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# Air Change Measurements Using Tracer Gases

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## 1. Introduction

Both comfortable and healthy indoor climate conditions can only be achieved by constant fresh air supply. However, the minimum of air change required to reach this goal is depending upon different perspectives. Measures for thermal insulation and energy savings are difficult to bring in line with air quality requirements that result from findings of epidemiological studies (Seppänen et al., 1999; Seppänen & Fisk, 2004; Wargocki et al., 2002), and supply air facilities considering the technical construction of buildings and safety aspects (Erhorn & Gertis, 1986). Moreover, construction deficits impairing the integrity of the building envelope, meteorological conditions (thermal and flow induced pressure differences) and, not least, the behaviour of the residents, affect the air change in a variety of ways (Heidt, 1987).

For a number of different reasons it is necessary and desirable to examine the real fresh air flow between indoor and outdoor climate under given circumstances. Air change processes are of particular importance in studies focusing on their relationship to indoor air pollutants.

For manifold reasons the air quality inside buildings has been intensively investigated since quite a long time. Of particular relevance are the following aspects (Seifert & Salthammer, 2003):

- In countries with a cold or temperate climate, inhabitants spend more than 50% of the time in their homes. In certain population groups (e.g. infants and the elderly), this proportion is even exceeding 90%.
- Indoor air contains a wide range of different organic and inorganic components. Therefore, the carbon dioxide concentration alone cannot always be regarded as an indicator of air quality (Fanger, 1988; Persily, 1997). Above all, organic compounds which are released from the building, furnishings, household and hobby devices, as well as by daily activities of the inhabitants, such as cooking, baking or frying, and especially smoking, altogether contribute to air pollution caused by volatile organic compounds.

The energy crisis in the 1970s caused an increase in energy costs, evoking an urgent need to reduce the consumption of heat energy. The prevention of heat loss in homes is a very effective way to save energy and related costs. For climate protection and the reduction of global CO<sub>2</sub> emissions the economical use of energy resources is of outstanding significance. Thermal insulation of the building envelope protects efficiently against heat

loss, but enhanced tightness includes the disadvantage of reduced air change, thereby increasing the indoor pollutant concentration. To evaluate the indoor air quality under conditions of natural ventilation, the air change rate (ACR) can be determined through standard tracer gas measurement.

For this purpose, a small amount of the tracer gas is released in the room (or building) under study and its concentration is then recorded as a function of time. Subsequently, using appropriate evaluation algorithms the ACR can be calculated from the data obtained.

The first section is introducing basic physical principles of air change processes between indoor and outdoor environment, followed by a description of tracer gases which are frequently used in daily routine, and methods determining the ACR. Moreover, the applicability of carbon dioxide as a tracer gas has been compared with results obtained by hexafluorobenzene or sulphur-hexafluoride. In addition, we are discussing the impact of weather conditions on ACR data obtained under natural ventilation. This section is followed by a review on ACR due to passive ventilation through facades in selected residences in Berlin. Finally, the relationship between air change and the concentration of selected volatile compounds under worst case conditions will be discussed.

## 2. Modelling air change in indoor rooms

According to VDI (2001) 4300, part 7, air change is defined as the ratio of air supply  $Q(t)$  into a zone (i.e. a room or space) in relation to the volume of this zone  $V_R$  (room volume) and is generally expressed as air change per hour [ $h^{-1}$ ] or [ACH]. The following equation expresses this definition:

$$\lambda(t) = Q(t)/V_R \quad (1)$$

$\lambda(t)$  is the ventilation rate or air change rate [ $h^{-1}$ ],

$Q(t)$  is the air supply into a room [ $m^3/h$ ],

$V_R$  is the room's volume [ $m^3$ ], and  $t$  = time [h].

### 2.1 Model assumptions

For the model described here, which is designed to calculate the time course of the tracer gas concentration, the following simplifying assumptions has been made:

- The tracer gas is considered to be chemically stable and inert; i.e. there will be no chemical reactions capable to alter the concentration of the tracer gas in the room.
- There will be no adsorption processes on walls, ceiling or furnishings of the room that may lower the concentration of the tracer gas in the room.
- The air is considered to be completely mixed throughout the measurements. Inside the room there are no concentration gradients, i.e. the concentration of the tracer gas at a given time is the same for the whole room.
- An exchange of tracer gas-containing air with ambient air only occur in those areas that are in contact with the outside, i.e. air change with other interior spaces is considered to be negligible. The room in which the tracer gas was released is considered a single zone system.
- The exchange processes that take place during the measurement period are assumed to be temporally invariant. The air supply rate  $Q(t)$  and, thus, the air change rate  $\lambda(t)$  are constant.  $Q(t)$  and  $\lambda(t)$  can be replaced by  $Q$  and  $\lambda$ .

## 2.2 Model equations

The basis for the description of the relationship between the mass or concentration of a gaseous substance in a space as a function of time is the mass balance equation. This equation expresses that the mass and – in a fixed volume – the concentration of a tracer gas can only change when either more tracer gas is added to the original amount or tracer gas is removed by elimination processes. Considering the above assumptions, the following supply and removal processes are significant for the tracer gas concentration in the room air (physical dimensions of these variables are given in brackets):

- Transport of tracer gas from the room air to the outside:  $Q \cdot C_i$  [mass per time unit]
- Transport of tracer gas from the outside air into the room air  $Q \cdot C_a$  [mass per time unit]
- (Constant) emission  $E$  of tracer gas into the space by a tracer gas source [mass per time unit]

Thus, the mass balance equation can be formulated as the following differential equation (Heidt & Werner, 1986):

$$V_R \cdot dC_i(t)/dt = -C_i \cdot Q + C_a \cdot Q + E \quad \text{or}$$

$$V_R \cdot dC_i(t)/dt = -(C_i - C_a) \cdot Q + E \quad (2a)$$

After dividing both sides of the equation by the volume  $V_R$ , we obtain an ordinary differential equation which describes the concentration change of a tracer gas in the room per time unit:

$$dC_i(t)/dt = -(C_i - C_a) \cdot Q/V_R + E/V_R \quad (2b)$$

$C_a$ : tracer gas concentration in outside air [mass / volume]

$C_i$ : tracer gas concentration in the indoor air [mass / volume]

$Q$ : exchange air flow between room and outside [volume / time unit]

$E$ : amount of tracer gas emitted per unit time [mass / time unit]

$V_R$ : room volume

$t$ : time

As stated above,  $Q/V_R$  is defined as the air change rate. When  $Q/V_R$  in Eq. 2b is replaced by  $\lambda$  (Eq. 1) we obtain Eq. 2c:

$$dC_i(t)/dt = -(C_i - C_a) \cdot \lambda + E/V_R \quad (2c)$$

Equation 2c expresses that under constant homogeneous mixing the concentration change of the tracer gas is proportional to the concentration difference between indoor and outdoor spaces ( $C_i - C_a$ ) at time  $t$ , the air change rate  $\lambda$ , and the amount of tracer gas emitted per time unit (emission rate  $E$ ).

By integration we obtain the starting conditions at time 0 ( $C(t=0) = C_0$ ):

$$C_i(t) = C_a + E/(\lambda \cdot V_R) + [C_0 - C_a - E/(\lambda \cdot V_R)] \exp(-\lambda t) \quad (3a)$$

Expanding and transposing yields:

$$C_i(t) = (C_0 - C_a) \exp(-\lambda t) + C_a + E/(\lambda \cdot V_R) [1 - \exp(-\lambda t)] \quad (3b)$$

Integration of the differential equation thus leads to an exponential function with the air change rate  $\lambda$  in the exponent. Equation 3b is the basis for the mathematical analysis of tracer gas measurements that are recorded as concentration-time curves. This function is

characterised by the following features: If no tracer gas is emitted into the room (i.e.  $E = 0$ ), and there is already a non-zero tracer gas concentration  $C_0$  present at the time  $t = 0$  which is higher than the outdoor air concentration  $C_a$ , then the expression  $(C_0 - C_a) \exp(-\lambda t) + C_a$  describes the elimination of the tracer gas out of the room. The curve starts with the initial concentration  $C_0$  and decays exponentially until the ambient tracer gas concentration  $C_a$  or any other constant background concentration is reached. If a tracer gas is used, which does not occur in the outside air (i.e.  $C_a = 0$ ) the concentration decreases over time to the value zero. Equation 3b can be simplified to:

$$C_i(t) = C_0 \exp(-\lambda t) \quad (3c)$$

If a source in the room is emitting tracer gas with a constant rate  $E$  and if the initial tracer gas concentration in the room at the beginning of the measurement  $C_0$  is equal to the outside concentration  $C_a$  ( $C_0 = C_a$ ), then the tracer gas will accumulate within the room. Starting with the concentration  $C_a$ , the mathematical function grows with increasing  $t$  asymptotically towards  $C_{eq}$ , which reaches the final value of  $C_a + E/(\lambda \cdot V_R)$  at infinity.

$$C(t) = C_a + [E/(\lambda \cdot V_R)][1 - \exp(-\lambda t)] \quad (4a)$$

When the outdoor air concentration can be neglected, then:

$$C(t) = [E/(\lambda \cdot V_R)][1 - \exp(-\lambda t)] \quad (4b)$$

For  $t \gg 1/\lambda$  Equation 4b can be simplified to:

$$C_{eq} = E/(\lambda \cdot V_R) \quad (4c)$$

$C_{eq}$ : equilibrium concentration

Equation 4c reflects the fact that under equilibrium conditions (i.e. emission equals elimination) the tracer gas concentration  $C_{eq}$  is proportionally dependent on the emission rate  $E$  but inversely proportionally dependent on both the air change rate  $\lambda$  and the volume of the room  $V_R$ . This relationship forms the basis for the determination of the air change rate with tracer gas measurements made under equilibrium conditions.

### 3. Determination of the air change rate (ACR) using tracer gases

In addition to the physical properties discussed in Section 2.1 (model assumptions) tracer gases should fulfil some other requirements in regard to their practical suitability. These include health safety aspects, low environmental burden, high availability and good handling in practical use at the lowest costs possible and, not least, tracer gases should be well recordable with established measurement techniques over a wide concentration range and with high selectivity (Raatschen, 1995).

#### 3.1 Requirements for ideal and commonly used tracer gases

In the past, a number of gaseous substances such as helium, hydrogen, oxygen, carbon monoxide, methane, acetone, and the radioactive noble gases argon-41 and krypton-85, have been studied and tested for the determination of ACR, mostly in comparison with other tracer gases. Reviews are given by Grimsrud et al. (1980), Shaw (1984), and Sherman (1990). Until the early 1990s, krypton-85 was still used for air change measurement (Schulze &

Schuschke, 1990), but later skipped for safety reasons (radiation protection). Nowadays, the following tracer gases are mainly used in practice (Raatschen, 1995):

- Nitrous oxide ( $N_2O$ )
- Sulphur hexafluoride ( $SF_6$ )
- Halogenated hydrocarbons, such as hexafluorobenzene ( $C_6F_6$ ) and perfluorocarbons (PFC).

### 3.1.1 Nitrous oxide

Formerly, nitrous oxide ( $N_2O$ ) was widely used for air change measurement in buildings, primarily in Europe (Heidt & Werner, 1986; Keller & Beckert, 1994; Salthammer, 1994; Wegner, 1983, 1984). In the U.S. it was rarely used because of its low TLV (threshold limit value) of 50 ppm (Lagus & Grot, 1997). Germany's equivalent of TLV, the MAK, is however 100 ppm. For precautionary reasons it should not be used in occupied buildings (Raatschen, 1995). Other disadvantages of  $N_2O$  are the ease of adsorption on surfaces at concentrations below 1000 ppm, and its high solubility in water, which means that the air change rate can be substantially overestimated in very airtight rooms (Schulze & Schuschke, 1990).

