

ORGANIC POLLUTANTS IN INDOOR AIR – BASICS AND PROBLEMS

ORGANISCHE VERUNREINIGUNGEN DER INNENRAUMLUFT – GRUNDLAGEN UND FEHLER

POLLUANTS ORGANIQUES DANS L'AIR INTERIEUR – BASES ET PROBLEMES

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SUMMARY

The concentration of volatile and semivolatile organic compounds (VOC and SVOC) in indoor air is the result of a complex interaction of sources and the specific situations within a contaminated building. It is influenced by a set of factors which should be taken into account to obtain comparable results. Even the same sources lead to different indoor air concentrations depending on structural situations, room climate, ventilation habits and the season. Indoor air is a dynamic system of which the composition is in principle varying. The parameters, which determine the concentration in indoor air are the source strength and the emission characteristics of the source. The source strength is a function of the diffusion potential and the vapour pressure of the pollutant/emitter and therefore a function of temperature and relative humidity. Besides this, the dilution factor with ambient air depending on varying air exchange rates and ventilation habits influences the concentration of organic pollutants in indoor air. Different results in the same building can normally be ascribed to these problems if all relevant parameters are documented in the test reports. In common the analytical problems determining organic compounds in indoor air after collecting on different absorbers are less important, compared to these influences. This leads to a situation that reported data, without precise information about the boundary conditions during the sampling period, can not be compared or evaluated. The standard deviation for the total analytical procedure (sampling and determination) differs for common pollutants (volatile aliphatic and aromatic organic compounds, persistent semivolatile organochloro-compounds) between 10 and 30 % even if the sampling is carried out under the same conditions with stan-

standardised methods. Higher differences between two measurements can normally be ascribed to differences of the room climate conditions existing while sampling. These differences lead to the fact that deviations of more than 100 % arise in practice. This specific situation complicates the estimation of an average annual concentration of pollutants in indoor air.

ZUSAMMENFASSUNG

Die Konzentration von flüchtigen und schwer flüchtigen organischen Verbindungen in Innenraumlufte ist das Ergebnis von komplexen Wechselwirkungen von Quellen und den jeweiligen besonderen baulichen Gegebenheiten in einem entsprechend belasteten Gebäude. Sie wird beeinflusst durch eine Reihe von Faktoren, die berücksichtigt werden müssen um vergleichbare und damit beurteilbare Daten zu erhalten. Sogar gleichartige Quellen führen in Abhängigkeit von baulichen und klimatischen Randbedingungen, sowie Lüftungsverhältnissen und Jahreszeiten zu unterschiedlichen Konzentrationen in der Innenraumlufte. Innenraumlufte ist ein dynamisches System, dessen Zusammensetzung grundsätzlich nicht konstant ist. Die Konzentration im Innenraum wird wesentlich von der Quellstärke und den charakteristischen Eigenschaften der Quelle bestimmt. Die Quellstärke ist eine Funktion des Diffusionspotentials und des Dampfdrucks der emittierenden Verbindung und wird dadurch von der Temperatur im Innenraum, der unter Umständen davon unterschiedlichen Temperatur der Quelle selbst und der relativen Luftfeuchte wesentlich bestimmt. Daneben ist die schwankende Verdünnung mit der Außenluft in Abhängigkeit von der durch bauliche Gegebenheiten bestimmten Luftwechselzahl und den Lüftungsgewohnheiten der Nutzer, die zudem wesentlich vom Außenklima (Wind) beeinflusst werden eine relevante Einflussgröße. Unterschiedliche Ergebnisse der Bestimmung der Raumluftkonzentrationen können im Regelfall auf diese Einflussgrößen zurückgeführt werden, wenn eine nachvollziehbare Dokumentation der Verhältnisse zum Zeitpunkt der Probenahme vorliegt. Die aus der Dynamik der Innenraumlufte resultierenden Schwankungen der Ergebnisse übersteigen die Schwankungen der Verfahren (Analyse plus Probenahme) signifikant. In der Praxis sind Unterschiede zwischen zwei vorliegenden Ergebnissen von 100 % durchaus nicht unüblich. Diese unterschiedlichen Randbedingungen zum Probenahmezeitpunkt führen dazu, dass Ergebnisse ohne eine entsprechende Dokumentation der Randbedingungen prinzipiell nicht beurteilbar sind. Basis für die Vergleichbarkeit ist die Standardabweichungen von Vergleichsmessungen unter gleichen Probenahmebedingungen. Je nach Konzentrationsbereich und zu untersuchender

Substanz sind für die üblichen flüchtigen und schwer flüchtigen organischen Verbindungen (Aliphate, Aromate, Ester, schwer flüchtige, persistente chlororganische Verbindungen) Schwankungen zwischen 10 und 30 % mit den gegebenen Normverfahren möglich.

