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Firefighters' multiple exposure assessments in practice

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HIGHLIGHTS

- Multiple exposures via multi-route exposure should be taken into account.
- PAHs in the air should be linked to the excretion level of 1-hydroxypyrene.
- The HI-method was a valuable tool for calculating the additive effects of chemicals.
- The Mixie uncovered reasons why future risk assessments should be very cautious.

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ABSTRACT

During the past decade, more research has focused on firefighters' multiple exposures via multi-route exposure. Multi-route exposure can alter the kinetics of chemicals; this has brought changes to the recommendations on biomonitoring. In addition, the possibility that the chemicals in smoke have additive and synergistic effects has not been consistently taken into account. In this study, biomonitoring and occupational hygienic measurements were used to determine smoke diving trainers' exposure to smoke in conventional and modern simulators. Biological action limit values (BALs) for 1-hydroxypyrene, linked with the ratio of pyrene to benzo[a]pyrene, were established for conventional and modern simulator types. The additive and synergistic effects for the main compounds detected in the air during the suppression of a fire were also calculated. According to the biomonitoring results, dermal exposure played a role in exposure to polycyclic aromatic hydrocarbons (PAHs), and it seemed to delay the excretion of 1-hydroxypyrene and 1-naphthol. The calculated BALs for 1-hydroxypyrene were 6 nmol/L and 53 nmol/L for the conventional and modern simulators, respectively. The combined cancer and eye disorders or upper respiratory tract irritation effects of volatile organic compounds (VOCs) in the conventional simulator were from 6.5 to 7.0-fold higher than in the modern simulator.

1. Introduction

Firefighters are potentially exposed to countless numbers of gaseous and vaporous agents and also ultrafine particles during the suppression of a fire. Chemical asphyxiants such as carbon monoxide, hydrogen cyanide, and hydrogen sulfide have been found to cause acute symptoms (Materna et al., 1992; Savolainen and Kirchner, 1998). Hydrogen chloride, nitrogen oxides, and sulfur dioxide have been found to be responsible for acute irritation effects (Bolstad-Johnson et al., 2000; Pośniak, 2000). Agents having long-term effects, such as benzene, polycyclic aromatic hydrocarbons (PAHs), formaldehyde, and 1.3-butadiene, have also been found in the air during the suppression of a fire (Feunekes et al., 1997; Caux et al., 2002; De Vos et al., 2006). In addition, smoke particles in the ultrafine range have been implicated as a risk factor for cardiovascular diseases and some studies suggest a possible association with lung cancer (Edelman et al., 2003; Swiston et al., 2008; IARC, 2010; Ward et al., 2010).

During the past decade, more attention has been paid to multiple exposures via multi-route exposure. Multi-route exposure has been demonstrated to change the kinetics of chemicals, which has posed a great challenge to occupational health care professionals in finding the right sampling time for biomonitoring tests (Feunekes et al., 1997; Caux et al., 2002; Laitinen et al., 2010). Besides being able to assess the additive, synergistic, and antagonistic effects of all of the chemicals present in smoke during the suppression of a fire, comprehensive information is needed about the interactions of these chemicals (Alarie, 2002). Although the evidence concerning multiple exposures is still being reviewed, it has been postulated that multiple exposures explain firefighters' increased mortality and

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morbidity as compared against controls (Kirchner and Savolainen, 1998; LeMasters et al., 2006; Golga and Weistenhofer, 2008).

In this study, we tested strategies for assessing firefighters' multiple exposures to 15 different PAHs and for interpreting the results of firefighters' biomonitoring after multi-route exposure to PAHs in different smoke diving simulators. A few useful techniques for evaluating the additive effects of volatile organic compounds (VOCs) and the synergistic effects of chemicals found during suppression of fire were also tested.

2. Materials and methods

2.1. Test persons

Male smoke diving trainers ($n = 13$) from the Emergency Services College served as the test group. The exposure tests were carried out in conventional (11 test persons) and modern (2 test persons) simulators. Smoke diving trainers performed three smoke dive tasks per day in both simulator types. Each smoke dive lasted an average of 30 min and the purpose of their dives was to monitor the fire fighting students' training. The first test was done immediately in the morning (09:00–10:00), the second just before lunch (10:00–11:00) and the last after lunch (13:00–15:00). There were three test days for the conventional simulator and one day for the modern. All test days followed the same action pattern. The test persons stayed in the rest room between exposure tests and they took away fire suits and personal protective equipment for that time. The trainers used full-faced masks and breathing air was taken from compressed air bottles (Dräger). In addition, they wore fire fighting suits (Bristol and Brage), and were all non-smokers.