### 3.1.2 Sulphur hexafluoride ( $SF_6$ )

After Gregory's observation in 1962, that sulphur hexafluoride ( $SF_6$ ) can be measured reliably on the nanogram scale with electron capture detection – ECD (Gregory, 1962), it has been widely used for air infiltration measurement in buildings since the early 1970s (Drivas et al., 1972; Hunt & Burch, 1975). Of all candidates, the characteristics of  $SF_6$  are nearest to the ideal of a tracer gas. Today,  $SF_6$  is the most frequently used tracer gas worldwide, which is confirmed by the number of relevant publications. In this chapter, only on a very limited selection of publications could be considered (Chuah et al., 1997; Howard-Reed et al., 2002; Kumar et al., 1979; Lagus & Grot, 1997; Raatschen, 1995; Shaw, 1984; Walker & Forest, 1995; Wilson et al., 1996).  $SF_6$  is very stable and only decomposes above 550 °C. Background concentration of  $SF_6$  in ambient air is  $\approx 1$  ppt ( $6 \text{ ng/m}^3$ ) (Raatschen, 1995). Although its density is about five times higher than that of air, this difference causes no systematically distorting effects on the results of air change measurements with concentrations usually applied in practice (Niemelä et al., 1991; Shaw, 1984).  $SF_6$  can be used in occupied buildings. It can be recorded with high accuracy within a wide concentration range. According to Raatschen (1995) the concentrations commonly used for indoor air change measurement are not exceeding one hundredth of the German MAK value which was defined to be 1000 ppm or  $6100 \text{ mg/m}^3$  (TRGS 900, 1999). It should be noted that this value is not a toxicity limit. It is simply defined as the still manageable analytical upper limit for gases which are not imminently toxic (BIA-Report, 2001). Due to its high stability  $SF_6$  is only very slowly degraded in the atmosphere and it belongs, like the perfluorinated hydrocarbons, to the climatic relevant greenhouse gases. Therefore, to avoid unnecessary environment hazard it should be used carefully and sparingly in concentrations as low as possible, like other tracer gases.

### 3.1.3 Hexafluorobenzene ( $C_6F_6$ ) and perfluorocarbon-hydrocarbon tracers (PFT)

Hexafluorobenzene, and perfluorocarbon-hydrocarbon tracers (PFT) such as perfluorodimethylcyclobutane, perfluoromethylcyclobutane, perfluorodimethylcyclohexane, and perfluoromethylcyclohexane are also appropriate as tracer gases and are

preferably used in the determination of ACR with the constant injection method applying the passive sampler technique. Enrichment for active sampling is also possible (Cheong & Riffat, 1995; Dietz & Cote, 1982; Dietz et al., 1986; Krooß et al., 1997; Mailahn et al., 1989; Salmon et al., 2000). Since these compounds, unlike SF<sub>6</sub>, adsorb well on activated carbon or Tenax they are frequently used in field studies (surveys) to determine indoor air change rates (Andersen et al., 1997; Bornehag et al., 2005; Hirsch et al., 2000; Lembrechts et al., 2001; Øie et al., 1997, 1998; Parker, 1986; Pandian et al., 1993; Ruotsalainen et al., 1992; Sakaguchi & Akabayashi, 2003). The disadvantage of these compounds, however, is that they tend to attach to room surfaces and the emission rates of these gases are strongly temperature dependent (Hill et al., 2000). Thus, a sufficiently long conditioning period is necessary as well as an accurate temperature control of the storage vessels.

### 3.2 Methods for the determination of ACR

Basically, three appropriate methods exist for the determination of ACR using tracer gases. According to VDI (2001) 4300, part 7, these are the concentration decay method, the constant injection method, and the constant concentration method.

#### 3.2.1 Concentration decay method

Tracer gas is injected into the room for a short period of time, either from a gas bottle with pressure reducer or manually from filled gas tanks. After mixing with the room air the tracer gas concentration is measured at regular time intervals. Because the decay curve of the tracer gas concentration  $C$  follows an exponential course when completely mixed with the room air (see Eq. 3c) each sampling will not only remove old air but also a certain amount of fresh air supply as well. This essentially means that there will be still  $\approx 37\%$  ( $100/e^1$ ) of the originally added tracer gas (37% old room air), after a complete air change cycle has occurred. The time ( $1/\lambda$ ), after which the air change cycle is completed is known as the nominal time constant  $\tau$  (Maas, 1997; Sherman, 1990). After  $3\tau$  ( $3/\lambda$ ),  $4\tau$  ( $4/\lambda$ ) and  $4.6\tau$  ( $4.6/\lambda$ ), the tracer gas concentration in the room volume under study is 5%, 2% and 1% of its initial value, respectively (Fig. 1a).

By using the concentration decay function

$$C(t) = C_0 \cdot e^{-\lambda t} \quad (5)$$

and applying non linear regression analysis we can determine the ACR  $\lambda$  ( $C_0$ : tracer gas concentration at time  $t=0$  (Sherman, 1990)). However, in most cases logarithmic concentration values are used to obtain a linear relationship between the logarithm of the tracer gas concentration  $C_i(t)$  and the time  $t$  (Fig. 1b):

$$\ln C_i(t) = \ln C_0 - \lambda \cdot (t_i) \quad (5a)$$

The ACR  $\lambda$  is then calculated via linear regression analysis according to Eq. 5a. Both evaluation options - the linear and the non-linear regression analysis - are particularly well suited for the examination of concentration-time curves recorded over a longer period of time. If only a few measurements are available, e.g. when sampling is performed with syringes or other appropriate devices, then the ACR can be determined using the following relationship

$$[\ln C_i(t=t_i) - \ln C_i(t=t_{i+1})] / (t_{i+1} - t_i) = \lambda. \quad (5b)$$

$C_i(t_i)$  is the tracer gas concentration at time  $t_i$

$t_{i+1} - t_i$  is the time interval between two measurements

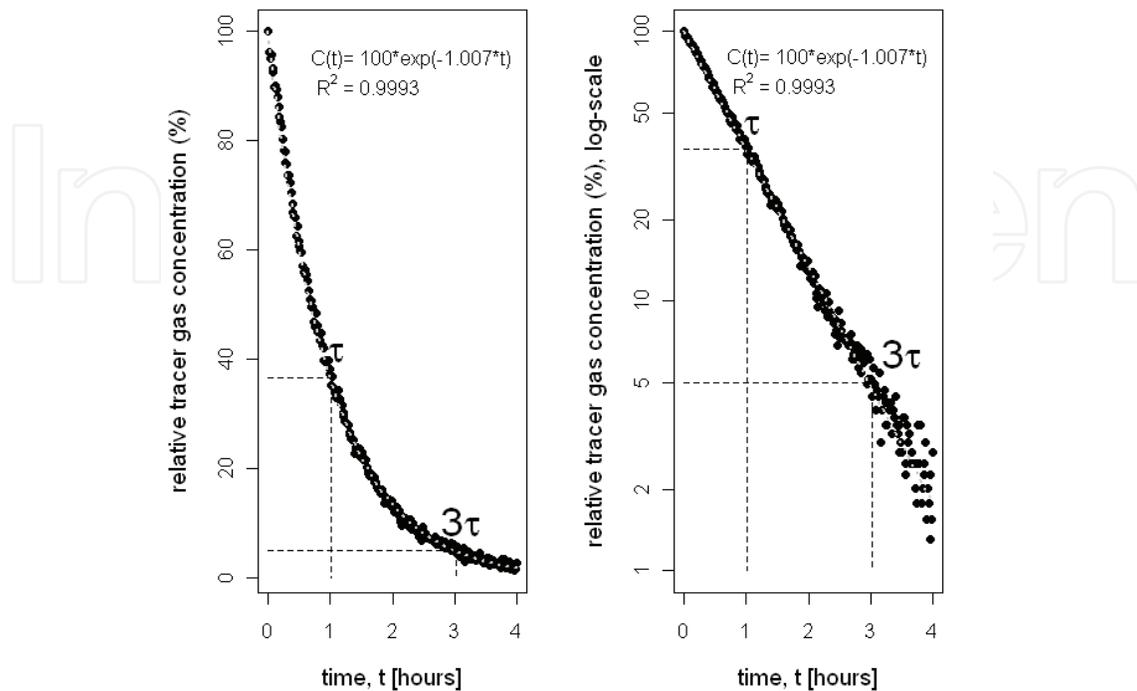


Fig. 1. Relationship between tracer gas concentration and time; a, untransformed ordinate; b, after logarithmic transformation.

An illustration of the procedure is given in Fig. 2a. Sulphur hexafluoride ( $\text{SF}_6$ ) was released in an office space (volume  $48 \text{ m}^3$ , equipped with two double glazed box windows). Initial concentration of the tracer gas was adjusted to  $\approx 5 \text{ mg/m}^3$ . The concentration was measured for 7 hours in 15 minutes intervals using a photo-acoustic infrared detector with selective filter (Brüel & Kjaer Single Gas Monitor). During the measurement the office was unoccupied; door and windows were kept closed. Tracer gas and room air were not continuously mixed with a fan or similar device. For the analysis,  $\text{SF}_6$  concentration values were chosen in such a way that each concentration-time pair -  $c(t_{i+1}), c(t_i)$  - covered exactly a time interval of one hour, meaning that the denominator was equal to 1 when determining the air change rate (ACR). After choosing the six measurement points depicted in Fig. 2a we calculated an average ACR of  $0.327 \text{ h}^{-1}$ . The standard deviation was  $0.038 \text{ h}^{-1}$ , which corresponds to a coefficient of variation of  $\approx 12\%$ . When the same measuring points were analysed by linear regression analysis we obtained an ACR of  $0.323 \text{ h}^{-1}$  (Fig. 2b). Both values are almost equal. The determination of ACR using concentration-time pairs is also known as the two-point method (Sherman, 1990). For this method, a number of at least five concentration-time pairs are recommended which should be evenly distributed over the entire measurement period (ASTM E 741 - 00; VDI, 2001). In order to reduce the statistical error to  $\leq 10\%$  the time interval between the first and the last measuring point should be in the order of magnitude of the nominal time constant (ASTM E 741 - 00; Heidt & Werner, 1986; Maas, 1997; Sherman, 1990). This prerequisite is, however, hardly fulfilled for ACR between  $0.1$  and  $0.2 \text{ h}^{-1}$  or even lower, since the required minimum decay time would be between 5 and 10 hours or more. Most probably, in the case of very low ACR a statistical

error of more than 10% must be accepted. Recommended minimum time intervals between the first and the last measurement and the corresponding measurement intervals are summarised in Table 1, according to ASTM E 741 – 00 and VDI (2001) .

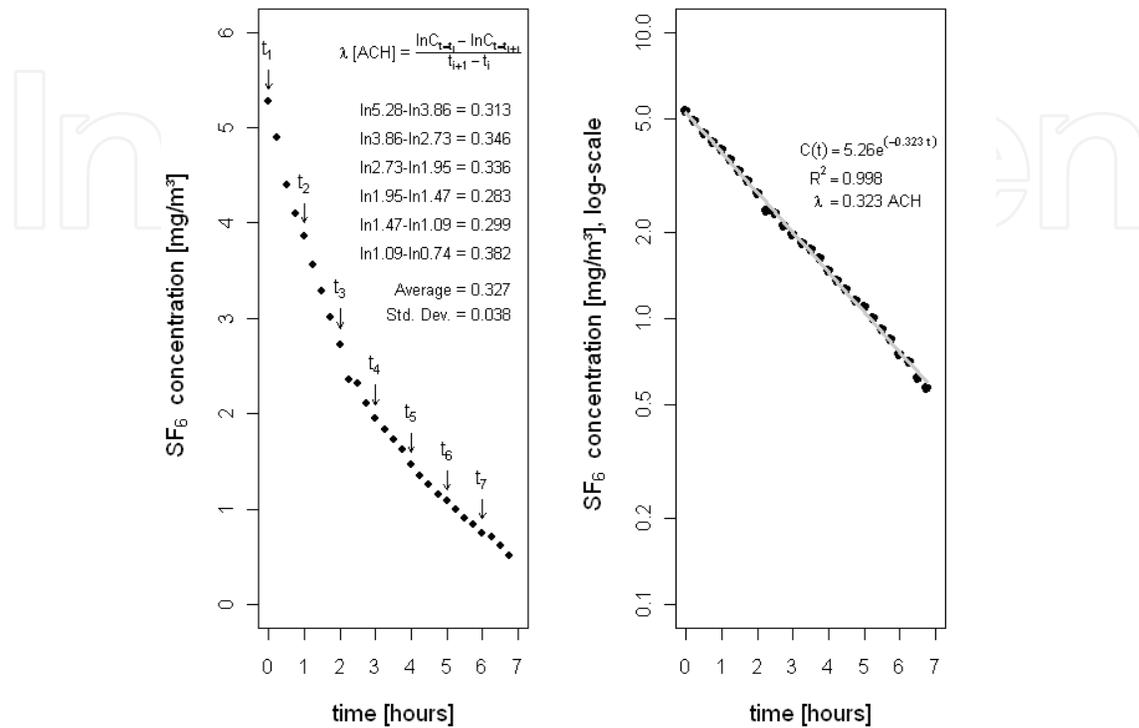


Fig. 2. ACR determination from concentration decay with the two-point method, time interval  $t_{i+1} - t_i$  between two measurements: one hour; a, linear scale; b, logarithmic scale, linear regression.

