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AIRWAY IRRITATION OF VOC MIXTURES BASED ON THE EMISSIONS OF THE FINISHING MATERIALS - PVC FLOORINGS AND PAINTS

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ABSTRACT

VOC emissions from building materials are assumed to cause irritation of eye and the upper airways (sensory irritation, SI) in the indoor environment. Four finishing products, two PVC floorings and paints, were selected to this study: PVC(+) and Paint(+) were accepted whereas PVC(-) and Paint(-) were not acceptable in the human sensory evaluation. SI potency of VOC mixtures representing the material emissions were tested by the mouse bioassay (ASTM E981-84). Both the paint mixtures were much poorer irritants than PVC(-), but stronger than PVC(+). In the PVC floorings, the mouse bioassay and the human sensory evaluation ended up the same rank order for the two materials. On the contrary, the Paint(+) mixture of the paint accepted in the human evaluation proved to be a more potent irritant than the Paint(-) mixture. Overall, formaldehyde was the main reason for SI responses caused by the mixtures.

KEYWORDS: sensory, human response, material emissions, VOC, formaldehyde, irritation, flooring

INTRODUCTION

Sensory irritation (SI), irritation of eye and the upper airways, is a symptom frequently detected in buildings with a poor indoor air quality. It is widely assumed that VOCs emitted from building materials are one of the main causes for SI in the indoor environment. Although estimates of SI have been suggested for many single VOCs, no threshold values have been given for VOC mixtures, e.g. material emissions. Because of difficulties with estimating irritation potency for VOC mixtures, it has been suggested that olfaction parameters (e.g. odor threshold or acceptability) can be used as an indicator of the SI potency [1].

In the Finnish Classification of Finishing Materials, the human sensory evaluation of material emissions is used besides measurements of chemical emissions for classifying materials into three categories [2]. Four finishing products, two PVC floorings and two paints, were selected to this study. All the materials, except Paint(-), fulfilled the chemical emission criteria for the best class of materials (M1), but only two of them {PVC(+) and Paint(+)} were acceptable for M1 class according to the human sensory evaluation. The aim of this study was to clarify if there is a relationship between the SI potency tested by the mouse bioassay (ASTM E981-84;

[3]) for VOC mixtures representing the material emissions and the acceptability of emissions in the human sensory evaluation.

METHODS

VOC mixtures representing emissions from materials

The human sensory evaluation tests were performed with a naive panel of five persons who evaluated the acceptability of the material emissions in a chamber (Climpaq) using a scale from not acceptable (-1...-0,1) to acceptable (+0,1...+1) [4]. The mean acceptability value for the PVC floorings were +0.87 {PVC(+)} and -0.17 {PVC(-)}, and for the paints +0.28 {Paint(+)} and -0.11 {Paint(-)}. The PVC (-) was described as 'unpleasant', 'disgusting', 'acrid' and 'stale', and the PVC(+) as 'good', 'sweet' and 'plastic-like'. The Paint(-) was perceived as 'unacceptable' and Paint (+) as 'sweet' and 'acceptable'. The emission factors and the calculated concentrations of chemicals in the Climpaq chamber (V=0.05 m^3; loading factor for flooring materials 0.67 m^2 and for paints 2.3 m^2; N= 3.13-3.42 m^3/h) are presented in Table 1.

Based on the proportions of the compounds (Table 1), the *basic* chemical mixtures (mixtures without formaldehyde and ammonia) were prepared for the experiments from the pure chemicals (supplied by Sigma-Aldrich, Germany). The diluted water solutions from 36.5% formaline (Riedel-deHaën, Germany) and 25% ammonia (Merck, Germany) or gaseous ammonia (Woikoski, Finland) were used.

Testing of SI - the mouse bioassay

The SI potency of the VOC mixtures was investigated by the standard mouse bioassay (ASTM E981-84 [3]). In each experiment, four naive OF1 mice were exposed (head-only exposure) to the known airborne concentration of the chemical mixture at the airflow rate of 23 l/min. Different concentration levels were tested for each mixture. The basic mixtures were tested first followed by the mixtures with formaldehyde and ammonia (total mixtures).