RESUME

La concentration en composés organiques volatiles et semi-volatiles dans l'air intérieur est le résultat d'interactions complexes entre les sources d'émission et les conditions spécifiques du bâtiment contaminé. Elle est influencée par une série de facteurs dont on doit tenir compte afin d'obtenir des données comparables et donc exploitables. Des sources d'émission similaires peuvent mener à des concentrations différentes, en fonction de la configuration du bâtiment, du climat intérieur, de l'aération et de la saison. L'air intérieur constitue un système dynamique dont la composition n'est en général pas constante. La concentration à l'intérieur dépend essentiellement de l'intensité et des caractéristiques de la source. L'intensité de la source est une fonction du potentiel de diffusion et de la pression de vapeur du polluant et par conséquent fonction de la température intérieure, de la température de la source et de l'humidité relative de l'air. En plus, la dilution variable par l'air extérieur en fonction du taux de renouvellement de l'air et des habitudes d'aération joue un rôle important, et est largement influencée par le climat extérieur (vent). Les divergences de résultats pour un même bâtiment peuvent en général être attribuées à ces facteurs, si tous les paramètres utiles ont été documentés lors de l'échantillonnage. Les fluctuations dues au procédé (échantillonnage et analyse) sont en général nettement inférieures à celles dues à la dynamique de l'air intérieur. En pratique, des divergences de 100 % entre deux mesurages sont courantes. Sans la documentation des conditions marginales lors de l'échantillonnage, les résultats des analyses ne sont ni exploitables ni comparables. La base pour la comparabilité est l'écart-type des mesurages comparatifs réalisés sur des échantillons prélevés dans les mêmes conditions. Selon la substance et sa concentration, des fluctuations entre 10 et 30 % sont possibles avec les méthodes standard pour les composés organiques courants (composés aliphatiques, hydrocarbures aromatiques, esters, composés organiques persistants).

KEYWORDS: Volatile and semivolatile organic compounds (VOC, SVOC), indoor air, basics, sampling and determination, limits, errors

1. BASICS

Changed life and working habits lead to the fact that the majority of humans in industrialised countries stay inside of buildings up to approx. 80 % per day [1]. Due to a lot of sources inside (paints, furniture, flooring materials, consumer products and other building materials) in combination with reduced or inadequate ventilation, the average concentrations of organic pollutants in indoor air are mostly 2 – 5 and occasionally more than 100 times higher than in ambient air [1]. This leads a lot of reports dealing with health and comfort problems, which have been associated with the presence of organic compounds in indoor air. Therefore, it should be of special interest to receive reliable data for the concentrations of organic pollutants in indoor air. This can be achieved by choosing suitable measuring and sampling procedures which are adapted to the different problems. Comparable and thus evaluated data can be obtained using the meanwhile existing technical guidelines in combination with a basic knowledge of physical and physicochemical behaviour of sources and emitters. This includes a documentation of the problem to be solved, as well as a complete documentation of structural and climatic boundary conditions during the sampling period in the reports.

In principle, the measurements of organic pollutants in indoor air demands to define the objectives of the measurement before beginning any measurement action. In practice different questions arise. Usually measurements in indoor air are carried out because of vague complaints of users in respect to a "bad" air quality. In this case normally no specific species and source of the indoor air pollution is known. This situation requires a screening for possible reasons of these troubles assumed. In this case no general strategy is possible and almost any individual case is different. Number and effectiveness of possible sources in indoor air are different in almost each individual case. In indoor air generally several hundred single organic compounds in a concentration range between few pg/m^3 up to mg/m^3 can be detected. These are emitted from different building materials, furniture, cleaning agents and consumer products [2-5]. To clarify the question whether there exists an unusual situation the knowledge of the average concentrations in indoor air is helpful [4]. Besides this, it is necessary to determine the room temperature, the relative humidity and the concentration of CO_2 in indoor air of rooms with reported "bad" air quality. In many cases the reported troubles can be traced back to those indoor air parameters. In indoor air of class rooms for example CO_2 -concentrations of 1500 up to 3000 ppm in

combination with temperatures above 25 °C during the lessons are not unusual [6,7].

