A weighted sum parameter to simplify discussion on aldehyde exposure

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A WEIGHTED SUM PARAMETER TO SIMPLIFY DISCUSSION ON ALDEHYDE EXPOSURE

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ABSTRACT
In the present work, the personal exposure of a French urban population to several aldehydes is assessed. For this purpose, a weighted sum parameter is used which takes into account the differences in sensory irritation of aldehydes.
The personal exposure of 17 volunteers was assessed with personal passive samplers during one week. Simultaneously, aldehyde concentrations were determined in different microenvironments.
The results show that formaldehyde is the most important sensory irritant of the aldehydes included in this study, due to a particularly high sensory irritation and because it is a ubiquitous compound found at high concentrations. Personal exposure is strongly linked to indoor exposure (homes and offices). By using a weighted sum parameter, the discussion of large data sets is considerably simplified. This sum parameter could be easily extended to other strong sensory irritants, leading to a weighted TVOC value for sensory irritation.

INDEX TERMS
TVOC, sensory irritation, personal exposure, passive sampler, microenvironments

INTRODUCTION
The indoor air can contain a large number of Volatile Organic Compounds (VOC), which are held responsible for health effects like irritation of the eyes, skin and upper respiratory tract (sensory irritation). A sum parameter based on the sum of all measured VOC concentrations (Total VOC, TVOC) is frequently used for a first evaluation of the indoor air quality (ECA-IAQ, 1997). However, TVOC values are often not comparable when obtained with different analytical techniques (Uhde and Salthammer, 2000) and the different health effects of different compounds are neglected.

Indoor VOC with strong sensory irritation typically contain one or more functional groups (for instance diisocyanates, organic acids, or aldehydes) and many of them show a high chemical reactivity. Reactive compounds are rarely detected indoors, because of their short lifetime, and because conventional sampling and analytical techniques are not appropriate (Wolkoff et al., 1997; Wolkoff and Nielsen, 2001).
Simple aldehydes, however, are relatively stable compounds, for which standard sampling and analytical techniques have been established. Aldehydes were subject of several personal exposure studies (for instance (Ullrich et al., 1999)) and numerous emission sources have been identified indoors (for example furniture) (Salthammer, 1999) and outdoors (for example photochemical reactions) (Viskari et al., 2000).

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In the present work, the personal aldehyde exposure of a French urban population is evaluated. For this purpose, a weighted sum parameter is used, taking into account the differences in sensory irritation of aldehydes.

METHODS

Weighted Sum Parameter. A weighted sum parameter K_{RD50} is formed by taking the sum of weighted aldehyde concentrations (Equation 1):

$$K_{RD50} = \sum_{i=1}^{n} v_i c_i \quad (1)$$

The compound-specific weighing factors v_i (TenBrinke et al., 1998) are based on the sensory irritation of a compound, which in turn can be expressed by its RD_{50} value. RD_{50} is the concentration of a compound inducing a 50 % decrease in respiratory rate in mice (Alarie, 1966). It has been shown that RD_{50} is predictive of human responses. A slight irritation would occur at a concentration of $0.1*RD_{50}$, and minimal or no effect would occur at $0.01*RD_{50}$ (Jensen and Wolkoff, 1996).

Weighing factors v_i are formed by dividing the compound-specific RD_{50} value by the RD_{50} value for formaldehyde, the most abundant of all aldehydes studied. RD_{50} values and weighing factors are summarised in Table 1. Note that RD_{50} for acetaldehyde is particularly high compared to its homologues.

RD_{50} values are not available for heptanal and octanal, but since v_i values for propanal, butanal pentanal and hexanal are in the order of magnitude of 0.001, it was decided to apply the same weighing factor for heptanal and octanal (TenBrinke et al., 1998).
K_{RD50} includes the compounds listed in Table 1.

Table 1. : RD_{50} values and weighing factors v

aldehyde	RD_{50} / [mg/m^3] (Jensen and Wolkoff, 1996)	weighing factor v
Formaldehyde	5	1
Acetaldehyde	6700	0.0007
Propanal	5800	0.0008
Butanal	3700	0.001
Pentanal	4100	0.001
Hexanal	4500	0.001
Heptanal	---	0.001
Octanal	---	0.001
Benzaldehyde	1600	0.003
Furfural	1000	0.005
Crotonaldehyde	12	0.42
Acroleine	4	1.25

When assuming additivity of the irritating effects (Flemming et al., 1996) – which seems reasonable given the low concentrations typically encountered– , K_{RD50} can be considered as a concentration in [µg/m3] of an aldehyde mixture having the same sensory irritation as formaldehyde at the same concentration.