Sensory irritants induce a reflex-based dose-dependent *decrease in respiratory rate* of mice (*RD*) due to stimulation of the nasal trigeminal nerve endings [5]. With the automated data collection system [6-8], a mean respiratory rate (f) of four mice was recorded for a 15 min control period (exposure to room air), for a 30 min exposure to a mixture and for a 15 min recovery period (exposure to room air) in each experiment. A baseline f was determined as an average of control period recordings and set equal to 100%. To separate SI from other respiratory effects, breaks after inhalation were also recorded during the experiments.

Risk evaluation

Occupational exposure limit (OEL) of a chemical can be obtained by multiplying RD_{50} value (i.e. a concentration which causes a 50% decrease in f) with 0.03 [9]. Recommended indoor air levels (RILs) [10] can be further estimated to be 1/4 - 1/40 * OEL, i.e. RD_{50}/133 - RD_{50}/1333.

Table 1. Emission factors of different compounds emitted from the PVC floorings and paints, and calculated concentration based on the emission factors in a Climpaq chamber.

Material / Compounds	Material emissions: Emission factor, $\mu g/m^2h$	Material emissions: Proportion, %	Calculated concentrations in Climpaq, $\mu g/m^3$
PVC (+), accepted			
• 1,2-propanediol	13	8.5	2.8
• phenol	5	3.3	1.1
• 2-(2-butoxyetoxy)ethanol	58	37.9	12.4
• 1-methyl-2-pyrrolidone	77	50.3	16.5
• ammonia,	-	-	-
• formaldehyde	-	-	-
SUM	153	100	32.7
PVC (-), not accepted			
• 2-ethylhexanol	13	18.1	2.6
• 2,2,4,6,6-pentamethylheptane	2	2.8	0.4
• 2-ethylhexanoic acid	35	48.6	6.9
• 1-butanol	5	6.9	1.0
• ammonia	12	16.7	2.4
• formaldehyde	5	6.9	1.0
SUM	72	100	14.2
Paint(+), accepted			
• Texanol	137	95.8	92.7
• ammonia	3	2.1	2.0
• formaldehyde	3	2.1	2.0
SUM	143	100	96.7
Paint (-), not accepted			
• 1,2-propanediol	260	69.5	191.1
• 2-(2-etoxyetoxy)ethan ol	100	26.7	73.5
• 2-amino-2-methyl-1-propanol	4	1.1	2.9
• (n-propylbenzamide)*	(3)	(0.8)	(2.2)
• ammonia	4	1.1	2.9
• formaldehyde	3	0.8	2.2
SUM	374	100	274.8

* n-propylbenzamide not included in the animal experiments because of supplying difficulties

RESULTS

SI was the main effect observed with the all mixtures. The *f* as a function of time in the experiments are shown in Figures 1 and 2. The PVC(+) mixture induced only slight SI on the levels of 300 and 580 mg/m^3 (Fig. 1). The basic PVC(-) mixture induced a dose-dependent decrease in f on the level of 87-306 mg/m^3 being almost 40% at the highest tested concentration. When formaldehyde and ammonia were added to the mixture, much stronger SI effects were observed. At the total mixture concentration of about 50 mg/m^3 with 11 mg/m^3 of NH_3 and 3.2 mg/m^3 of HCHO, the maximum decrease in *f* was 40-48% following moderate fading by the end of the exposure as seen earlier in the exposures to formaldehyde only [11].

The basic Paint(-) mixture did not cause any decrease in f below 600 mg/m^3, whereas a slight (<15%) decrease in respiratory rate was observed at the basic Paint(+) mixture levels of 140-180 mg/m^3 (Fig. 2). When ammonia (2.9 mg/m^3) was added to this mixture (140 mg/m^3), no

major changes in the response were detected. However, the addition of formaldehyde to the both basic mixtures induced the response significantly. Overall, both the paint mixtures were still much more poorer irritants than PVC(-), but stronger than PVC(+) mixture.