Air change rate [ACH]	Minimum time span of tracer gas measurement [hours]	Air change rate [ACH]	Sampling interval [min]
0.05	20		
0.125	8		
0.25	4	< 0.5	30 - 40
1	1	0.5 to 1	20 - 30
2	0.5	1 to 2	10
4	0.25	2 to 5	5
10	0.1	> 10	< 2

Table 1. Examples for minimum time spans between first and last sampling based on recommendations of ASTM E 471 – 00 and recommended sampling intervals (VDI, 2001) for air change measurement with the decay method.

The concentration decay method is the most commonly used one in practice. It is particularly well suited for the determination of ACR up to  $10 \text{ h}^{-1}$  (10 ACH) in indoor rooms with a volume below  $500 \text{ m}^3$  (VDI, 2001). If data loggers are used to record the concentration decay, the measurement intervals can be shortened down to seconds and in consequence  $\text{ACR} > 10 \text{ h}^{-1}$  can be determined.

### 3.2.2 Constant injection method

To determine ACR with this method a diffusion tube containing  $C_6F_6$  or PFT is frequently used as tracer gas source. A defined amount of tracer gas is constantly emitted over a certain period of time. Thus, the tracer gas concentration increases with time and reaches a stable value (equilibrium concentration) which depends on the room volume  $V_R$ , the air change rate  $\lambda$ , and the emission rate  $E$ . At that time point, one or more air samples are taken and the tracer gas concentration is determined for each sample. The ACR can be calculated after solving Eq. 4c for  $\lambda$ . Fig. 3a depicts an example of the constant injection method.

Crucial for this method is, however, that sampling can only be started when the tracer gas concentration is near the equilibrium. If sampling starts too soon, an overestimation of the ACR will be the consequence. The time to reach approximate equilibrium conditions depends on  $\lambda$  (see Fig. 3b).

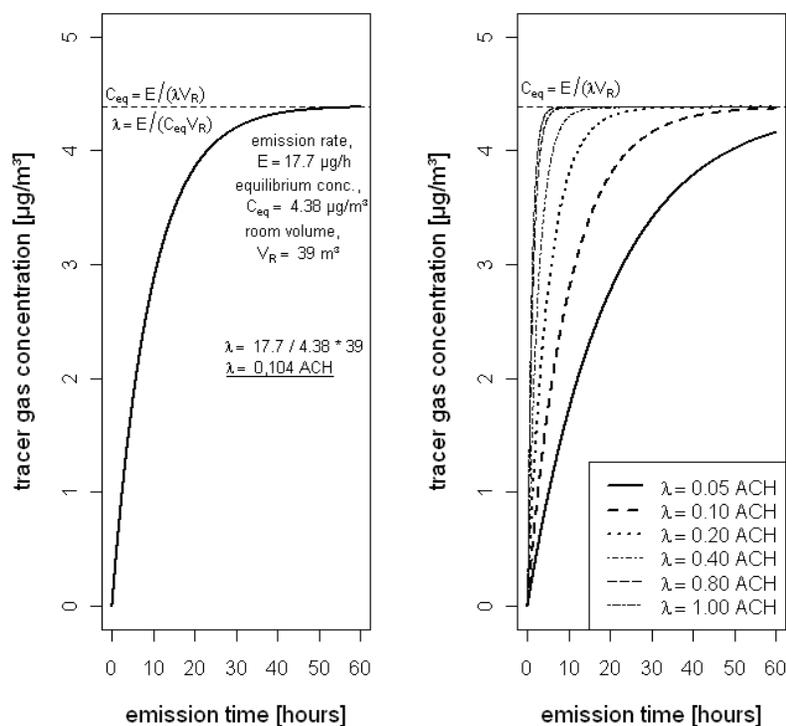


Fig. 3. Determination of ACR; a, constant injection method; b, concentrations after constant injection for different ACR.

According to ASTM E 741 - 00, the tracer gas concentration should have reached at least 95% of the equilibrium concentration before measurements can be started. The time until approximate equilibrium is reached can be estimated applying Eqs. 4b, 4c. However, under worst case conditions in rooms with very low air changes, it will be difficult to correctly measure ACRs with this method, because the build-up time to reach nearly equilibrium concentrations is too long (Table 2).

The time intervals for the tracer gas concentration to reach 95%, 98% or 99% of the target equilibrium value ( $C_{\text{eq}}$ ) are  $3/\lambda$ ,  $4/\lambda$  and  $4.6/\lambda$ , respectively (cf. Table 2).

As Table 2 shows, the time periods needed to reach approximate equilibrium can be several days when the ACR is only  $0.1 \text{ h}^{-1}$  or even lower. For this reason, the constant injection method is suitable only for short-term measurements of ACR above  $0.3 \text{ h}^{-1}$  when diffusion

tubes are used as tracer gas source. Under usual "worst case" conditions (i.e. last active window ventilation 10-12 hrs before sampling) and ACR  $<0.3 \text{ h}^{-1}$  the tracer gas concentrations are still too far away from the equilibrium. The advantage of the constant injection method is that it can be used for indoor hygiene studies addressing the relationship between pollution and air change in daily used rooms. In this case, tracer gas measurements can be performed with passive samplers and exposure times range from days to weeks.

Air change rate [hours]	Time span [hours] to reach nearly equilibrium concentration $C_{eq}$		
	95% $C_{eq}$	98% $C_{eq}$	99% $C_{eq}$
0	-	-	-
0.01	300	400	480
0.05	60	80	92
0.1	30	40	46
0.2	15	20	23
0.3	10	13	15
0.4	7.5	10	11.5
0.5	6	8	9.2
1	3	4	4.6
1.5	2	2.7	3
3	1	1.3	1.5

Table 2. Time spans [hours] needed to reach 95%, 98% or 99% of equilibrium concentration ( $C_{eq}$ ) for a wide range of air change rates.

### 3.2.3 Constant concentration method

During constant and thorough mixing with the indoor air, tracer gas is released in the room until a predefined concentration is reached. During the entire measurement the tracer gas concentration is kept constant with an automated dosing and control system. Under the condition of constant tracer gas concentration the air supply is proportional to the tracer gas supply rate. The air supply rate can then be calculated from the ratio of the tracer gas supply to the tracer gas concentration. If the room volume is known, the ACR can be calculated from this ratio (Chao et al., 2004; Kumar et al., 1979; Maas, 1997). An advantage of the constant concentration method is that even short-term changes of air supply can be detected. Compared to the previously described methods the technical equipment required for this method is, however, rather expensive and thus this method is comparatively rarely used for indoor air quality evaluation.

### 3.3 The use of carbon dioxide as a tracer gas

Carbon dioxide ( $\text{CO}_2$ ) is one of the gaseous organic compounds always detectable in the indoor air. Since humans exhale metabolic carbon dioxide in considerable quantities, its concentration can increase to several thousand ppm ( $\text{ml}/\text{m}^3$  room air) within a short time.  $\text{CO}_2$  concentration is often used to assess the air quality of occupied rooms. In this context we remind of Pettenkofer's reference concentration. Already in 1858 the German chemist and hygienist pointed out that a  $\text{CO}_2$  concentration of 1000 ppm (0.1 vol %) is the upper tolerable limit in indoor environments. Nowadays  $\text{CO}_2$  measurements are often used for the determination of the indoor ACR, because it can be easily quantified and the required

devices are reasonably priced and easy to operate. Moreover, CO<sub>2</sub> fulfils a number of the above mentioned specifications of a good tracer gas.

A huge number of studies are published testing the feasibility of exhaled human carbon dioxide as tracer gas in air change settings. Next to the already mentioned work of Pettenkofer (1858) we would like to allude to the studies done by Penman (1980), Penman & Rashid (1982), and Smith (1988) which are of special importance. Results of Dols & Persily (1992), Nabinger et al. (1994), and Persily (1997) have, however, demonstrated that ACR cannot be reliably determined from spot, peak or average values of the CO<sub>2</sub> concentration inside buildings, because these values are strongly influenced by the number of occupants in the rooms, their times of stay, and, hence, the incessantly changing carbon dioxide supply rates. Depending on the amount of natural ventilation the ACR is sometimes over-estimated up to 2-fold of the real value. The reason is that the air-tightness of modern buildings and the usual sojourn times of the occupants prevent in most cases that the equilibrium concentration can be approached. In practice it is much better to derive the ACR from the decay or build-up curve as was already shown (Barankova, 2005; Bekö et al., 2010; Chao et al., 1997; Chung & Hsu, 2001; Guo & Lewis, 2007; Menzies et al., 1995; Roulet & Foradini, 2002; Schulze & Schuschke, 1990; Sekhar, 2004; Shaw, 1984).

### 3.3.1 Statistical evaluation of CO<sub>2</sub> decay curves (with examples)

After CO<sub>2</sub> is released in a room, either as exhaled breath or via a gas container, its concentration will decay exponentially, if no further CO<sub>2</sub> supply occurs. To exemplify this, the CO<sub>2</sub> decay recorded in a bedroom of an older building (built 1908) is depicted in Fig. 4. In 1990 this bedroom was equipped with a double box window; the room volume is 30 m<sup>3</sup>. The room was doped by a person with exhaled carbon dioxide, then the measurement was started and the room was left. During the entire procedure the room was unoccupied - door and window were kept closed. Measurement of CO<sub>2</sub> concentration was done by a CO<sub>2</sub> sensitive probe with infrared absorption. A Testo 400 device (Testo, Lenzkirch, Germany) was used for data logging and as control unit.

Unlike other tracer gases, CO<sub>2</sub> has the particularity that there is always a certain amount of CO<sub>2</sub> in the outdoor air meaning that the background concentration cannot be neglected. In general the outdoor air concentration of CO<sub>2</sub> is between 350 and 450 ppm or even higher, depending on the season. Thus, the CO<sub>2</sub> decay curve will not decline to zero. Instead, the CO<sub>2</sub> concentration decreases to values which are near to that of the outdoor air. This must be taken into account when analysing CO<sub>2</sub> decay curves. Eq. 5 cannot be applied; the air change rate (ACR) must be determined using the term derived from Eq. 3b:

$$C(t) = (C_0 - C_a) \cdot e^{-\lambda \cdot t} + C_a \quad (6)$$

C<sub>0</sub>: initial concentration, C<sub>a</sub>: concentration in the ambient air (i.e. background concentration)  
It is possible to examine the linearised curve when the background concentration is subtracted before linearising. Therefore, the background concentration must be determined by an additional measurement.

$$\ln(C(t) - C_a) = \ln(C_0 - C_a) - \lambda \cdot t \quad (6a)$$

With this relation the ACR can be determined by linear regression analysis. In practice, however, this will be difficult in most cases since it is not feasible to record the CO<sub>2</sub>-concentration until the decay curve reaches background level. Modern buildings are in

general designed to achieve high airtightness resulting in a very slow decay, and hence the linearisation of the decay curve using the logarithm of the concentration differences is not possible. Furthermore, the background concentrations are often unknown or not accessible to direct measurement. In these cases, the air change rate  $\lambda$  can be determined only by non-linear regression with iterative calculation methods according to the model as given in Eq. 6. Iterative calculation methods start with initial values (raw values) specified by the user for the function parameters, which are then improved iteratively by using the method of least squares until the model function is fitted optimally to the measured curve.