Besides these orientating or screening measurements or as a consequence of a first screening the question whether a defined limit or guide line value in indoor air is exceeded. To answer these questions in general, a increased effort is required, especially when a concentration in the range of the limit or guide line value is expected. The guide line value for formaldehyde in indoor air in Germany is given by 120 $\mu\text{g}/\text{m}^3$. The confidence interval for these measurements based on two parallel samplings is given by 45 $\mu\text{g}/\text{m}^3$ (VDI 4300 Bl. 3). This means that all results obtained between 100 and 140 $\mu\text{g}/\text{m}^3$ may describe the same indoor air concentration. This means, that it is not possible to decide whether the given guide line value is exceeded or not based on only two obtained results. This principle problem has to be considered always, when the concentration in indoor air is close to a given limit or a guideline value. Currently, for example, a guide line value for dioxin-like PCBs of 4,7 pg WHO-TEQ/ m^3 is discussed in Germany. The measurement uncertainty for those dioxin-like PCBs (sum of dioxin-like PCBs calculated as WHO-TEQ) in indoor air is about 0,6 pg WHO-TEQ/ m^3 . This means, that for results within the range of 4,1 and 5,3 pg WHO-TEQ/ m^3 it is in principle not to decide whether this guide line value discussed is exceeded or not (see table 1) [6]. This means, that any exceeding of a given limit or guide line value based on only one result can not be assessed in principle if the concentration in indoor air is in the range of a limit or guide line value. In practice frequently exceedings of a given limit or guide value are assessed based on just one result. Based on these considerations three different questions and thus three measurement strategies have to be differentiated.

Table 1: Standard deviation of the determination of dioxin-like PCBs in indoor air (sampling and determination) [6]

						MEAN	STANDARD DEVIATION (ABS.)
Average Temp. [°C]		23,8		23,5			
PCB-Congener	WHO - TEF						
		PCB in indoor air [ng/m ³]					
77	0,0001	0,7	0,7	0,6	0,7	0,68	0,05
105	0,0001	2,5	2,4	2,3	2,7	2,48	0,17
114	0,0005	0,1	0,1	0,08	0,07	0,09	0,02
118	0,0001	14,2	13,6	10,8	12,2	12,70	1,52
126	0,1	0,02	0,02	0,01	0,02	0,02	< 0,01
156	0,0005	2,7	2,4	2,6	2,7	2,60	0,14
157	0,0005	0,3	0,3	0,2	0,2	0,25	0,06
167	0,0000 1	1,1	1,1	0,9	0,9	1,00	0,11
169	0,01	< 0,1	< 0,1	< 0,1	< 0,1	< 0,1	--
Σ (min) ng/m ³		22,4	21,4	18,2	20,2	20,55	1,81
Σ (max) ng/m ³		22,4	21,5	18,2	20,2	20,56	1,82
WHO-TE min in pg/m ³		5,2	5,5	4,2	4,9	4,9 ₅	0,56
WHO-TE max in pg/m ³		5,2	5,6	4,2	5,0	5,00	0,59

- Checking the compliance with a guideline or limit value. Usually the sampling conditions for this problems are clearly fixed by standards as for example in the VDI 4300 Bl. 3.
- Determination of an average concentration during longer periods of time while using the building. Therefore it is often required to define the measurement and sampling conditions, depending on the specific situation of the building and the way how this buildings are used. For example usual ventilation habits differ between classrooms and offices or living spaces. This means, however, that a sampling procedure that is suitable for continuous sampling during longer periods has to be chosen. Usually passive collecting systems are suitable for this. If no passive collecting systems are available it

is necessary to develop a strategy for representative sampling under consideration of seasonable variability [6,8].