2.2. Simulators

The burning materials in the conventional simulator were chipboard, conifer plywood, or pure spruce wood. Kerosene, solvent naphtha or ethanol was used as the igniting liquid. The conventional simulator was a large block of flats (three floors) made from concrete and the test situation was as similar to a real fire as possible.

In the modern simulator (gas simulator) tests, the burning material was propane and the artificial smoke was made with a smoke generator using mineral oil. The simulator was constructed from metal and had been built on two different floors. The size of the simulator was less than 50 m^2 per floor. The test situation was artificial and the smoke behaved in a totally different way than it would in a real fire.

2.3. Methods

2.3.1. Biomonitoring

Biomonitoring and occupational hygienic measurements were used to determine the trainers' exposure to chemicals. To be able to measure the trainers' total exposure, their urinary 1-hydroxypyrene (Jongeneelen et al., 1987) and 1-naphthol (Keimig and Morgan, 1986) excretion was measured. According to general sampling guidelines for the biomonitoring of 1-naphthol and 1-hydroxypyrene, urine samples should be taken immediately after the work shift if inhalation exposure is the main exposure route. In this study, inhalation exposure was not thought to be the main exposure route because the firefighters used full-faced masks and air was taken from compressed air bottles. A more comprehensive sampling strategy was therefore needed. The first urine sample was taken before exposure in order to check the background concentrations, the second sample was taken as recommended by the general guidelines, and the third and fourth samples were taken 6 h after exposure and the next morning to reveal the possible delay in excretion patterns. The term exposure in this case means exposure during the whole day. The urinary 1-hydroxypyrene was detected in samples from 13 test persons after four test days and urinary 1-naphthol was detected in samples from 5 persons after two test days. The difference in the number of samples taken was due to the limited budget of the study.

2.3.2. Occupational hygienic measurements

The concentrations of PAHs and VOCs were measured at stationary sites inside the simulators during training. Fifteen different PAH-compounds; naphthalene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene and dibenz[a,h]anthracene were collected to XAD-2 adsorbents and glass-fiber filters and analyzed using high-performance liquid chromatographic (HPLC) separation and multiple-wavelength shift fluorescence detection (Mäkelä and Pyy, 1995). VOCs were collected by pumps to Tenax TA adsorbents and analyzed according to ISO 16000 Standard using GC–MS technique (ISO Standard, 2004). Six PAHs and five VOCs samples were collected from the conventional simulator and two of each sample were collected from the modern simulator. Sampling times for VOCs varied from 20 min to 58 min. The same figures for the PAHs varied from 43 min to 188 min.

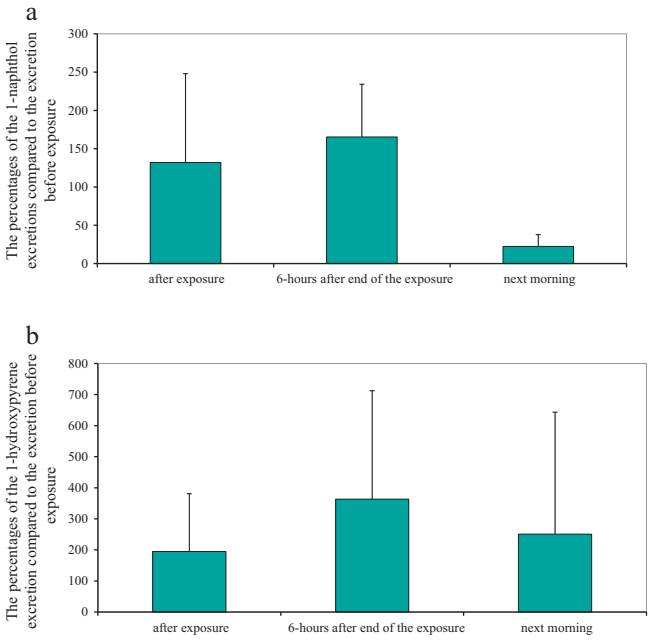

Fig. 1. (A) The percentages of the firefighting trainers' urinary 1-naphthol excretions (mean + S.D.) at the time points immediately after exposure, 6 h after the end of exposure, and the next morning compared to excretion before exposure (B) The percentages of the firefighting trainers' urinary 1-hydroxypyrene excretions (mean + S.D.) at the time points immediately after exposure, 6 h after the end of exposure, and the next morning compared to excretion before exposure.

