 (<1)
Climate parameters				
Temperature (°C)	28 (28)	30 (29)	31 (30)	30 (29)
Relative humidity (%)	50 (48)	58 (53)	69 (53)	59 (50)
CO_2 (ppm)	990 (1185)	3776 (1319)	6405 (1385)	3752 (1295)

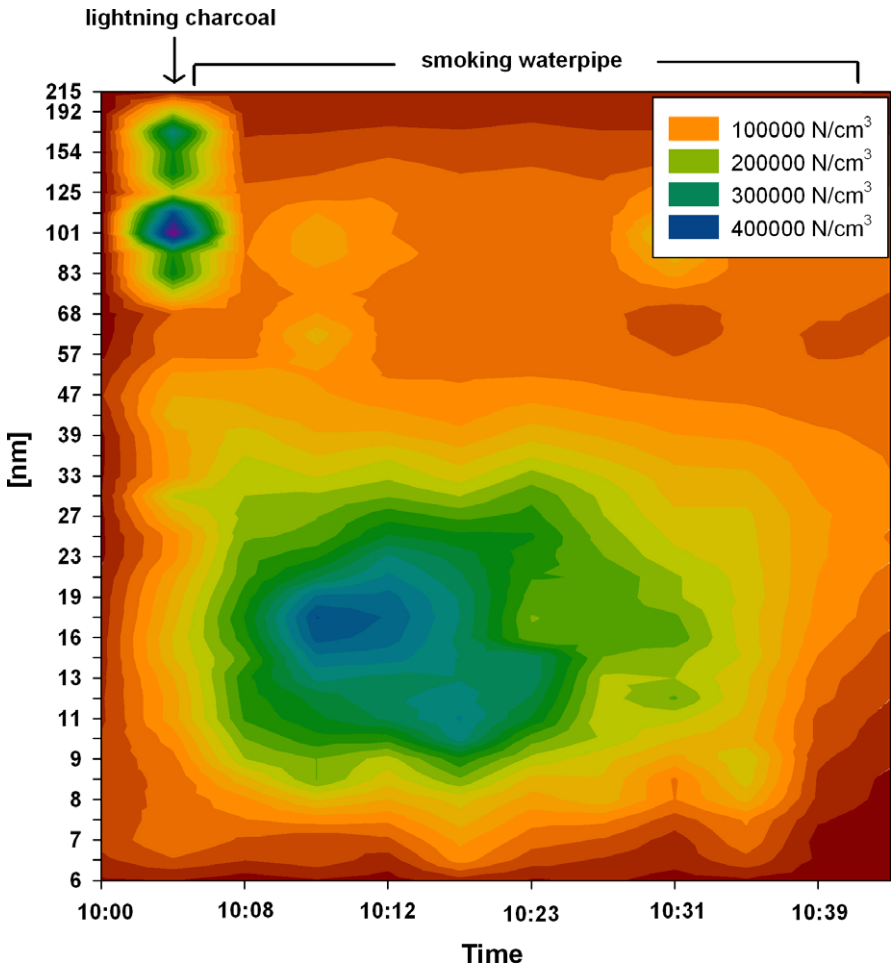

Fig. 2. Particle number concentration (N/cm^3) by size classes (electrical mobility diameter) during the first waterpipe smoking session.

moved slightly above the detection limit for the whole smoking session right from the beginning, the NO-values showed only a slow increase reaching a plateau after 2 h. The same tendency was monitored with regard to CO. CO readings during the smoking experiment are given in Fig. 1.

Table 2 summarizes the results for different organic substances. A distinct increase was especially found for benzene (0.11 vs. 15.0 μg/m^3), 2,5-dimethylfuran (<0.05 vs. 8.8 μg/m^3), nicotine (<0.05 vs. 18.0 μg/m^3) and total volatile organic compounds (730 vs. 1800 μg/m^3) when comparing both 4-h sampling periods. The sum of all measured 16 gaseous and particle-bound PAH were approximately twice as high during the smoking session, compared with the prior control session. This total concentration of PAH was dominated by the more volatile naphthalene, phenanthrene, acenaphthene and fluorene. With regard to the seven PAHs classified as probable carcinogens by the US-EPA, the concentration increased from 1.86 ng/m^3 during the nonsmoking session to 4.86 ng/m^3 during the smoking session.

The concentrations for elements and metals found in the indoor air showed a significant increase for various elements, especially the rare-earth elements lanthanum and cerium (Table 3). The concentrations of these elements in the indoor air increased by a factor of ~500 during the smoking session. This observation is in agreement with measurements conducted in a previous study on smoking activities (Bolte et al.,2008), whereas concentrations in outdoor air are usually well below 0.5 ng/m^3 for lanthanum and below 1 ng/m^3 for cerium. Our findings indicate that lanthanum and cerium can be used as sensitive and specific tracers for smoking intensity.

For the toxic and potentially carcinogenic elements cadmium, lead, arsenic and thallium, a significant increase was observed. We found concentrations in both study periods of <0.1 vs. 0.38 ng/m^3, <3 vs. 11.2 ng/m^3, <0.2 vs. 0.35 ng/m^3 and <0.1 vs. 1.14 ng/m^3, respectively.

4. Discussion

Our study was organized against the background of the recent debate on smoke-free legislation in Germany and the lack of indoor air quality data during waterpipe smoking. By analysing different particulate and gas-phase constituents and components of indoor air, we found significant levels of toxic and/or carcinogenic substances in this environment during a smoking event.