Experimental. Average concentrations of several aldehydes (formaldehyde, acetaldehyde, acroleine, benzaldehyde, furfural, pentanal, hexanal, heptanal and octanal; averaged over 5 days) in various urban environments and with personal samplers were obtained. Due to analytical limitations, crotonaldehyde, propanal and butanal were not quantified.

Passive sampling devices (Cocheo et al., 1996)) equipped with dinitrophenyl hydrazine (DNPH) sampling cartridges and ozone scrubbers (Bates et al., 2000), were used to sample the compounds listed in Table 1. Preliminary uptake rates were experimentally determined for butanal, pentanal, hexanal, heptanal, octanal, nonanal, furfural and benzaldehyde.

The measurement campaign was carried out in a medium-sized French town and consisted of two parts. The weather conditions during the first part were characterised by rain episodes. Throughout this part, 22 samplers were exposed at outdoor background sampling sites. In addition, samplers were installed at 8 sites where people spend more of their time.
During these first five days, 17 volunteers carried a personal sampler on them. Half of them were office workers and the other half worked outdoors (gardeners). All participants installed samplers indoors in their sleeping rooms (were the participants are supposed to spend most of their time at home) and in their offices.

After this first part, the campaign lasted for another five days, where 10 samplers were installed outdoors. This time, the weather was dry and rather sunny.

Analytical and further technical details are given in (Meininghaus et al., 2001).

RESULTS AND DISCUSSION

Influence of aldehydes other than formaldehyde. Crotonaldehyde and furfural were never detected. Acroleine was present in only two samples and benzaldehyde in 12 samples, whereas the other compounds were frequently detected. Concentrations of individual compounds were already published (Meininghaus et al., 2001).

Figure 1 shows a very good linear correlation between K_{RD50} values obtained from all samplers and the corresponding formaldehyde concentrations. The figure clearly illustrates that formaldehyde is by far the most important compound, due to a particularly low RD_{50} value (high sensory irritation) and because it is a ubiquitous compound found at high concentrations. Aldehydes with high sensory irritation (in particular acroleine and crotonaldehyde) were rarely detected and only at very low concentrations (this may be due to limitations of the sampling and analytical techniques). On the other hand, other frequently detected aldehydes show relatively high RD_{50} values.

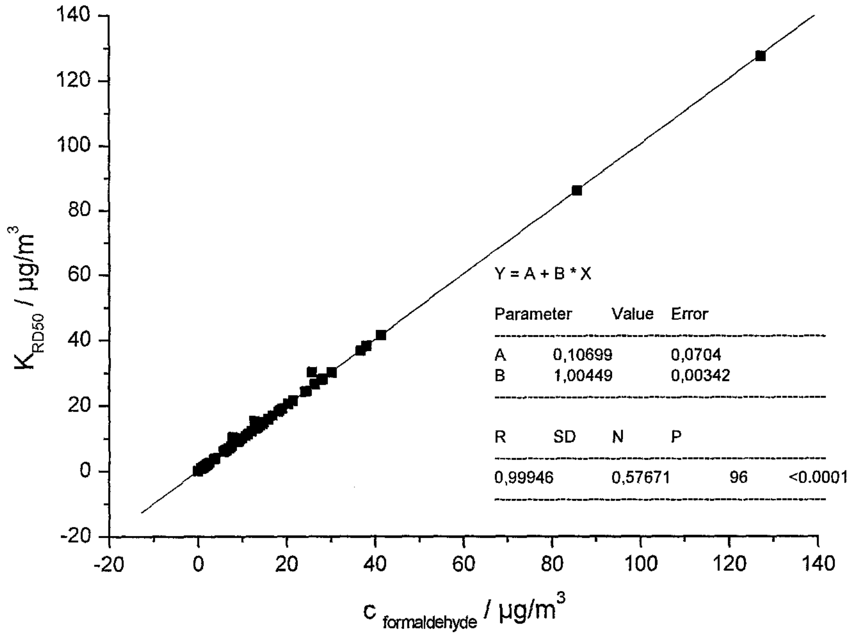

Figure 1. K_{RD50} values of all samplers over formaldehyde concentrations of all samplers

Although K_{RD50} values could be simply replaced by formaldehyde concentrations, they will be used throughout this paper, in order to illustrate how a weighted sum parameter – which could be easily extended to other compounds – helps to simplify the discussion on exposure to complex mixtures.