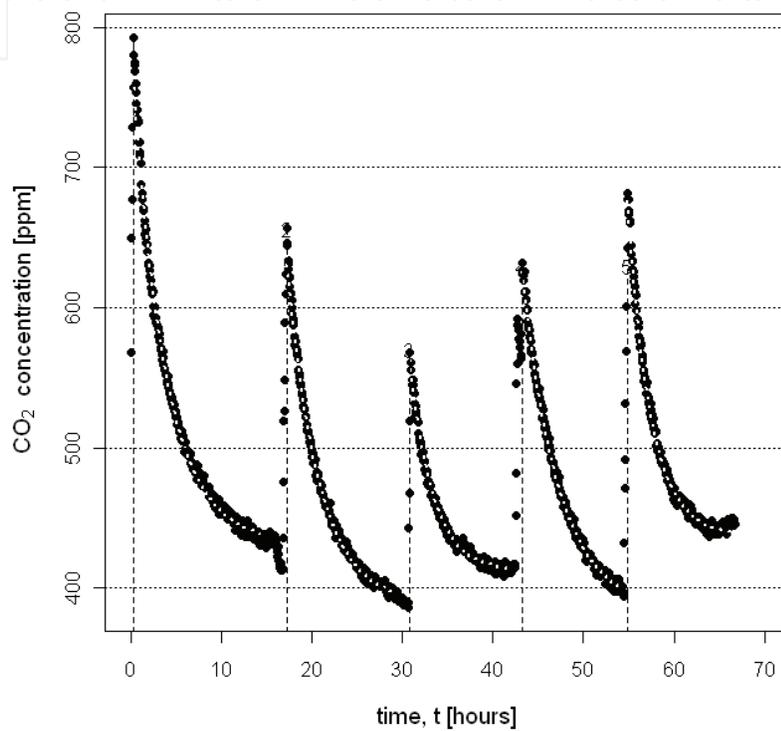


Fig. 4. Decay curves of CO<sub>2</sub> (black glyphs) in a bedroom equipped with double box window and located in a building built in 1908, estimated with nonlinear regression (white curves).

curve	time [hours]	air change $\lambda$ [ACH] (95% CI)	background conc. [ppm] (95% CI)	goodness of curve fit (R <sup>2</sup> )
1	14.5	0.250 (0.246 – 0.254)	428 (427 – 429)	0.997
2	12.8	0.301 (0.295 – 0.306)	391 (390 – 392)	0.997
3	11.3	0.394 (0.383 – 0.406)	412 (411 – 413)	0.992
4	11.0	0.249 (0.243 – 0.254)	384 (382 – 385)	0.998
5	10.8	0.400 (0.393 – 0.407)	438 (437 – 439)	0.998
Avg. 95% CI		0.319 (0.226 – 0.411)		

Table 3. Air change rates in a bedroom located in an older building (built in 1908) equipped in 1990 with wooden framed double box window, decay method and CO<sub>2</sub> as tracer gas. Avg.: Average, CI: confidence interval

The results obtained from the CO<sub>2</sub> decay curves depicted in Figure 4 were evaluated using the method of nonlinear regression and are shown in Table 3. The calculated ACR ranged from 0.25 to 0.4 h<sup>-1</sup> (mean 0.32 h<sup>-1</sup>). The goodness of fit of the regression model is very high, since more than 99% of the variability of the CO<sub>2</sub> concentration can be explained by the respective regression functions. This is also reflected by high accuracy of the estimates for the various ACR. The 95% confidence intervals are very narrow and deviate only about 2 – 3% downward and upward from each individual value. These results were due to both the low variance of the CO<sub>2</sub> values from the fitted curves (white curves in Fig. 4) and the extensive measurement periods (several hours) which yielded high numbers of nodes (concentration-time data pairs) at measuring intervals of 3 minutes. However, the differences between day-to-day measurements are much larger. The 95% confidence interval for the mean ACR ranges from 0.226 to 0.411 h<sup>-1</sup>. A possible explanation for this relatively high day-to-day variation is given below (see section 3.5). ACR, which were determined under the terms of exclusive joint ventilation (i.e. windows and doors closed), vary considerably and show extremes that span two orders of magnitude. This is shown in Figures 5a and 5b where six examples of CO<sub>2</sub> decay curves are depicted which can typically be recorded indoors. For illustrative reasons decay curves were selected which start at about almost the same initial concentration of 1500 – 1800 ppm and were recorded over a measurement period of more than 10 hours (Fig. 5a). Another selection criterion was the existence of stable weather conditions over the entire measurement period. Both figures demonstrate that CO<sub>2</sub> decay curves can decrease exponentially over a period of 50 hours and longer under stable weather conditions (Fig. 5a).

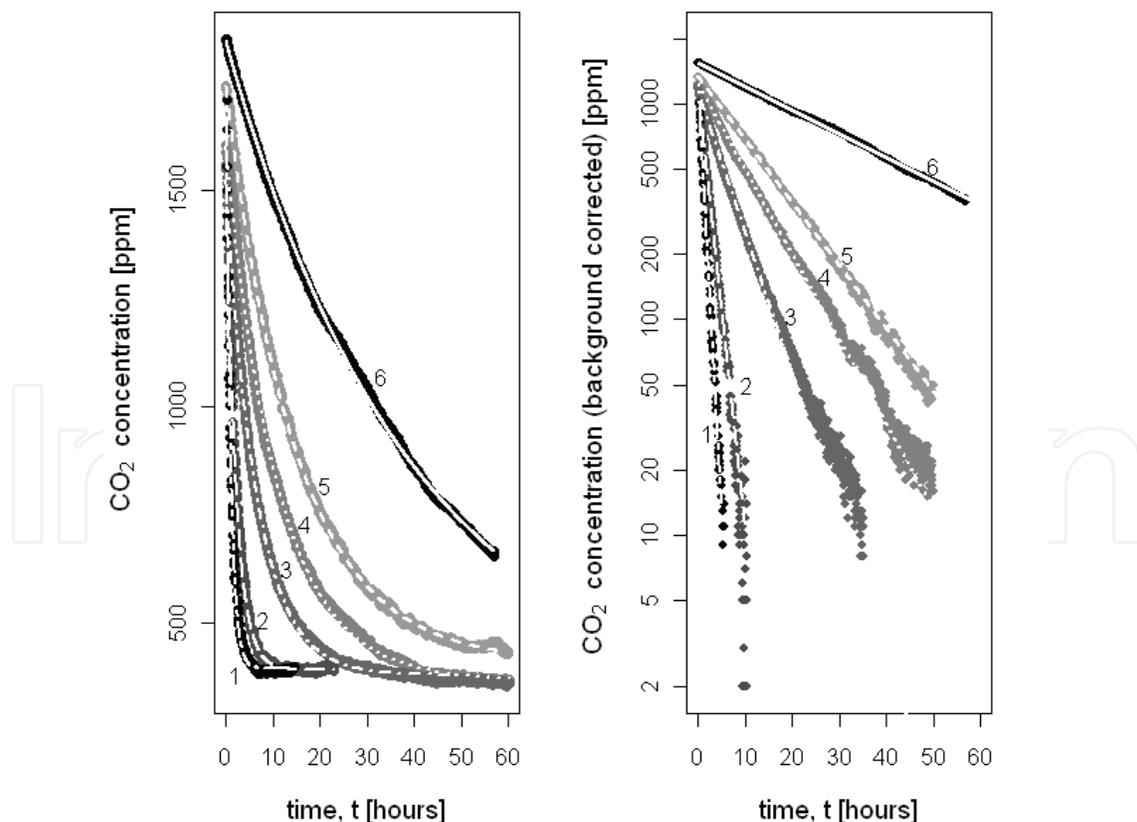


Fig. 5. CO<sub>2</sub> decay curves (black) in 6 rooms of different air tightness. White curves show results of regression analyses; a, nonlinear regression; b, linear regression of logarithmic values

This is an indication that tracer gas and room air can remain homogeneously mixed over such long periods without the need of a fan. The ACR estimated from the decay curves are given in Table 4, and the analysis of the curves was done with non-linear regression analysis (see Eq. 6). In addition to the air change rate  $\lambda$ , the background concentration  $C_a$  was also determined with the regression model and is presented in Table 4. After subtracting the respective background concentration from the measured  $\text{CO}_2$  concentration values, the logarithmic plot of the concentration differences shows a nearly perfect linear concentration-time relationship (Fig. 5b).

curve	time [hours]	air change, [ACH] (95% CI)	background conc. [ppm]	goodness of fit ( $R^2$ )
1	14.5	0.942 (0.931 – 0.953)	393 (391 – 394)	0.998
2	23.0	0.465 (0.463 – 0.468)	392 (391 – 393)	0.999
3	48.0	0.154 (0.153 – 0.155)	380 (379 – 381)	0.998
4	60.3	0.091 (0.091 – 0.092)	367 (366 – 368)	0.999
5	60.0	0.067 (0.066 – 0.067)	406 (405 – 407)	0.9996
6	57.0	0.0255 (0.0253 – 0.0257)	357 (350 – 365)	0.999

Table 4. ACR and background concentrations with their 95% confidence intervals (95% CI) measured in 6 rooms with differing natural ventilation (windows and doors closed); estimated from  $\text{CO}_2$  decay curves with the statistical method of nonlinear regression (time interval between two concentration values: 3 min).

### 3.3.2 Determination of ACR from the concentration increase of carbon dioxide

When  $\text{CO}_2$  is supplied at a constant rate, it is possible to determine the ACR from the increasing concentration values (Figure 6). Evaluation of the concentration curve is the same as with the constant injection method. The air change rate  $\lambda$  is calculated with the non-linear regression model approach  $y = a_0 + a_1[1 - \exp(-a_2 \cdot t)]$  (cf. Eq. 4a), where  $a_0 = C_a$ ,  $a_1 = E/(\lambda \cdot V_R)$ , and  $a_2 = \lambda$ . When the ACR is small, the build-up curve will approach the equilibrium only slowly, and this will lead to a quasi-linear concentration curve if the measurement period is too short. Therefore, the ACR calculated from this curve can be afflicted with large uncertainties. For this reason, to obtain a more reliable estimate of the ACR the concentration build-up must be recorded over a period as long as several hours, with intervals of a few minutes. When metabolic  $\text{CO}_2$  is used, measurements should be done best during sleep (of the occupants), because this comes nearest to the requirement of a constant  $\text{CO}_2$  supply. Figure 6 also shows the ACR determined from the analysis of the  $\text{CO}_2$  respective build-up curves depicted here.

These  $\text{CO}_2$  build-up curves were recorded in the same room as the decay curves in Figure 4. The measurement of the  $\text{CO}_2$  increase occurred, however, four years before, when the room was occupied by 3 persons (two adults, one child). The ACR of both series of measurements, decay and build-up curves, differ only slightly from each other taking daily variation into account.

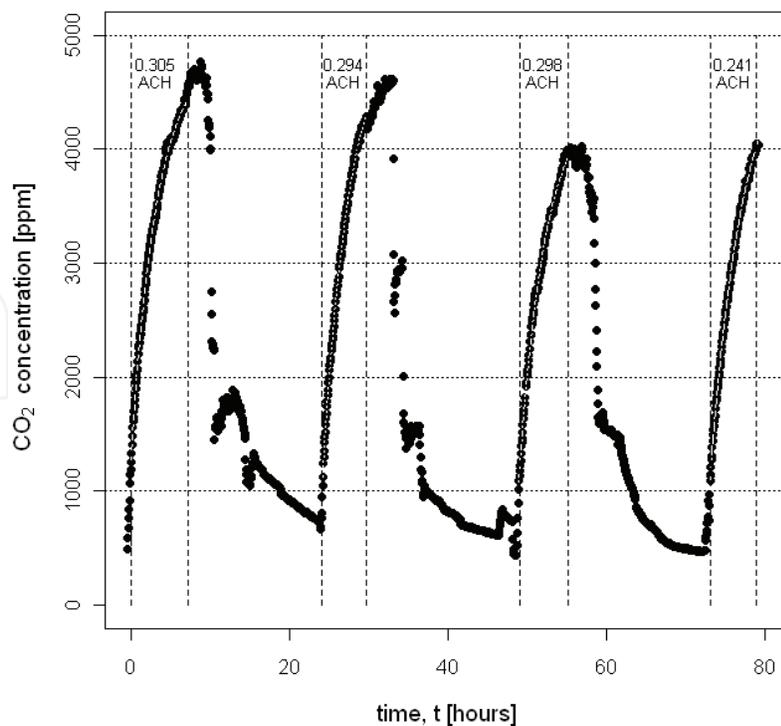


Fig. 6. CO<sub>2</sub> concentration build-up curves in a bedroom equipped with a double box window and located in a building built in 1908. Corresponding ACR, estimated with nonlinear regression (fitted values: white curves). Constant-injection method, room volume 30 m<sup>3</sup>, occupancy: 2 adults, one child.

### 3.4 Comparison of different tracer gases for the determination air change rates

The above examples have shown the applicability of determining ACR from CO<sub>2</sub> concentration curves, when either the influence of the background concentration can be mathematically eliminated or a constant supply of CO<sub>2</sub> is provided. It can, however, not be concluded that conventional and established tracer gases are dispensable. Compared to CO<sub>2</sub> these have some indisputable advantages, namely a much wider range of possible applications and virtually negligible background concentrations. Both of which greatly simplifies the determination of ACR.