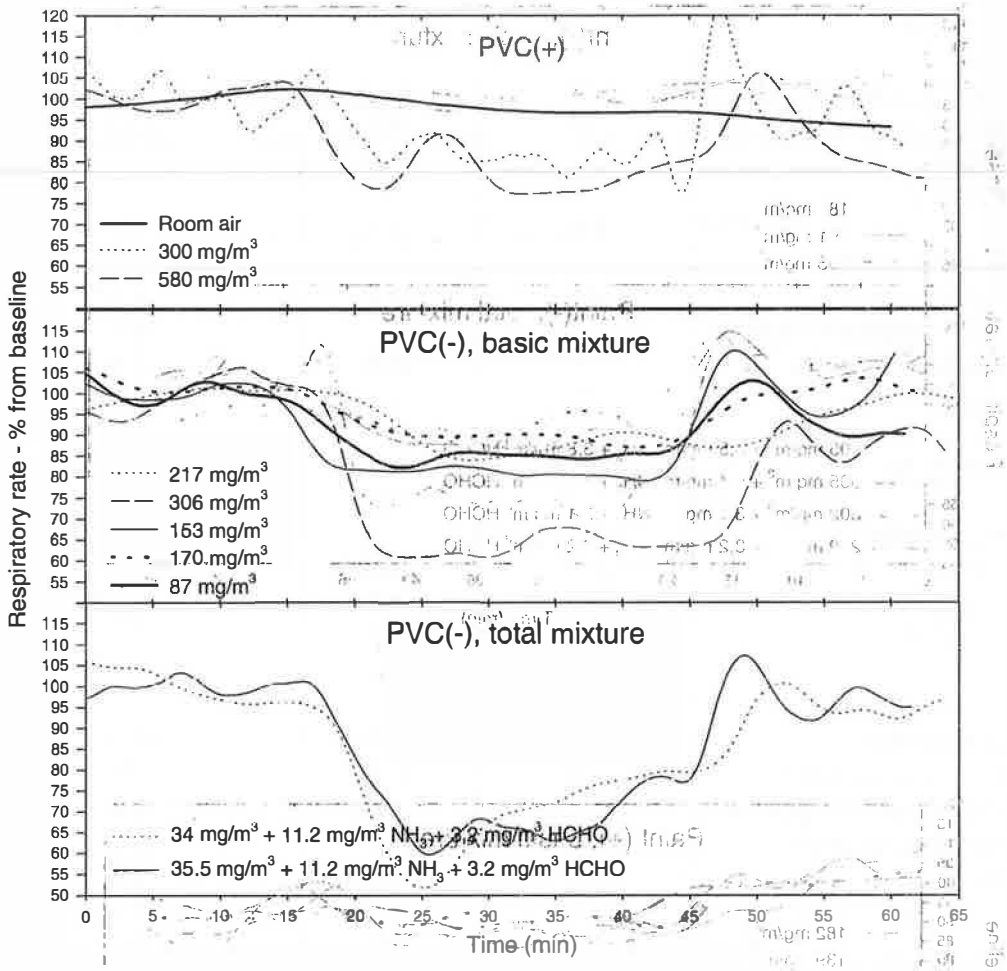

Figure 1. Time-response curves for the PVC mixtures

DISCUSSION

The PVC(-) mixture was clearly more potent sensory irritant than the mixtures of PVC(+) and the paints, whereas the difference between the paints was not so clear. This was true even for the basic mixtures, probably caused by two main components: 2-ethylhexanol and 2-ethylhexanoic acid. RD_{50} value of 2-ethylhexanol is 233 mg/m^3 [9]. For 2-ethylhexanoic acid, RD_{50} is not determined, but due to structural similarity it could be close to that of 2-ethylhexanol. The PVC(+) mixture and the basic Paint mixtures consisted of much poorer irritants.

Ammonia and formaldehyde are well-known sensory irritants [9, 11]. However, their SI potencies differs by factor of 100: the RD_{50} for ammonia is 303 - 790 mg/m^3 and for formaldehyde 3.9-6.5 mg/m^3 [9, 11]. In the mixtures studied, the concentrations of ammonia,