3. Results and discussion

3.1. Effect of multi-route exposure on the kinetics

1-Naphthol has been used as a biomonitoring indicator of exposure to shorter-chain PAHs (Bieniek, 1997; Laitinen et al., 2010). Moreover, 1-hydroxypyrene has been used as a biomonitoring indicator of exposure for longer-chain PAHs (Jongeneelen, 1992, 2004; Bouchard and Viau, 1999; Pavanello et al., 2000; Laitinen et al., 2010). Both biomonitoring methods are used for the exposure assessment of PAHs.

After the urine sample results from both simulators were combined, the highest average 1-naphthol excretion was detected 6 h after the end of exposure ($n = 5$ firefighters). The excretion was 170% of the excretion before exposure (Fig. 1a). The same pattern was found for the excretion of 1-hydroxypyrene, for which excretion was 360% of the excretion before exposure ($n = 13$ firefighters, Fig. 1b). The firefighters' excretion of 1-naphthol and 1-hydroxypyrene before exposure was below or very near the limits of the unexposed population in Finland (30 mmol/L for 1-naphthol and 3 nmol/L for 1-hydroxypyrene). The firefighters' average 1-naphthol and 1-hydroxypyrene concentrations after training in the conventional simulator were 135 mmol/L and 5 nmol/L, respectively. The same results after training in the modern simulator were 65 mmol/L for 1-naphthol and 1.2 nmol/L for 1-hydroxypyrene (under the limit of 1-hydroxypyrene for the unexposed population in Finland).

3.2. Setting the BAL (biological action limit) for 1-hydroxypyrene

1-Hydroxypyrene is only an indicator of exposure to pyrene and benzo[a]pyrene and it is not responsible for the toxic effects of these compounds. In addition, benzo[a]pyrene is 1000-fold more toxic than pyrene; this has to be taken into account somehow in the risk assessment of PAHs. Table 1 shows the list of toxicity equivalency factors for common PAHs compared to benzo[a]pyrene (Collins

Table 1
Toxicity equivalency factors (TEF) and IARC classification for polycyclic aromatic hydrocarbons (PAHs).

PAHs	TEF	IARC classification	Update year
Naphthalene	0.001	2B	2002
Acenaphthene	0.001	3	2010
Fluorene	0.001	3	2010
Phenanthrene	0.001	3	2010
Fluoranthene	0.001	3	2010
Pyrene	0.001	3	2010
Chrysene	0.01	2B	2010
Benzo[ghi]perylene	0.01	3	2010
Benz[a]anthracene	0.1	2B	2010
Benzo[b]fluoranthene	0.1	2B	2010
Benzo[k]fluoranthene	0.1	2B	2010
Benzo[a]pyrene	**1**	**1**	**2012**
Indeno[1,2,3-cd]pyrene	0.01	2B	2010
Dibenz[a,h]anthracene	**1**	**2A**	**2010**

According to Collins et al. (1998) and IARC (2012).
Group 1: The agent is carcinogenic to humans.
Group 2A: The agent is probably carcinogenic to humans.
Group 2B: The agent is possibly carcinogenic to humans.
Group 3: The agent is not classifiable as to its carcinogenicity to humans.

et al., 1998). The IARC classifications of PAHs are also listed (IARC, 2012).