Under certain circumstances the use of established tracer gases is not possible, and in these cases we have to resort to the CO<sub>2</sub> method. This may be necessary in patients with environmentally related health problems, who will not accept the use of tracer gases in their homes because they fear health hazards. In such cases, the CO<sub>2</sub> method is a valuable alternative. It is therefore necessary to examine the extent to which results obtained with CO<sub>2</sub> differ from those obtained with conventional and established tracer gases. To achieve this we performed a number of comparative studies by which an established tracer gas like hexafluorobenzene (C<sub>6</sub>F<sub>6</sub>) or sulphur hexafluoride (SF<sub>6</sub>) was used in parallel to CO<sub>2</sub>.

#### 3.4.1 Determination of air change rates with CO<sub>2</sub> and C<sub>6</sub>F<sub>6</sub>

The measurements described below were done in two different rooms, firstly, in a bedroom (24 m<sup>3</sup> effective volume) of a terraced house (built December 1997) equipped with double glazed windows and insulating rubber seals, conducting measurements over a period of 5

days, and secondly, in an office space (72 m<sup>3</sup> effective volume) of an old brick building (built ≈1900) equipped with two double-box windows. Windows and doors were closed during the experiment. A total of two measurement cycles were performed at intervals of two months.

At the start of the conditioning phase a diffusion tube which contained hexafluorobenzene was placed in both rooms. First sampling on Tenax-tubes occurred about 72 hours after the windows and doors were closed, further samples (two samples each) were taken every 24 hours. Room air was collected using sampling tubes (1 litre per tube) with a bellows pump, type Accuro (Dräger, Lübeck, Germany). ACR were calculated from the known emission rate of the C<sub>6</sub>F<sub>6</sub>-tubes, the equilibrium concentration and the effective room volume, according to the procedure for the constant injection method as described in VDI (2001), using Eq. 4c.

In the bedroom, CO<sub>2</sub> concentration was measured continuously over the entire study period with measuring intervals of 3 minutes using the Testo 400-device equipped with CO<sub>2</sub> probe, and the measured values were stored in the internal data logger. In the office space, the CO<sub>2</sub> concentration was measured with a second device in a 24 hour rhythm according to the American standard ASTM D 6245-98. Measurements occurred mainly at nighttime in the then unoccupied office building. Prior to the start of the measurement, the CO<sub>2</sub> concentration in the office space was set to ≈600 ppm by the experimenter. The calculation of the ACR was achieved with non-linear regression analysis using the statistical package SPSS, where the measured concentration values were fitted to exponential terms with consideration of the calculated background concentrations.

Results of the CO<sub>2</sub> measurements:

The CO<sub>2</sub> time-concentration curve recorded for the bedroom is depicted in Figure 7. The bold black line represents the measured concentration values, whereas the grey line describes the graph which was obtained by means of section-wise non-linear regression analysis. Above the individual curve sections, the calculated ACR are plotted. Given the high air-tightness of the bedroom ACR as low as 0.05 to 0.1 h<sup>-1</sup> were determined. It is obvious that the CO<sub>2</sub> concentration increases very quickly during the night (one adult) to levels greater than 4000 ppm and decay exponentially during the day to values of 1500 - 2000 ppm. Due to the low ACR, the equilibrium concentration of CO<sub>2</sub> is not compassed during a 7 hours' sleep. The statistical analysis of the CO<sub>2</sub> curves obtained for the office space equipped with double box windows resulted, however, in significantly higher ACR, which ranged between 0.2 and 1.4 h<sup>-1</sup> because of different window and door features.

Results of the hexafluorobenzene measurements:

In Figure 8, the values of the hexafluorobenzene concentrations from a series of measurements in both rooms are shown together with calculated curves. The solid lines represent the concentration curves which are calculated using Eq. 4b on the basis of the known emission rate  $E$ , the room volume  $V_R$  and the given air change rate  $\lambda$ , whereas the points represent the concentrations that have been measured (bedroom: triangles; office space: squares). It is evident that the measurements in the bedroom oscillate about values which would be expected for an ACR of about 0.05 h<sup>-1</sup>. The reason for the large deviation of the two first measurements from the curve is probably due to largely differing flow resistances of the Tenax tubes used. In comparison to the bedroom, the CO<sub>2</sub> values in the office (lower curve) show a much lower level despite the double emission rate. On the one hand, the lower concentration increase is caused by the larger room volume, but another reason is the significantly higher ACR of 0.45 h<sup>-1</sup>. The calculation of the ACR from the individual concentration values measured led to values ranging from 0.05 to 0.14 h<sup>-1</sup> for the bedroom and from 0.2 to 1.7 h<sup>-1</sup> for the office space, respectively.

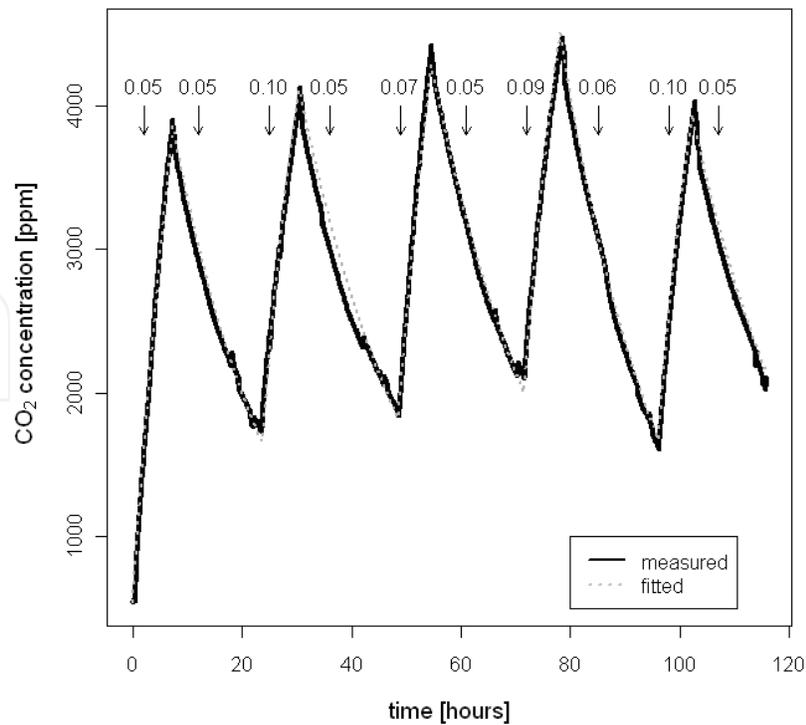


Fig. 7. Course of CO<sub>2</sub> concentration in a bedroom equipped with an insulating glass window with rubber sealing. Window and door closed. Measured values (black) with corresponding air change rates [in ACH], dotted curve: theoretical values. Room volume: 24 m<sup>3</sup>, occupancy: one adult.

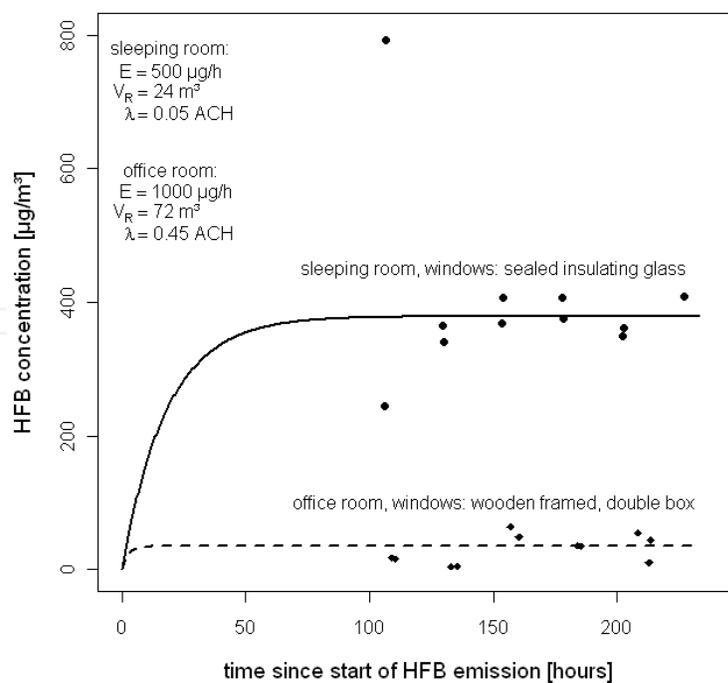


Fig. 8. Course of hexafluorobenzene concentration (HFB) in two rooms with different volumes and different window types, theoretical curves calculated from given emission rates  $E$ , air change rates  $\lambda$ , known room volumes  $V_R$ . Black glyphs: measured values.

The results of the tracer gas comparison are shown in Table 5 for both test series. At large, a very good agreement was found for ACR determined with the two different tracer gases.

building	Window construction	CO <sub>2</sub> (ACH)		Hexafluorobenzene (ACH)	
		serie 1	serie 2	serie 1	serie 2
new	Insulating glass with rubber sealing	0.05 to 0.08	0.05 to 0.1	0.05 to 0.06	0.09 to 0.14
		n = 10	n = 10	n = 5	n = 6
old	wooden framed double box	0.3 to 0.5	0.2 to 1.4	0.2 to 0.9	0.3 to 1.7
		n = 10	n = 10	n = 5	n = 6

Table 5. ACR results of comparative measurements with CO<sub>2</sub> and hexafluorobenzene as tracer gases. Windows and doors closed.

### 3.4.2 Parallel testing of CO<sub>2</sub> and SF<sub>6</sub>

In another series of investigations an office space with a volume of 48 m<sup>3</sup> and two double box windows was doped with CO<sub>2</sub> and SF<sub>6</sub> in parallel on 6 consecutive days, and the ACR were determined from the decay curves. Measurement of SF<sub>6</sub> was done in the same way as described in the above section "decay curve". An example of the parallel concentration decay of both tracer gases is given in Figure 9; results of the 6 experiments, which differ only slightly, are compiled in Table 6.

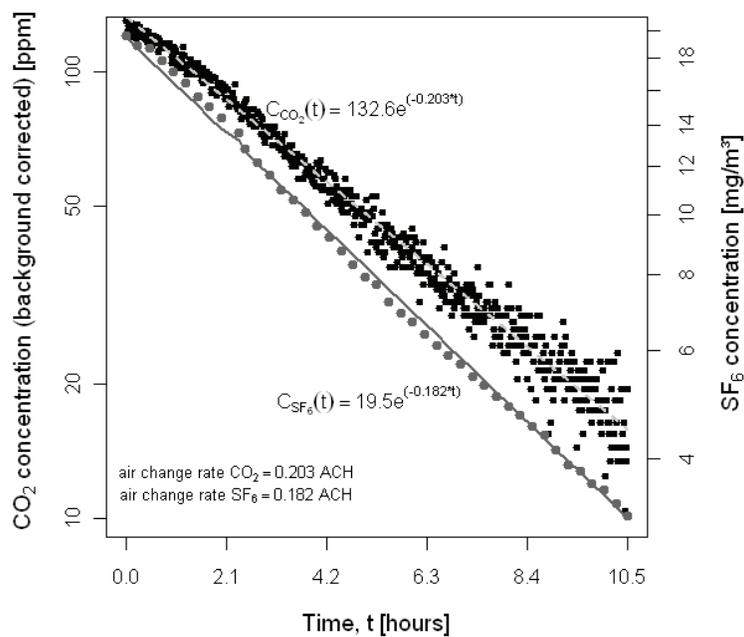


Fig. 9. Decay curves of CO<sub>2</sub> and sulphur hexafluoride (SF<sub>6</sub>) and curve fits; simultaneous measurements in the same room.

measurement	air change rate [ACH]		Quotient of air changes
	CO <sub>2</sub>	SF <sub>6</sub>	ACR <sub>CO<sub>2</sub></sub> / ACR <sub>SF<sub>6</sub></sub>
1	0.211	0.240	0.879
2	0.447	0.438	1.021
3	0.330	0.327	1.009
4	0.203	0.182	1.115
5	0.330	0.320	1.031
6	0.300	0.280	1.071
average	0.304	0.298	1.021
std.dev.	0.090	0.087	0.080
COV (%)	29.7	29.2	8.0

Table 6. Comparative measurements with CO<sub>2</sub> und SF<sub>6</sub>; office space (volume 48 m<sup>3</sup>, two double box windows); COV (%): coefficient of variation = 100\*standard deviation (std.dev.) / average.