- Determination of a maximum concentration ("worst case"), where the conditions are to be co-ordinated to the physicochemical characteristics of the parameter which should be analysed, e.g. vapour pressure (equilibrium concentration in indoor air at normal room temperature).

2. PRELIMINARY TEST METHODS

In normal cases the expected low concentrations of organic pollutants in indoor air do not permit the use of direct monitoring measuring systems. Methods like flame ionisation detector (FID), photo ionisation detector (PID) and photo-acoustic sensor (PAS) for volatile organic compounds can be applied with restrictions. Under defined conditions, these methods may give a fast overview of a possibly existing air pollution. In practice however, these methods show substantial deviations from the actual concentrations in indoor air. These methods just give hints. For semivolatile organic compounds (SVOC) like pesticides, polychlorinated biphenyls (PCB), softeners (phthalates) and various flame retardants in indoor air no directly indicating methods exist up to know. Test tubes which are commercial available, are normally used for occupational air. The detection limits for those tubes are normally within the range of mg/m³. Those tubes are suitable as preliminary test methods, if high indoor air concentrations are expected. Known cross-sensitivities for example for compounds of the classes of aliphates, aromatics, ketones or esters have to be regarded (VDI 4300 Bl. 6). In general preliminary test cannot be used to determine an exact concentration of organic compounds in indoor air.

3. SAMPLING METHODS

The low concentrations of organic pollutants in indoor air requires an enrichment step via a sampling system normally. Sampling of indoor air is usually accomplished at average indoor temperatures of approximately 18 - 24 °C and air humidity between 30 and 60 % (comfort range (VDI 4300 Bl. 6)). In principle active and passive sampling techniques are applicable. Active sampling techniques with a pump are usually preferred. The disadvantage of these sampling techniques are, that they give just a "snapshot" of the indoor air situation during the sampling period. Usual changes of the concentration of organic pollutants in indoor air, resulting from user behaviour (ventilation habits) and changes of the

source strength can not be detected. The sampling volume depends on the sampling technology and the expected concentration and varies between 0.001 – 400 m³. Small volumes from 1 to max. 10 l are used in combination with thermal desorption techniques to determine various VOC. Air volumes up to 400 m³ are necessary to determine polychlorinated dibenzodioxins and dibenzofurans (PCDD/F) in indoor air. When collecting volumes of 400 m³ it has to be noticed that the sampling air volume drawn in per hour shall not exceed 1/10 of the room volume. Otherwise a dilution via ambient air can not be excluded. Using passive collecting systems an overview of the average indoor air concentration of a longer period is possible. Passive sampling systems are available for aliphatic and aromatic organic compounds as well as aldehydes [9]. These methods do not allow to control the indoor conditions (ventilation habits, room climate) during the sampling period. Temperature and air humidity can be measured additionally using small continuous registratrating systems . It has to be regarded, that users very often are interested to create worst-case situations to prove that their health problems do have an objective background. Applications and limits of these sampling techniques and aspects of the measuring strategy are given in the standards VDI 4300 Bl. 1 - 8, DIN ISO 16000 Bl. 1-4 as well as E DIN 14412 in detail.

4. ANALYTICAL METHODS

Organic compounds in indoor air were usually detected after absorption on fixed phases, either directly by thermal desorption techniques or after extraction with different solvents with capillary gas chromatography (GC) with different detection systems (e.g. flame ionisation detector FID; Photo ionisation detector PID, mass-spectrometers (MS with low or high resolution). For reactive compounds, such as aldehydes reaction absorption systems (2,4 Dinitrophenylhydrazin DNPH as fixed phase) are used which forms a stable hydrazone with aldehyde. After extraction the determination of the aldehydes is carried out with high performance liquid chromatography (DIN ISO 16000 Bl.3 and 4). Meanwhile, the appropriate technical standards for a set of indoor air pollutants are existing and are constantly extended. For common volatile organic compounds (VOC) the standard deviation for the total procedure (thermal desorption sampling and determination) in concentration-ranges between 110 µg/m³ (n-hexane) and 16.8 µg/m³ (m-xylene) vary between 5,6 % (n-hexane) and 0.9 % (m-xylene). (DIN ISO 16017 Bl.1). Comparable data can be achieved by sampling