In 2010 the Finnish Institute of Occupational Health established a BAL value of 12 nmol/L for 1-hydroxypyrene (Priha et al., 2010) and this new value is based on research studies conducted in coke ovens (Pyy et al., 1997). This value represents the ninetieth percentile of all analyzed 1-hydroxypyrene samples taken in Finland immediately after work shifts in 2009. According to the studies in coke ovens the average pyrene versus benzo[a]pyrene ratio in the air has been 2.5 (Pyy et al., 1997; ACGIH, 2005). Because the ratios of pyrene versus benzo[a]pyrene varies at different workplaces, ACGIH has given a formula (1) for the recalculation of new values for unexposed populations for risk evaluation purposes in various environments (ACGIH, 2005).

$$\text{New value}_{\text{BAL}} = (12\,\text{nmol/L}) \times \left[\frac{(\text{ratio of pyrene/BaP})}{2.5}\right] \quad (1)$$

This information was also applied for calculations of the corresponding BAL value for 1-hydroxypyrene by using the measured ratios of pyrene versus benzo[a]pyrene in the air of conventional and modern simulators. The current Finnish BAL-value was used as the base value. The ratios of pyrene versus benzo[a]pyrene were 11 for the modern simulators and 1.3 for the conventional simulators (Fig. 2a and b). According to the formula (1) the firefighters' exposure to the mixture of pyrene/BaP (ratio = 11) after training in the modern simulator, should be evaluated against the BAL-value 53 nmol/L of 1-hydroxypyrene. On the other hand the firefighters' exposure to the mixture of pyrene/BaP (ratio = 1.3) after smoke diving in the conventional simulators should be evaluate against the BAL-value 6 nmol/L. The highest single concentration of 1-hydroxypyrene 6 h after the end of the training in conventional simulators was almost 3-fold higher than the new proposed value. The corresponding value after trainings in the modern simulator was under the limit of the unexposed population.

3.3. The evaluation of the additive effects of VOCs

VOCs are a large group of different volatile organic compounds having various effects. The Hazard index method is one possible tool for calculating the additive effects of VOCs (U.S. EPA, 1986; ATDSR, 2004). It is essential to remember that the Hazard index method can only be used for compounds which have a similar effect or target organ. In this example, the method was used for calculation of the additive effects of the most important measured

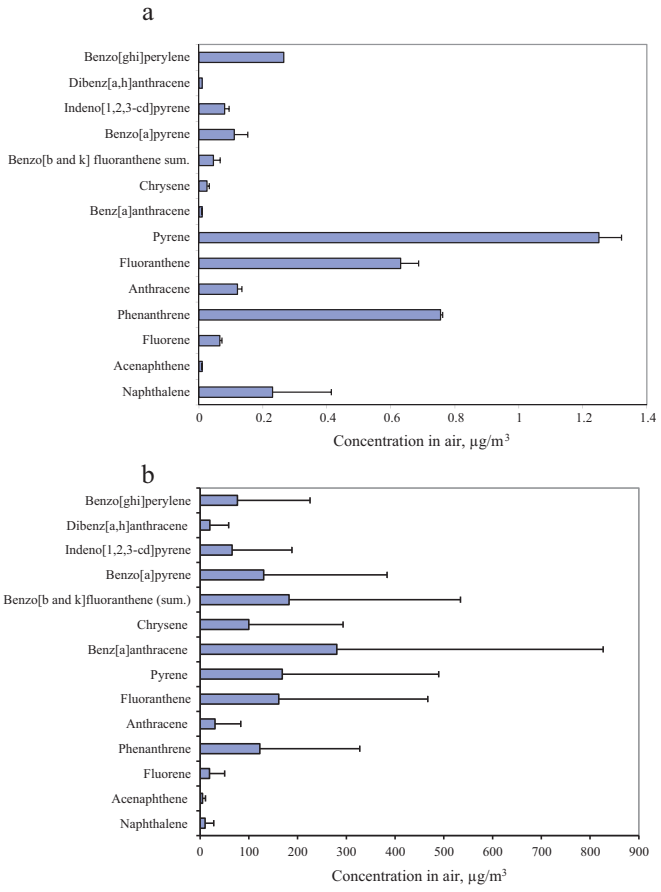

Fig. 2. (A) The profile of polycyclic aromatic hydrocarbons (PAHs) in the air of a modern simulator. (B) The profile of PAHs in the air of a conventional simulator.

volatile organic compounds (12 compounds) in eye disorders or upper respiratory tract irritation and for calculation of cancer risk. The Hazard index method follows an Eq. (2) which is the sum of the following fractions.

$$\frac{C_1}{\text{STEL}_1} + \frac{C_2}{\text{STEL}_2} + \frac{C_3}{\text{STEL}_3} \ldots < 1, \quad (2)$$

In this equation, C denotes the airborne concentration of the component and STEL denotes the Finnish short term exposure limit value (15 min time-weighted average) for the same component. Multiple exposures to VOCs at the workplace should not exceed the sum value of 1.