The average ACR was 0.304 h<sup>-1</sup> for CO<sub>2</sub> (SD ± 0.09 h<sup>-1</sup>) and 0.298 h<sup>-1</sup> for SF<sub>6</sub> (SD ± 0.09 h<sup>-1</sup>). To check for systematic differences between the individual determinations we calculated the ratio of the two air change rates and got a mean ratio of 1.02 with a standard deviation of ± 0.08. Thus, no statistically significant systematic differences between the compared tracer gases could be detected. Since our sample size was rather small (only 6 determinations for each tracer gas) we included results from other studies (Guo and Lewis 2007; Roulet and Foradini 2002; Shaw, 1984) for comparison. After inclusion of these data the difference was statistically significant, albeit the systematic deviation between the ACR, determined under the same conditions with either CO<sub>2</sub> or SF<sub>6</sub>, was very small (Fig. 10). A similarly good agreement was found by Stavova (Baránková) et al. (2006) comparing CO<sub>2</sub> and Freon 134a. The mean ratio of the two air change rates (CO<sub>2</sub>/SF<sub>6</sub>) is about 1.06 (95% CI: 1.01 - 1.12). Standard deviation ranged from 0.93 to 1.21. The ≈6% overestimation of the ACR, as obtained with CO<sub>2</sub>, is yet much smaller than the 50 percent overestimation obtained by Schulze and Schuschke (1990) who used krypton 85. On the basis of existent data it is so far not possible to conclude with certainty whether or not very small ACR (i.e. 0.1 h<sup>-1</sup>) are responsible for higher systematic deviations.

### 3.5 Wind and temperature effects on the natural air flow in case of joint ventilation

Natural ventilation is driven by the air-pressure difference between the interior and external environment which prevails on the room's opening areas like windows and doors. The air change rate is also influenced by the tightness of windows and doors versus joints and cracks in the building envelope. The air-pressure difference is predominantly caused by temperature differences between indoor and outdoor climate, and by local wind effects. On the windward side of a building, the wind is trapped reaching its highest dynamic power when the flow angle equals 90°. On the sides of a building that parallel the wind direction and on the far side, negative air-pressures occur regularly. These air-pressure differences result in raised rim hole rates, particularly in case of leaks or open windows on opposite sides of a building, due to suction and pressure cycles of the wind (i.e. cross ventilation).

The entire air-pressure onto the façade is dependent on both the wind speed and squalls, as well as the building's constructional quality, namely the tightness of the façade.

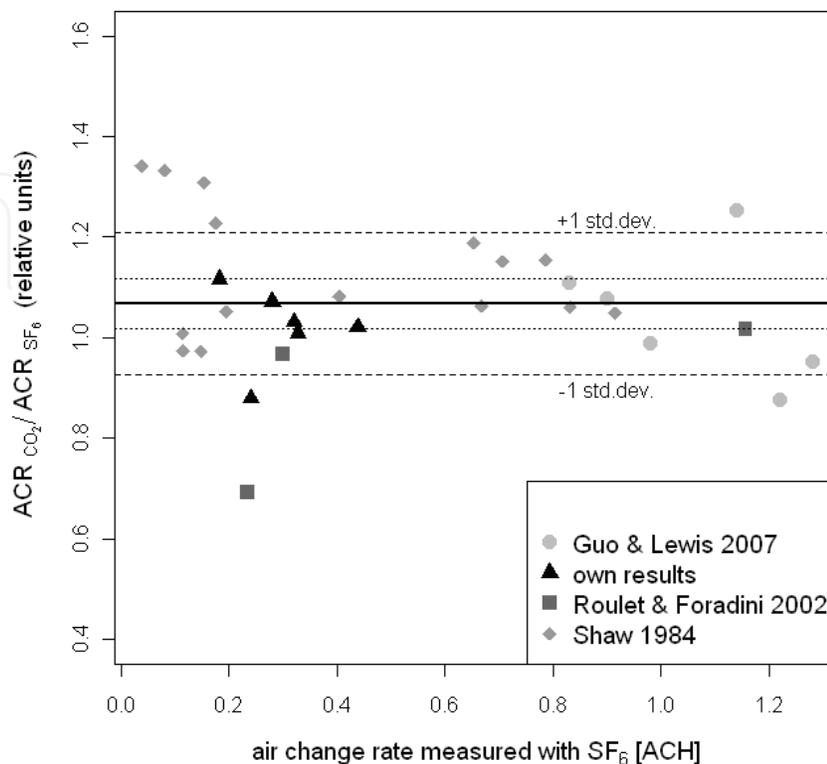


Fig. 10. Comparative air change determinations with  $\text{CO}_2$  and  $\text{SF}_6$  as tracer gases. Results from other authors and own results. Solid line: mean, dotted lines: 95% confidence interval of the mean, dashed lines: interval of  $\pm 1$  standard deviation.

### 3.5.1 Wind influences

To study the effects of weather conditions on the ACR, measurements were conducted in two different office buildings of the Robert Koch Institute, Berlin, Germany. Building #1 is an older brick building (built ca. 1900), surrounded by allotment gardens (to the north and south) and large open spaces. On the east side is a three-storey row house, and on the west side a four-storey office building. Due to the location of the building, unhindered wind flow is limited to south-east or south-west directions. All of the five rooms which have been selected in building #1 were located on the south side and equipped with double box windows (rooms A – D and F; room volumes between 30 and 90  $\text{m}^3$ ).

The rooms in building #2 constructed in the late 1970s were equipped with tightly closing windows featuring single glazing and insulating rubber seals. The room selected for the measurements had a large window sill with a couple of windows in north-east direction and a room volume of 40  $\text{m}^3$  (room E). In front of the windows was an open space (length  $\approx 150$  m), limiting wind flow to solely from north-east to south-east directions. On the south side, room E was protected by a cross-building located in a distance of  $\approx 10$  m. Likewise, on the north side, the building was protected by a north-east to south-west oriented wing of the same building (in a distance of about 70 m from room E).

Between January 1999 and December 2010 a total of 611 air change measurements were performed in the five rooms of building #1 (A – D and F) using the  $\text{CO}_2$  decay method. The

decay curves were mainly recorded at night or on weekends according to ASTM D 6245-98, when the building was unoccupied. Analogously, 388 air change measurements were carried out in room E of building #2 between July 2002 and September 2008. During the measurements all doors and windows were kept closed. Data about meteorological conditions during the tests, such as outside air temperature, wind speed and wind direction were obtained from the web sites of local Berlin weather stations. In room E of building #2 the indoor temperature was recorded during the measurements. Table 7 provides an overview of the air change rates determined. For rooms with double box windows individual values of ACR varied from  $<0.05 \text{ h}^{-1}$  to  $1.8 \text{ h}^{-1}$  (i.e. a factor of  $\approx 40$ ), whereas the medians (50<sup>th</sup> percentile) obtained for the rooms A - D and F varied only between 0.3 and  $0.6 \text{ h}^{-1}$ . Significant differences between the individual rooms could not be detected.

room	sample size	Minimum [ACH]	Median (95% CI)	Maximum [ACH]
A	15	0.209	0.457 (0.369 - 0.715)	1.320
B	26	0.163	0.445 (0.351 - 0.552)	1.370
C	11	0.090	0.470 (0.126 - 0.696)	0.720
D	42	0.079	0.512 (0.352 - 0.639)	1.426
F	517	0.043	0.342 (0.317 - 0.374)	1.841
E	388	0.016	0.078 (0.070 - 0.085)	0.403

Table 7. Air change rates [ACH] in office spaces with windows of different construction, rooms A-D, F: wooden double box windows, room E: insulating glass window with plastic frame and rubber sealing

Significantly lower ACR were determined for room E in building #2 equipped with double glazed windows for which the median was  $0.08 \text{ h}^{-1}$ . The individual values varied between  $0.016$  and  $0.40 \text{ h}^{-1}$ , i.e. maximum and minimum differ more than a magnitude. Wind influences, especially wind speed, were identified as the main reason for the variability of the ACR in the two buildings (Fig. 11).

In building #1 (rooms A - D and F) the ACR ranged from  $<0.1 \text{ h}^{-1}$  when wind speed was lower than  $5 \text{ km/h}$  to  $1.2\text{--}1.4 \text{ h}^{-1}$  at a wind speed up to  $30\text{--}40 \text{ km/h}$ . The strong influence of the wind caused significant and uncontrollable heat losses through leaky windows which, in turn, caused unhealthy draught effects.

Surprisingly, the results obtained for room E in building #2 equipped with insulated windows and rubber seals, were significantly dependent on wind influences, as well, although the wind effects were much smaller than those of building #1. For room E, ACR were less or equal to  $0.05 \text{ h}^{-1}$  when wind speed was lower than  $<5 \text{ km/h}$  but increased up to  $0.25 \text{ h}^{-1}$  with a wind speed of more than  $40 \text{ km/h}$ . The relations described here essentially confirm the results formerly obtained by Wegner (1983). Besides the wind speed, the flow direction of the wind had a significant impact on the amount of the ACR. This is evident from the graphical presentation in Figure 12, grouping ACRs according to the wind direction prevailing in test periods.

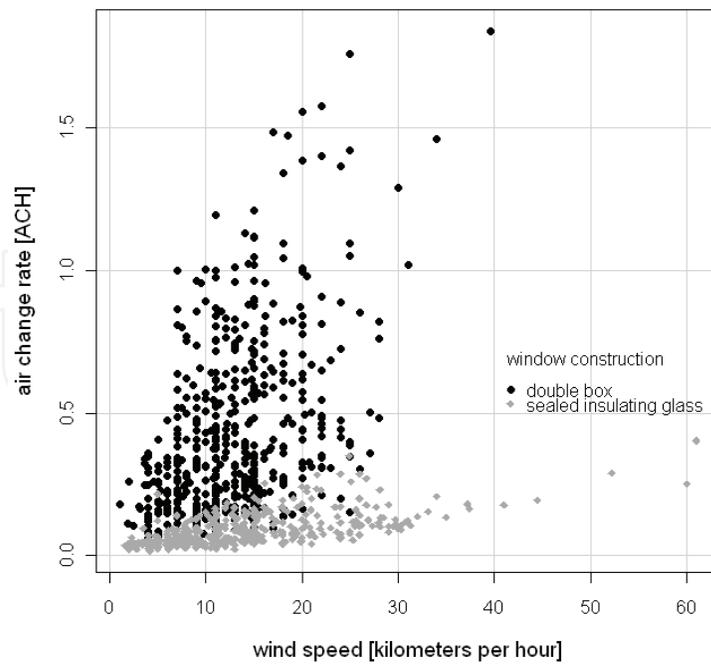


Fig. 11. Influence of wind speed on ACR in office spaces equipped with windows of different construction. Windows and doors closed. Rooms with double box windows (A-D, F: n= 611), room with insulating glass window: n= 388).

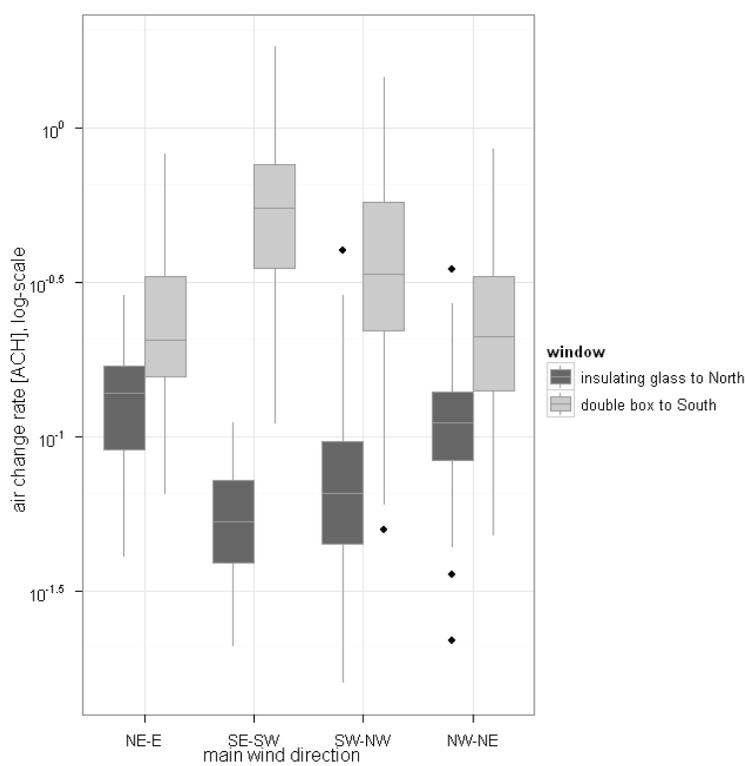


Fig. 12. Wind direction and ACR in office spaces equipped with windows of different construction. Windows and doors closed. Rooms with double box windows (A-D, F: n= 601), room with insulating glass window: n= 388).