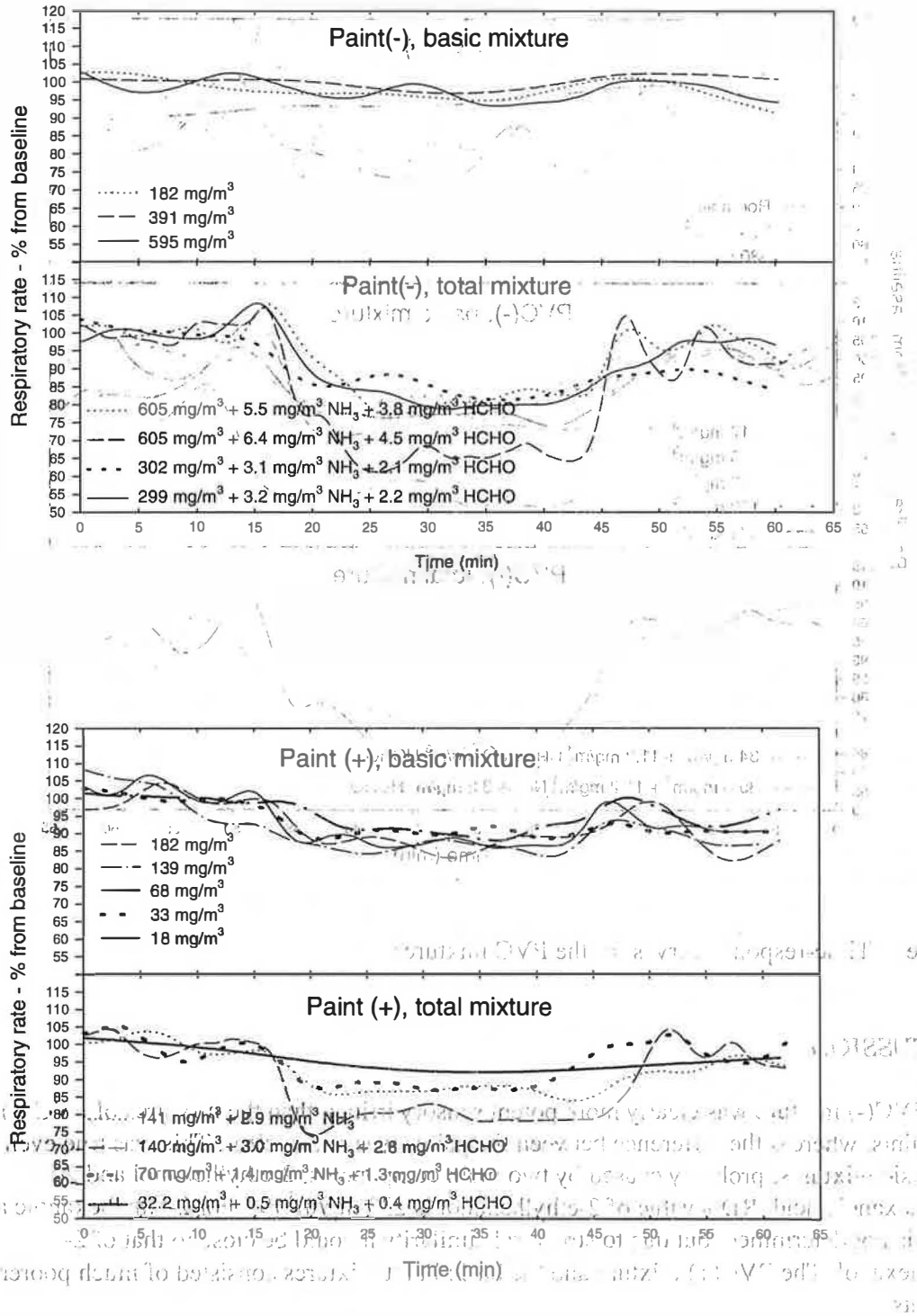

Figure 2. Time-response curves for the Paint mixtures

were far below its RD_{50}, whereas the highest concentrations of formaldehyde tested were close to the RD_{50}. Thus, formaldehyde was the main reason for stronger SI responses caused by the total mixtures compared to those induced by the basic mixtures.

In the PVC floorings, the mouse bioassay and the human sensory evaluation ended up the same rank order for the two materials. On the contrary, the Paint(+) mixture from the paint accepted in the human evaluation seemed to be more potent irritant than the Paint(-) mixture. The concentration of the Paint(-) emissions in a Climpaq chamber, however, was about 2.8 times higher than that of the Paint(+) (Table 1), which could explain the results of the human sensory evaluation. In addition, odor sensation may play a more significant role than SI in human perception. In the case of the Paints and PVC(+), the RD_{50} level was not reached. However, the concentration of 50 mg/m^3 for the PVC(-) is close to the RD_{50} value. Based on this concentration, RIL for PVC(-) mixture is between 38 and 375 µg/m^3. The lower RIL value is still over two times higher than the estimated concentration in a Climpaq chamber during the human sensory evaluation of the PVC(-) flooring (Table 1). Thus, it is not likely that SI was involved in the 'rejection' of the material.

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