on activated charcoal and desorption with different solvents (VDI 2001 Bl. 2). This shows that the common accuracy of the determination procedures including the sampling procedures are sufficient for common VOC. The determination of semivolatile organic compounds (e.g. biocides, flame retardants) by gaschromatography/ massspectrometry after absorption on PU foam shows recovery rates between 92 % (γ -hexachlorocyclohexane; γ -HCH) and 100 % (p, p, DDT) with a standard deviations between 8 % (γ -HCH) and 21 % (DDT). The use of PU foams for compounds with a boiling point below 200 °C (e.g. 1,2,3 trichlorobenzene) is not adequate. The recovery rates drops under 10 % (EPA TO-4A). Examples of the use, the problems and the limits of the existing methods are given in the available standards DIN ISO 16000 Bl. 3,4 and 6; DIN EN 16017 Bl. 1 and 2; E DIN 14412; VDI 2001 Bl. 1-3; VDI 3864 Bl. 1 und 2; VDI 3875 Bl. 1 VDI 3498 Bl. 1 and 2; VDI 4301 Bl. 1 - 3 as well as EPA Compendium Methods TO-4A and TO -10A in detail. The use of these standards is recommended. The detection limits of the analytic procedures applied varies as a function of the method, the compounds and the sampling volume between few pg/m^3 (PCDD/F or dioxin-like PCB) [6] up to the $\mu\text{g}/\text{m}^3$ range (aldehydes, usual VOCs)[2-4,6,9].

5. ERRORS AND LIMITS

In practice, despite of existing technical standards the results obtained in the same polluted building often vary significantly. In common these differences are not the result of an analytic error but due to the dynamics of indoor air. Very often it is possible to explain those differences as a result of different climatic conditions during sampling in combination with the physicochemical behaviour of the interesting compound. The concentration of organic compounds in indoor air depends on a line-up of varying factors. Indoor air is in principle a dynamic system. The parameters, which determine the concentration in indoor air are the source strength, the physicochemical characteristics of the emitter and the dilution with ambient air depending on the air exchange rates or ventilation habits given. The source strength depends on the diffusion potential and the vapour pressure of the emitter. Therefore, emission is as a function of the temperature of indoor the air and the source, which might be different because of influences of structural conditions like heaters/radiators or direct sunshine on buildings materials. The strength of sources is often also depending on the relative humidity of indoor air, especially for all organic compounds which are steam volatile.

Tab. 2 and 3 give examples for the effects on the concentration of dioxin-like PCBs and PCP in indoor air.

Table 2: Concentration of selected dioxin-like PCBs in indoor air of 5 PCB contaminated buildings in dependence on the season (winter times/summer times) [6]

	SAMP- LING PERIOD	T [°C] Ø	PCB 105	PCB 118	PCB 126	Σ PCB*	WHO- TEQ
			Concentration in indoor air [ng/m ³]				pg/m ³
A	summer	23,6	4,8	38,2	0,02	56,2	10,0
A	winter	22,5	3,8	17,2	0,02	20,6	5,6
B	summer	25,0	4,9	64,1	0,02	85,7	13,6
B	winter	23,0	4,1	41,3	< 0,01	57,4	8,1
C	summer	25,1	5,5	66,6	0,01	90,2	13,7
C	winter	21,5	2,1	18,8	< 0,01	25,6	3,5
D	summer	21,8	2,5	37,1	< 0,01	48,5	7,1
D	winter	17,5	1,2	13,4	< 0,01	18,2	2,6
E	summer	23,9	4,6	12,1	< 0,01	21,8	2,5
E	winter	18,9	2,3	3,8	< 0,01	8,5	1,0

T = temperature ; Σ PCB* = Sum dioxin-like PCB

Table 3: Concentration of Pentachlorophenole (PCP) in indoor air of a public library measured under standardised conditions (VDI 4300 Bl. 2)

ROOM TEM- PERATURE	REL. HU- MIDITY	DATE OF SAM- PLING	CONCENTRATION OF PCP
28 °C	52 %	August	480 ng/m ³
24 °C	45 %	November	75 ng/m ³
19 °C	36 %	January	15 ng/m ³