Adverse wind blowing against the window or in parallel to the window front are of higher impact than wind flow from the backside of the building, the latter causing lower ACR than the two former (see Table 8).

room	wind direction	sample size	ACR [ACH]	Quotient <sub>raw</sub>	Quotient <sub>adj</sub>
			Median (95% CI)	Median (95% CI)	Median (95% CI)
A - D, F	NE-E	122	0.202 (0.171 - 0.243)	0.92 (0.779 - 1.107)	1.06 (0.941 - 1.189)
	SE-SW	242	0.55 (0.505 - 0.596)	2.50 (2.295 - 2.709)	2.89 (2.601 - 3.207)
	SW-NW	163	0.329 (0.294 - 0.395)	1.50 (1.340 - 1.801)	1.38 (1.239 - 1.547)
	NW-NE	74	0.22 (0.179 - 0.261)	1 (Ref.)	1 (Ref.)
E	NE-E	83	0.138 (0.109 - 0.153)	2.09 (1.651 - 2.317)	2.21 (2.038 - 2.399)
	SE-SW	81	0.054 (0.047 - 0.063)	0.82 (0.714 - 0.957)	1.10 (1.013 - 1.198)
	SW-NW	171	0.066 (0.056 - 0.072)	1 (Ref.)	1 (Ref.)
	NW-NE	53	0.111 (0.095 - 0.135)	1.68 (1.438 - 2.043)	1.92 (1.742 - 2.115)

Table 8. Dependence of ACR on wind direction, raw and adjusted ACR quotients, Rooms A-D, F: windows to south, room E windows to north-east.

In case of the rooms with double box windows, this led to a relative threefold increase of air change, expressed as ratio  $ac(luv) : ac(lee)$ , for wind directions from south to west. In the room with double glazed windows, we measured an almost doubled increase of air change (70 - 100%) for wind directions from north to east. The overall air change ratios depending upon the relative wind direction, respectively were not remarkably influenced by the wind speed (Quotient<sub>adj</sub> in Table 8). Summarising wind effects we could demonstrate that regardless of wind strength and the window type an air flow directed to the window front can yield a 1.5- to more than 2-fold increase of the ACR in comparison to wind from the backside.

### 3.5.2 Temperature influences

The temperature difference between inside and outside is one of the major forces driving ventilation. We investigated the impact of the temperature gradient (room air temperature minus outside air temperature) on the ACR in room E and found a statistically significant effect on the extent of air change (Fig. 14a). Whereas the average air change rate  $\lambda$ , after correction for the wind influence, was only  $0.06 \text{ h}^{-1}$  at a temperature difference  $\Delta T$  of 0 K, it increased up to  $0.1 \text{ h}^{-1}$  when the temperature gradient rose to about 30 K. However, as compared to wind effects (Fig 14b), the exponential relationship between ACR and temperature gradient (Fig. 14a), turned out to be much weaker, at least for room E with its tightly closing window.

### 3.6 Air change rate during window ventilation

Wind action and temperature gradient both proved to be significant factors influencing the amount of air change through joints and cracks in buildings. For indoor spaces in buildings with high leak tightness, window ventilation plays a crucial role in the removal of excess humidity, carbon dioxide, odors and other volatile compounds. The effectiveness of

ventilation via opened windows is not only dependent on their number, size and arrangement in the room, but also on the modus and frequency of active window ventilation. Windows that are only opened partway (i.e. tilted) ventilate less effectively than fully opened windows. Cross-ventilation can be achieved when windows on opposite sides of a room are opened simultaneously. Cross-ventilation removes moisture and contaminants most effectively from the indoor air to the outside. The effectiveness of the different modes of window ventilation expressed in terms of the ACR, was examined with a couple of tests in room E, the results of which are shown in Figure 13 in conjunction with the joint ventilation results.

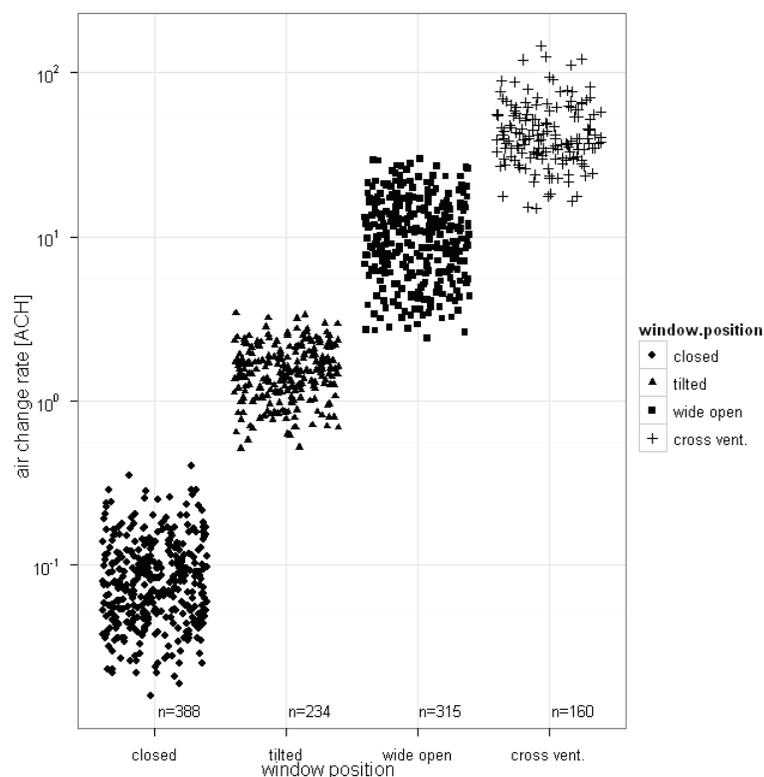


Fig. 13. ACR and window position. Office room E.

ACR determinations were carried out by the decay method; duration of the measurements varied between 0.5 h and 3 h with recording intervals of 12 s to 1 min. After doping the room with  $\text{CO}_2$ , one window in the room (dimensions: width 0.8 m and length 1.2 m) was either in tilted position (i.e. 10 cm wide upper opening) or widely open (i.e. opening angle  $80^\circ$ ). To achieve cross-ventilation, an opposite window was also widely open. For both ventilation modes – tilted or full open – the following ACR were determined (Table 9a): With the tilted window a total of 239 measurements was performed; the average ACR (i.e. median) was  $1.5 \text{ h}^{-1}$  with day-to-day variations that ranged from  $0.5 \text{ h}^{-1}$  to  $3.4 \text{ h}^{-1}$ . In the case of full open window, the median ACR was  $10 \text{ h}^{-1}$ , with a minimum of  $2.4 \text{ h}^{-1}$  and a maximum of  $30 \text{ h}^{-1}$  (321 measurements). For cross ventilation a number of 179 measurements were performed. The ACR varied between  $15 \text{ h}^{-1}$  and  $146 \text{ h}^{-1}$  (median:  $40 \text{ h}^{-1}$ ). On the basis of these data we calculated the ventilation times which are necessary to reduce an indoor  $\text{CO}_2$  concentration of 1500 ppm by 98% (corresponding to a  $\text{CO}_2$  concentration that is about 30 ppm higher than outside) and yielded 2 hours for the tilted window (range 1

- 7 hours),  $\approx 0.3$  hours for the wide open window (range 0.1 - 1.5 hours) and 0.1 hours for cross-ventilation (range 1 - 14 min; see also Table 9b).

room	window construction	window position	Air change rate [ACH]					sample size
			Min	05. P	Median	95. P	Max	
A - D, F	double box	closed	0.043	0.111	0.36	1.030	1.841	602
E	insulating glass	closed	0.016	0.029	0.078	0.200	0.403	388
		tilted	0.52	0.70	1.50	2.60	3.43	239
		wide open	2.4	3.5	10.4	23.4	29.7	321
		cross vent.	15.0	22.5	40.1	104.7	146.4	179

Table 9a. Dependence of ACR on window construction and opening

room	window construction	window position	Ventilation time [hours]				
			Max	95. P	Median	05. P	Min
A - D, F	double box	closed	84	32.4	10	3.5	2.0
E	insulating glass	closed	225	124	46	18	8.9
		tilted	6.9	5.1	2.4	1.4	1.0
		wide open	1.5	1.0	0.35	0.15	0.12
		cross vent.	0.24	0.16	0.09	0.03	0.02

Table 9b. Ventilation time needed to reduce CO<sub>2</sub> concentration of 1500 ppm by 98%

These results clearly confirm the great importance of ventilation with fully open windows and, even more, of cross-ventilation for the efficient removal of indoor pollutants. The broad variation in ACR can be explained to a great deal by the effect of the temperature gradient between indoor and outside (Fig. 14a).

Compared to the temperature gradient, wind effects played only a minor role at least for the window openings "tilted" and "widely open" tested here (Fig. 14b). The dependence of the ACR on the temperature gradient could be estimated by non-linear exponential curve fitting for all tested window opening modes (Fig. 14a). From these findings we can conclude that in case of ventilation via tilted window, with a temperature gradient of 10 K, an ACR of  $0.92 \cdot \exp(0.045 \cdot 10) = 1.4 \text{ h}^{-1}$  can be expected for the room examined here (95% prediction interval:  $1.0 \text{ h}^{-1} - 2.0 \text{ h}^{-1}$ ). With a temperature gradient of 20 K an average ACR of  $2.3 \text{ h}^{-1}$  can be expected (95% prediction interval  $1.6 \text{ h}^{-1} - 3.1 \text{ h}^{-1}$ ). For ventilation with fully open window an ACR of  $4.5 \cdot \exp(0.071 \cdot 10) = 9.2 \text{ h}^{-1}$  ( $\Delta T = 10 \text{ K}$ ) can be expected and  $18.6 \text{ h}^{-1}$  ( $\Delta T = 20 \text{ K}$ ) with 95% prediction intervals  $5.5 \text{ h}^{-1} - 15.3 \text{ h}^{-1}$  and  $11.1 \text{ h}^{-1} - 31.2 \text{ h}^{-1}$ , respectively. In the case of cross ventilation an average ACR would expected to be  $39.0 \text{ h}^{-1}$  and  $50.2 \text{ h}^{-1}$  for  $\Delta T = 10 \text{ K}$  and  $20 \text{ K}$ , with 95% prediction intervals of  $23.0 \text{ h}^{-1} - 65.8 \text{ h}^{-1}$  and  $29.6 \text{ h}^{-1} - 85.1 \text{ h}^{-1}$ , respectively.