Some studies characterized the constituents of mainstream waterpipe smoke showing that higher levels of metals (e.g. arsenic, nickel, cobalt, chromium and lead), carbon monoxide, "Tar", nicotine, polycyclic aromatic hydrocarbons and aldehydes could be observed compared to cigarette smoke (Shihadeh, 2003; Monn et al., 2007; Shihadeh and Saleh, 2005; Sepetdjian et al., 2008; Monzer et al., 2008; Al Rashidi et al., 2008).

One interesting result of these studies was the higher proportion of fluoranthene, pyrene and phenanthrene (Sepetdjian et al., 2008; Monzer et al., 2008) or chrysene, fluoranthene and phenan-

Table 2
Concentrations of organic substances during a waterpipe session and the day before without any smoke exposure (4-h averaging time).

Substance	No smoking	During smoking session
Volatile organic compounds (VOC) (μg/m³)		
Acetonitrile	<0.05	<0.05
Acrylnitrile	<0.05	<0.05
Benzene	0.11	15.0
2,5-Dimethylfuran	<0.05	8.8
3-Ethenylpyridine	<0.05	<0.05
Nicotine	<0.05	18.0
TVOC	730	1800
Polycyclic aromatic hydrocarbons (PAH) (ng/m³)		
Naphthalene	36	110
Acenaphtylene	4.2	11
Acenaphthene	34	42
Fluorene	13	23
Phenanthrene	40	66
Anthracene	2.3	4.3
Fluoranthene	4.0	5.3
Pyrene	4.2	5.0
Benz(*a*)anthracene[a]	<0.1	0.18
Chrysene[a]	0.21	0.47
Sum of benzo(*b*)- and benzo(*k*)-fluoranthene[a]	0.46	1.40
Benzo(*a*)pyrene[a]	0.45	0.61
Indeno(1,2,3-*cd*)pyrene[a]	0.41	1.90
Dibenzo(*a*,*h*)anthracene[a]	0.28	0.30
Benzo(*g*,*h*,*i*)perylene	0.47	3.50
Sum of B2-PAH	1.86	4.86
Sum of all PAH	140	270

[a] Classified by US-EPA as probable human carcinogen (B2) based on sufficient evidence of carcinogenicity in animals.

Table 3
Descriptive statistics of metals for a 4-h session smoking a waterpipe compared with a control session with no smoking (in ng/m³).

Substance	No smoking	During smoking session
Al	<200	<200
As	<0.2	0.35
Bi	<0.05	0.112
Ca	<2000	<2000
Cd	<0.1	0.38
Ce	<0.2	129
Co	<0.5	<0.5
Cr	<10	<10
Cu	<10	<10
K	<1000	3435
La	<0.1	63.1
Mg	<100	114
Mn	<5	7.78
Mo	<10	<10
Na	<1000	48,576
Ni	<5	<5
Pb	<3	11.2
Sb	<2	<2
Sn	<5	<5
Ti	20.9	<20
Tl	<0.1	1.14
V	<10	<10
Zn	<500	<500

threne (Shihadeh and Saleh, 2005) found in waterpipe mainstream smoke, compared with cigarettes. This is consistent with our results demonstrating high concentrations of phenanthrene in indoor air. Even though the PAH concentrations were dominated by the more volatile PAH, we observed concentrations of probable carcinogenic PAH during the session in indoor air which were 2.6 times higher than on the control day.

Moreover, the higher concentrations of metals and CO found in mainstream waterpipe smoke by the aforementioned researchers were consistent with our findings. Our CO mean concentration of 48 ppm over the 4-h experiment nearly doubled the threshold limit value (TLV) of 25 ppm for an 8-h working shift in the US (ACGIH, 1994). Furthermore, the findings considerably exceeded the air quality guidelines set by the WHO (for example 25 ppm for 1 h of exposure and 10 ppm for 8 h) (WHO, 2000).

To our best knowledge this study is the first measurement of various organic and inorganic substances and particles during a waterpipe session showing high concentrations of smoking-related target analytes.

Up to now, only results from two other studies were available presenting data for particulate matter in indoor spaces. In the first indoor air study, the concentration of waterpipe smoke was quantified using a continously operating aerosol monitor (Maziak et al., 2008b). In their study, $PM_{2.5}$ and PM_{10} was measured during a laboratory (room space: ~34 m^3) session in which 20 individuals used a waterpipe and 20 others smoking a cigarette. The mean $PM_{2.5}$ background concentrations and $PM_{2.5}$ levels during waterpipe smoking were 48 and 264 $\mu g/m^3$ with maximum values up to 908 $\mu g/m^3$. Smoking a cigarette leads to similar average concentrations in the test room. The second study measured $PM_{2.5}$ in eight German waterpipe cafés in 2007 using the same equipment as in our study (BA Kbg, 2007). The measuring time of 30–60 min in each location resulted in mean $PM_{2.5}$-levels between 51 and 2727 $\mu g/m^3$.

In conclusion, the observed indoor air contamination of different harmful substances during a waterpipe session is high, and exposure may pose a health risk for smokers but in particular for secondhand smoke exposed non-smokers. The results indicated that the exposure level is similar to that found in German hospitality locations where smoking is allowed (Bolte et al., 2008). More research on this topic is urgently needed, but by now the public and especially the policymakers should be more informed about the health consequences of this smoking practice.

Conflict of interest statement

The authors declare that there are no conflicts of interest.

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