In many cases it is not considered, that the temperature of the sources depends on structural condition and can vary between 10 and 60 °C depending on the season. Besides these factors, the dilution with ambient air depending on varying air exchange rates influences the concentration of organic pollutants in indoor air basically. This has a substantial influence on the determinable contents of the emitters in indoor air at comparable ambient temperatures. These effects increase with the boiling point of the emitters (see tabs. 2 and 3). This dominating influence can be shown in detail for polychlorinated Biphenyls, especially for dioxinlike PCBs (tab. 2). The PCB-concentration in the indoor air increases with the temperature and is significantly higher in summer as in winter even at almost similar room temperatures. An increase of temperature of approximately 5 °C doubles the PCB concentration in indoor air (tab. 2 and fig. 1) [6,7].

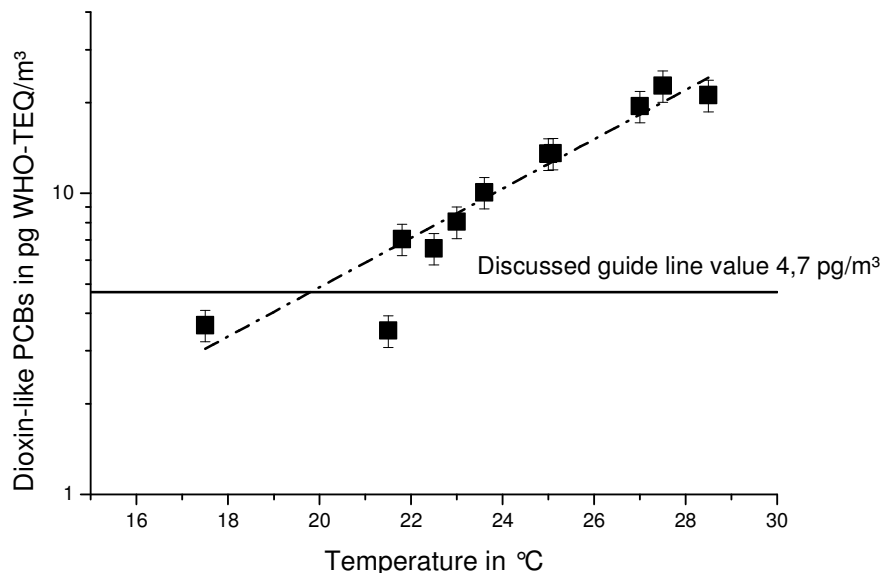


Fig. 1: Temperature dependence of the emission of dioxin-like PCBs. Average temperature during the sampling period. [6]

The dependence of the emission characteristics on vapour pressure/boiling point can be demonstrated by the different emission characteristics of n-propylbenzole (boiling point 159°C) versus diethyleneglykole (boiling point 245 °C) (see fig. 2) [5]. VOCs with boiling points between 80 and 180 °C lead in general to short term, intensive pollution of the indoor air immediately after application of the products. Compounds with high boiling-points (> 250 °C) lead to smaller, but long lasting emission, as well as to the generation of secondary contaminated surfaces.