The additive effect of cancer risk was calculated for benzene, solvent naphtha group 1, solvent naphtha group 2, styrene and ethylbenzene. The solvent naphtha group 1 means aliphatic and alicyclic hydrocarbons, which have a boiling point from 0 to 120 °C. Group 2 means aromatic, aliphatic and alicyclic hydrocarbons, which have a boiling point from 150 to 220 °C.

The additive effect of eye disorders or upper respiratory tract irritation was calculated for benzene, 2-butanone, phenol, solvent naphtha group 1, solvent naphtha group 2, styrene, toluene, ethylbenzene, 2-propanol, xylene, acetone and ethanol. The classifications of chemicals according to effects were done using the MIXIE-program, which will be introduced in the next section.

The additive effect of cancer risk for the above mentioned compounds was 0.39 + 0.21 (Hazard index) in the working environments of the conventional simulator and 0.06 + 0.01 in the working environment of the modern simulator (Table 2). The additive risk for eye disorders and upper respiratory tract irritation

Table 2
The additive effects of the most important VOCs in conventional and in modern simulators.

Simulators	Conventional simulator			Modern simulator		
	Concentration, μg/m³	Additive effect		Concentration, μg/m³	Additive effect	
Compounds	Mean ± SD	Carcinogenic effect	Eye disorders/Upper respiratory tract irritation	Mean ± SD	Carcinogenic effect	Eye disorders/Upper respiratory tract irritation
Benzene	1200 ± 610	0.376	0.376	180 ± 21	0.054	0.054
Phenol	430 ± 410		0.021	49 ± 1		0.002
Solvent naphtha, group 2	2400 ± 3000	0.012	0.012	240 ± 0	0.001	0.001
Solvent naphtha, group 1	2800 ± 3600	0.006	0.006	630 ± 200	0.001	0.001
Toluene	370 ± 270		0.001	120 ± 0		0.000
Styrene	230 ± 130	0.001	0.001	29 ± 3	0.000	0.000
2-Butanone	170 ± 130		0.001	18 ± 4		0.000
Ethylbenzene	60 ± 40	0.000	0.000	27 ± 1	0.000	0.000
2-Propanol	70 ± 20		0.000	13 ± 1		0.000
Xylene[a]	18 ± 27		0.000			0.000
Acetone	76 ± 52		0.000			0.000
Ethanol[a]	180 ± 160		0.000			0.000
Hazard indices		0.39 ± 0.21	0.42 ± 0.23		0.06 ± 0.01	0.06 ± 0.01

[a] These chemicals have supra-additivity properties. The human studies indicate that ethanol ingestion reduces the metabolism of xylene, which can lead to the potentiation of the neurotoxic effects associated with xylene.

was 0.42 + 0.23 in the conventional and 0.06 + 0.01 in the modern simulator.

3.4. Additive and synergistic effects of chemicals

Contaminants with similar effects on the same organs or organ systems of the human body should be taken into account by adding them together rather than considering them individually. Other interactions, such as synergy and antagonism, should also be taken into account. Adolf Vyskocil of the University of Montreal and Daniel Drolet of the Institut de recherche Robert-Sauvé en santé et en sécurité du travail in Quebec led the project that developed the MIXIE program for evaluating multiple exposure. This program contains information on about 700 chemicals, their effects, the mechanisms of their effects, and their target organs. This database also contains information about the additive and synergistic effects of these chemicals. The effects of these chemicals are categorized into 32 different classes. This program is freely available on the Internet at http://www.irsst.qc.ca/en/_outil_100037.html.

The most potentially hazardous chemical agents (carbon monoxide, naphthalene, benzene, benzo[a]pyrene, solvent naphtha, hydrogen chloride, hydrogen cyanide and formaldehyde) detected in the air during the normal suppression of a fire were entered into MIXIE. The program found five combinations where additive effects were possible and suggested two combinations where synergistic effects might be possible. The first combination of additive effects was cancer risk; this category contained the following chemicals: solvent naphtha group 1 and group 2, benzene, formaldehyde, and benzo[a]pyrene. The second combination of additive effects was central nervous system disorders; solvent naphtha group 1 and group 2 and carbon monoxide fell into this category. The third group of additive effects was the eye disorders combination; solvent naphtha group 1 and group 2, naphthalene, hydrogen chloride, and formaldehyde fell into this category. The chemicals in the fourth group were solvent naphtha group 1 and group 2, hydrogen cyanide, hydrogen chloride, and formaldehyde, all compounds irritating the upper respiratory tract. The fifth group consisted of chemicals that are potential disruptors of oxygen transport; this category included carbon monoxide, hydrogen cyanide, and naphthalene.