Comparison of Microenvironments. K_{RD50} values (minimum, median, average and maximum values) are presented in Table 2 for different environments and time periods and allow for comparing different environments.

Table 2. : Minimum, median and maximum K_{RD50} values of different microenvironments and time periods

environment	K_{RD50} / [µg/m³]			
	minimum	median	average	maximum
outdoors: background sites part I	0.7	1.2	1.3	1.9
outdoors: frequently visited sites part I	1.3	1.9	2.0	2.6
Outdoors: part II	1.0	1.3	1.4	1.9
Indoors: sleeping rooms	6.4	14.4	27.6	127.4
Indoors: offices	5.8	14.4	14.5	24.2
Indoors: personal samplers	6.4	14.5	16.7	38.2

Statistically significant differences (Mann-Whitney rank sum test, $p < 0.05$) were observed between outdoor and indoor K_{RD50} data sets, and between outdoor and personal sampler data sets. The results confirm that highest aldehyde concentrations prevail indoors, even with weather conditions favouring aldehyde formation by photochemical reactions. Personal exposure will hence mostly be determined by indoor exposure.

Aldehyde Exposure. The time spent in a microenvironment t_{env} multiplied by the respective K_{RD50} value is an exposure in this microenvironment (Equation 2):

$$Exposure_{env} = \sum t_{env} \times K_{RD50,env} \qquad (2)$$

The time t_{env} was obtained from a questionnaire distributed among the participants.

An estimated total exposure may be formed by summing up individual exposures of different microenvironments. This may then be compared to the calculated total exposures based on personal sampler measurements.

Figure 2 contains exposures of individual microenvironments and calculated exposures (personal sampler) of the participants. The total aldehyde exposure is strongly related to the exposure at home and to a lesser extent to exposure in the office. Outdoor exposure is negligible, even for the participants working outdoors (l-u).
Differences between estimated and calculated exposure may be due to the fact that not all the relevant microenvironments (transport, shopping mall, bar…) were included in this study. Moreover, the selected sampling sites (close to emission sources like furniture) may not be representative for the microenvironment.

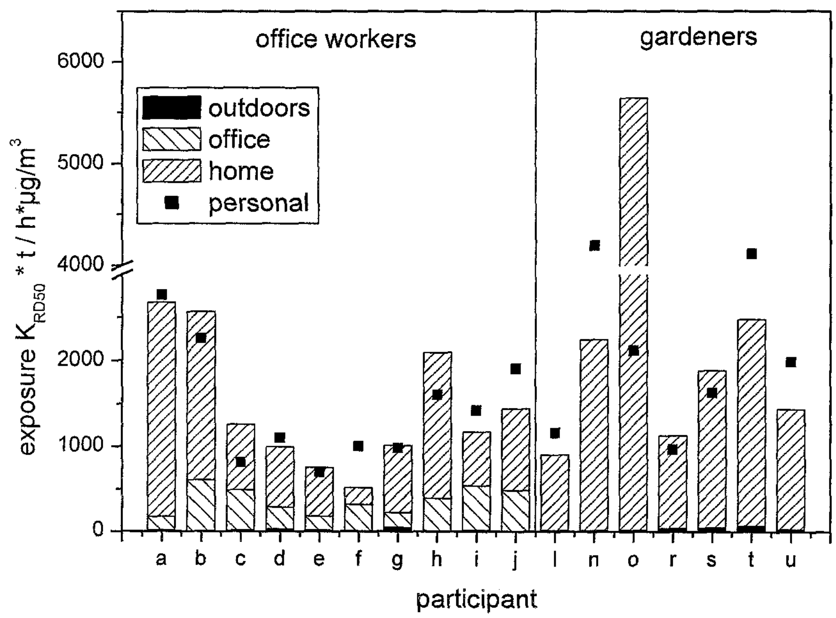

Figure 2: Aldehyde exposure in different environments and personal exposure.

CONCLUSIONS

By using a weighted sum parameter, the discussion of large data sets is considerably simplified.

With regard to personal exposure, formaldehyde is the most important sensory irritant of the aldehydes included in this study, and the exposure at home is much more important than outdoor exposure.

The weighted sum parameter contains information on the physiological effect of a mixture of compounds (aldehydes) and could be easily extended to other strong sensory irritants. This would finally result in a weighted TVOC value for sensory irritation.

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