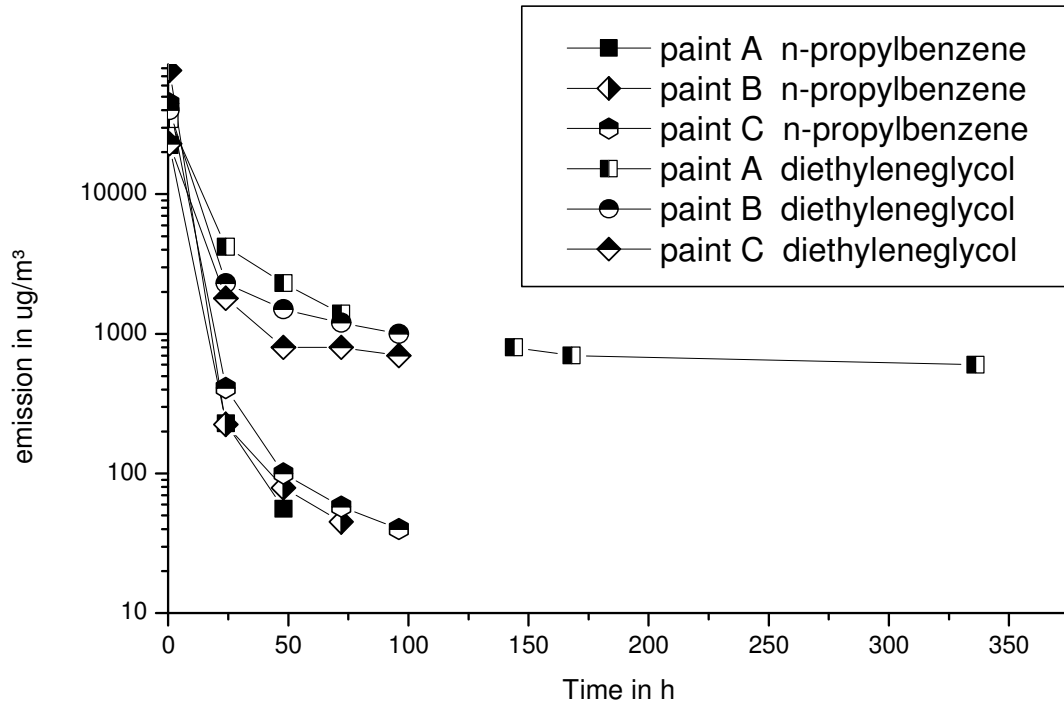


Fig. 2: Emission characteristics of *n*-propylbenzene (boiling point 159 °C) and diethyleneglycol (boiling point 245 °C) used as solvents in different paints [5]

Besides emission processes especially for semivolatile compounds adsorption processes have to be considered. Sinks like material surfaces, dust particles have to be taken into account. The lower the vapour pressures of semivolatile compounds is, the more important are these sink processes. The surface concentration of these secondary contaminated surfaces increases over the time. Especially for persistent organic compounds like PCB this effect leads to an indoor situation, where after several years the secondary sources form about 50 % of the emission and thus 50 % of the indoor air concentration [10,11]. As already mentioned house dust is a very effective sink. Thus house dust may directly influence the measurable concentration of semivolatile organic compounds in indoor air.

An example for the significant influence of whirled up dust as a result of indoor activities, here for example a cleaning procedure with a broom, is given in table 4. In house dust the concentrations of PCBs with lower vapour pressures such as PCB 101, PCB 153, PCB 138 and PCB 180 are enriched. These PCB contaminated dust particles whirled up, can be collected on PU-foam as well as the PCBs in the gaseous phase emitting from the given sources [12]. The effect of indoor activities is a significant increase of the PCB concentration in indoor air. Whirling up house dust can also be achieved by normal activities in

class rooms during lessons. The result of this activities may be an increase of the indoor air concentration, compared to measurements carried out under standardised conditions without any inside activities.

Table 4: Polychlorinated Biphenyls (PCB) in indoor air ; Comparison of the sampling technique VDI 4301 Bl. 2 and EPA TO-10 A 2; A = Conditions according VDI 4301 Bl. 2; B = During the sampling a cleaning procedures with a broom was performed [12]

PARAMETER	SAMPLING METHOD ACCORDING TO			
	VDI 4301 BL. 2		EPA COMPENDIUM METHODE TO-10A	
	Concentration in ng/m ³			
	A	B	A	B
PCB 28	25	30	50	25
PCB 52	35	40	25	50
PCB 101	15	120	10	100
PCB 153	10	210	8	180
PCB 138	5	240	5	310
PCB 180	5	170	5	240
Sum PCB	95	790	103	905

The concentration of volatile and semivolatile organic compounds in indoor air is the result of a complex interaction of sources and the specific situation inside of contaminated buildings and the climatic conditions in the surrounding of these buildings. Therefore it varies depending on the present situation. It is influenced by a set of factors which should be considered to obtain comparable results. Even the same sources lead to different indoor air concentrations depending on structural situations, room climate, ventilation habits, activities and the season. Hygienic appraisal of indoor air are normally based on an average annual indoor air concentrations. The given examples for the influences of various parameters on the indoor air concentration prove the difficulties of any estimations of an average annual indoor air concentration.

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