There were also two notes about possible synergistic effects. The first one was about effects between carbon monoxide and hydrogen cyanide. An additive effect for mortality and synergy for physical incapacity has been found in two short-term rat studies. Both findings were recorded at exposure levels exceeding the short-term exposure limit (STEL) for carbon monoxide. In the absence of other studies, the program was very careful in its statement; it recommended considering the first-level analysis. The second suspicion of synergistic effects was between formaldehyde and hydrogen chloride. Two studies evaluating the interaction in nasal cancer, done in the same laboratory, were identified. In the first study, the substances were not individually tested. In the second study, the authors did not report any influence of hydrogen chloride on the cancer induced by formaldehyde. In the absence of other studies, the program was again very careful and made the same recommendation as for carbon monoxide and hydrogen cyanide. However, these examples showed that risk assessments for these chemicals still face many uncertainties. Until consistent information is available, all evaluations of the synergistic effects of these chemicals should take the worst possible scenario into account.

4. Conclusion

Firefighters' multiple exposures via multi-route exposure should be taken into account in the evaluation of exposure. Dermal exposure plays a role in exposure to PAHs during smoke diving, as was revealed by the excretion patterns of 1-hydroxypyrene and 1-naphthol. In order to measure dermal exposure, a second urine sample should be taken 6 h after the exposure has ended. To improve risk assessment, the profile of PAHs in the air should be measured and linked to the excretion level of 1-hydroxypyrene. In this study, it was essential to know the ratio of pyrene versus benzo[a]pyrene in the air in order to carry out risk assessment for PAHs. The ratios of pyrene versus benzo[a]pyrene in the air were on average 1.3 in the conventional simulator and 11 in the modern simulator, which recommends using BAL-values 6 nmol/L and 53 nmol/L in the respective simulators for risk evaluations with these PAH-mixtures. Firefighters are therefore exposed to a more toxic mixture of PAHs in conventional simulators than in modern simulators. This example described how occupational hygienic measurements can support the interpretation of biological monitoring results and risk assessments. In the future the challenging task will be how to take other PAHs into account in the interpretation of biomonitoring results. Hopefully TEFs linked to benzo[a]pyrene can bring some kind of solution for this problem.

The Hazard indices method was a valuable tool for calculating the additive effects of chemicals, although this method had its

limitations. The combined cancer and eye disorders or upper respiratory tract irritation effects of volatile organic compounds (VOCs) in the conventional simulator were from 6.5 to 7.0-fold higher than in the modern simulator. The comparison of the two simulator types was the most important task to done in this study and we did not take air samples from the most hazardous places in the simulators. That is one reason why calculated indices were quite low.

The MIXIE program helped to form chemical groupings around combined health effects found in smoke. The program found five combinations where additive effects were possible. It also suggested two combinations where synergistic effects might be possible. The combinations of additive effect were cancer risk, central nervous system disorders, eye disorders, upper respiratory tract irritation, and potential disruptors of oxygen transport. There were also two notes about possible synergistic effects. The first was between carbon monoxide and hydrogen cyanide, for which an additive effect has been detected for mortality and synergy for physical incapacity. The second was between formaldehyde and hydrogen chloride and concerns interaction with regard to nasal cancer. These data uncovered more reasons why future risk assessments should be very cautious until adequate, consistent information about the synergistic effects of these chemicals is available.

The purpose of this study was to show what possibilities are available for conducting multiple exposure evaluation in the field in practice. Although the number of the air samples was limited for the indices calculation, simultaneously taken biomonitoring samples supported the findings of occupational hygienic measurements well. The findings of this study gave indicative information about firefighters' training conditions and the risks in conventional and modern simulators.

Conflict of interest statement

The authors declare no conflict of interest.

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