### 3.7 Air change rates in Berlin's housing stock

The results presented so far do not allow general statements on the air change in buildings, since the number of rooms examined was far too small. As part of site inspections and indoor air investigations during 1999–2005, ACR were determined for a further number of different buildings and rooms applying the CO<sub>2</sub> concentration decay method. A total of 198 living and working spaces, situated in 152 buildings in the Berlin area and in the immediate surrounding were studied, comprising of 143 residential buildings of different

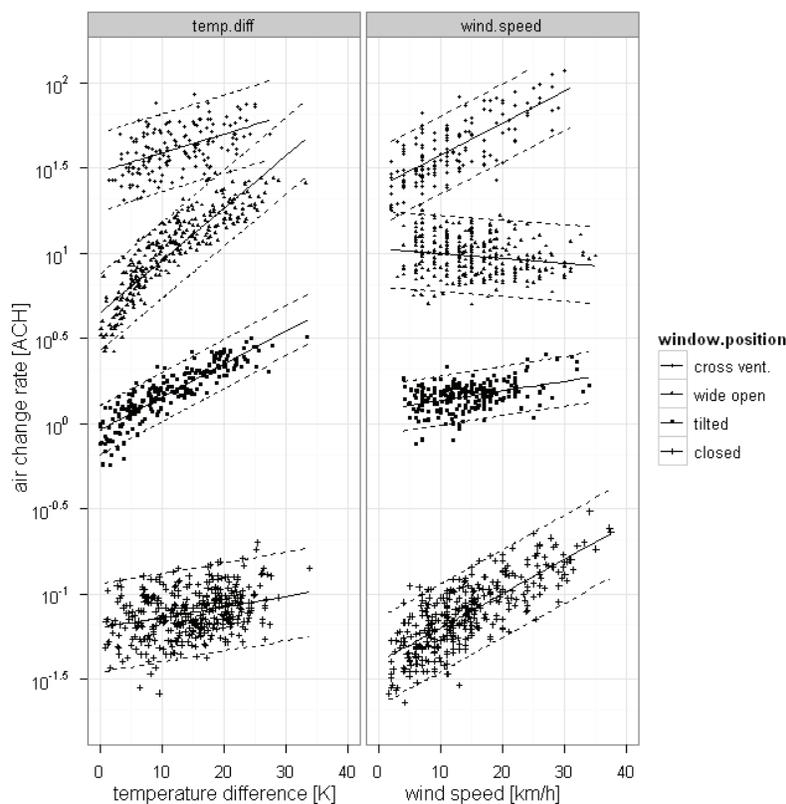


Fig. 14. a. Influence of temperature gradient (indoor to outdoor) on ACR, corrected for wind effects, b. Influence of wind speed on ACR, corrected for temperature effects, in an office space (room E) for different ventilation conditions: natural ventilation: "closed", "window tilted", "widely open" and "cross ventilation", (from bottom to top); room volume  $40 \text{ m}^3$ , solid lines: fitted curve, dashed lines: 95% prediction intervals.

age and storey numbers, and nine different office buildings. None of the rooms had an additionally installed ventilation system. Tenants were asked not to enter the rooms during the tracer gas measurements and to keep windows and doors closed. The duration of the tracer gas measurements varied between 2 hours and 24 hours. To obtain decay curves with a high number of nodes we choose time intervals of 1 to 3 minutes between  $\text{CO}_2$  measurements. Two Testo 400 devices with  $\text{CO}_2$  probe (infrared absorption) were used for recording and data logging. The results show that under worst case conditions the vast majority of the ACR are below the range of  $0.5 \text{ h}^{-1} - 1 \text{ h}^{-1}$  which was recommended by the former German Federal Health Office (Bundesgesundheitsamt, 1993) for common living quarters. The ACR of the studied rooms ranged from minimum  $0.02 \text{ h}^{-1}$  to maximum  $1.98 \text{ h}^{-1}$ . Of the 198 naturally ventilated rooms the majority (167=84%) had ACR of  $0.5 \text{ h}^{-1}$  or below. ACR of  $0.8 \text{ h}^{-1}$  or higher could be determined for only 12 of the rooms (6%). The 50<sup>th</sup> percentile (median) of the ACR was  $0.2 \text{ h}^{-1}$  with a 95% confidence interval (CI) of  $0.17 \text{ h}^{-1} - 0.24 \text{ h}^{-1}$ ; arithmetic mean was  $0.31 \text{ h}^{-1}$  (95% CI  $0.27 \text{ h}^{-1} - 0.36 \text{ h}^{-1}$ ) and geometric mean was  $0.22 \text{ h}^{-1}$  (95% CI  $0.20 \text{ h}^{-1} - 0.25 \text{ h}^{-1}$ ), respectively (see Table 10). Thus, our studies essentially confirm the results obtained by Münzenberg (2004) and Salthammer et al. (1995), who conducted ACR measurements under similar methodological conditions in Germany, and received results consistent with those from surveys in Scandinavia by Andersen et al. (1997); Bornehag et al. (2005) and Harving et al. (1992).

Measurements performed in the same rooms using diffusion tubes and hexafluorobenzene ( $C_6F_6$ ) or PFT as a tracer gas yielded somewhat higher ACR compared to that obtained with the  $CO_2$  concentration decay method. This can be explained by necessarily longer exposure times of collection tubes ranging from several days to weeks, which essentially means that there have been one or more active ventilation phases during the measurement period influencing the air change (Table 10). However, even in these studies, the proportion of rooms with ACR below  $0.5\ h^{-1}$  was remarkably high. Studies conducted in the USA also show a high proportion of buildings (50 percent and more) with ACR below  $0.5\ h^{-1}$  (Persily et al., 2010), suggesting that a basically desirable ACR of at least  $0.5\ h^{-1}$  is difficult to achieve under normal conditions. Similar conclusions have been drawn by Erhorn and Gertis (1986) and Münzenberg (2004).

Stratification for window types showed significantly higher ACR (factor  $\approx 2$ ) for rooms with double box windows or composite windows than for rooms with specially insulated windows (double glazing and continuous rubber seal; Table 11). However, also for rooms with double box windows or composite windows the majority of the ACR determinations yielded values  $< 0.5\ h^{-1}$ . The 50<sup>th</sup> percentile (median) was  $0.33\ h^{-1}$ , and the 75<sup>th</sup> percentile was about  $0.56\ h^{-1}$ . In comparison, rooms with double glazed windows had a median ACR of  $0.16\ h^{-1}$  and the 75<sup>th</sup> percentile was  $0.25\ h^{-1}$ .

study	method	tracer gas	sample size	Min [ACH]	Median [ACH]	Max [ACH]	AM [ACH]	GM [ACH]	< 0.5 ACH (%)
Salthammer et al. (1995)	concentration decay	$N_2O$	150	< 0.1	0.25	1.7	0.36	n.a.	> 70
Münzenberg (2004)		$CO_2$	80	< 0.05	0.18	> 1.5	0.26	n.a.	90
own results		$CO_2$	198	0.02	0.21	1.98	0.31	0.22	84
Harving et al. (1992)		TFE	114	< 0.1	0.28	> 1.5	n.a.	n.a.	72
Bekó et al. (2010)	concentration decay and build-up	$CO_2$	300 cases		0.44		0.62	0.46	57
			200 bases		0.42		0.62	0.46	
Hirsch et al. (2000)	constant-injection (diffusion tubes)	$C_6F_6$	78					0.73 <sup>1)</sup>	
								0.52 <sup>2)</sup>	
Andersen et al. (1997)		PFT	117	0.16	n.a.	0.96	0.37	0.34	82
Ruotsalainen et al. (1992)		PFT	242	0.07	$\approx 0.5$	1.55	0.52	n.a.	48
Øie et al. (1997)		PFT	38	0.15	n.a.	1.4	n.a.	n.a.	58
Øie et al. (1998)		PFT	344	n.a.	n.a.	n.a.	n.a.	n.a.	36
Bornehag et al. (2005)		PFT	320 <sup>3)</sup>				0.36 <sup>3)</sup>		80
			43 <sup>4)</sup>				0.48 <sup>4)</sup>		60

Table 10. ACR measurements in Germany and Scandinavia, results of surveys. TFE: 1.1.2-trichloro-1.2.2-trifluoroethane, <sup>1)</sup> before reconstruction, <sup>2)</sup> after reconstruction, <sup>3)</sup> single family houses, <sup>4)</sup> multi-family houses, AM: arithmetic mean, GM: geometric mean, n.a. not available,  $< 0.5\ h^{-1}$ : proportion (%) of air change rates  $< 0.5\ h^{-1}$

window construction	Sample size	Min	05. P	25. P	Median (95 % CI)	75. P	95. P	Max	AM (95 % CI)	GM (95 % CI)
Double box/ composite window	80	0.05	0.11	0.20	0.33 (0.27 – 0.44)	0.56	1.50	1.98	0.47 (0.37 – 0.57)	0.34 (0.28 – 0.41)
Insulating glass	118	0.02	0.05	0.10	0.16 (0.13 – 0.20)	0.25	0.54	1.00	0.22 (0.18 – 0.25)	0.16 (0.14 – 0.19)

Table 11. ACR in naturally ventilated rooms (windows and doors closed) equipped with different window types. Min: Minimum. Max: Maximum. 05. P: 5<sup>th</sup> percentile. 25. P: 25<sup>th</sup> percentile. 75.P: 75<sup>th</sup> percentile. 95. P: 95<sup>th</sup> percentile. 95% CI: 95% confidence interval. AM: arithmetic mean. GM: geometric mean

The empirical frequency distribution of the ACR for the total sample had the typical shape of a log-normal distribution with a steep rise on the left and a long tail on the right side (Fig. 15a). A good fit to the normal distribution could be achieved by calculating the logarithms of the ACR (Fig. 15b).

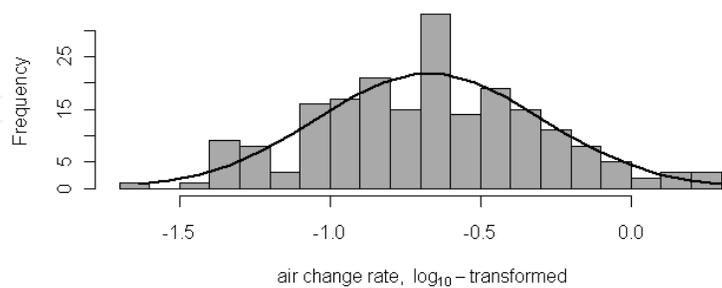
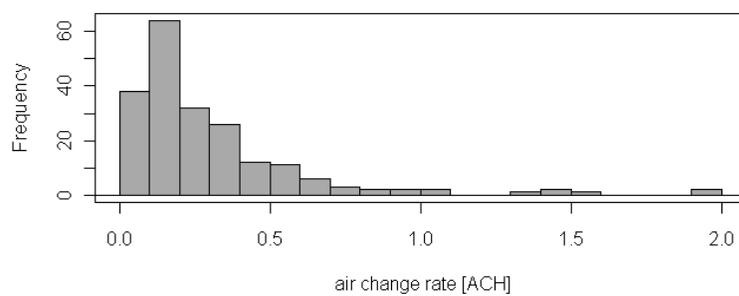


Fig. 15. Empirical frequency distribution of ACR (natural ventilation) for 198 rooms in the City of Berlin; a, values as determined; b, log-transformed values. Solid line represents normal distribution.

This is consistent with the results of Scandinavian studies by Andersen et al. (1997); Bekö et al. (2010); Harving et al. (1992); Øie et al. (1998) and Ruotsalainen et al. (1992) who found the same form of empirical frequency distributions of ACR in Danish, Finnish and Norwegian residential buildings.

### 3.8 Ventilation and pollutant concentrations

Adequate ventilation is the cheapest and easiest way to effectively remove indoor pollutants. Previous studies have shown that insufficient air change is mainly due to renovation and other structural changes, namely in most cases the installation of new airtight windows (Wegner 1983, 1984). The results of our own investigations presented here have led to similar conclusions, apart from significantly lower ACR as compared to previous studies (Wegner 1983, 1984). Generally insufficient fresh air supply resulted in a considerable increase of indoor air pollutants. This is the case for formaldehyde (Li et al., 2002; Salthammer et al., 1995; Wegner, 1983) and other volatile compounds (Hodgson et al., 2003) as well as for the radioactive noble gas radon (Andersen et al., 1997; Chao et al., 1997; Sentikova, 1999). Our own studies also found a correlation between the concentration of volatile organic compounds and the ventilation rate in naturally ventilated rooms, as described below.

We have chosen some frequently found indoor air pollutants, which may be taken as typical representatives of different substance classes, namely formaldehyde, acetaldehyde, acetone, 2-ethyl-1-hexanol, hexanal, d-limonene,  $\alpha$ -pinene, decamethylcyclopentasiloxane (D5-siloxane), and toluene. Air sampling to analyse these contaminants was done in the rooms under study (closed windows and doors), either after a CO<sub>2</sub> tracer gas measurement was finished or directly before the start of a CO<sub>2</sub> decay recording with Aircheck samplers (model 224-PCXR8) SKC Inc., Eighty Four, PA, USA. For enrichment we used the adsorption media recommended for these pollutants. Air volumes varied from 90 l to 120 l and the sampling air flow was 1.5 – 2 l/min. Formaldehyde was somewhat special since it was measured semi-quantitatively in situ in most rooms. An Interscan 4000 device (Ansyco, Lenzkirch, Germany) was used to carry out the measurements. Prior to the determination of the formaldehyde concentration, a zero balance was produced by pumping air through an upstream activated carbon filter for 10 minutes. Thereafter, the concentration value was recorded when the reading remained constant. Comparison of measurements for 24 different rooms yielded a good agreement of concentration levels obtained with the electrochemical formaldehyde method using the Interscan 4000 and the laboratory results obtained by analysis of the air samples that were collected with DNPH cartridges.

Statistics for the pollutants investigated here are listed in Table 12; if the sample size was sufficient the 95% confidence intervals were also included. Acetone, formaldehyde and acetaldehyde showed the highest concentrations, with median values of 75  $\mu\text{g}/\text{m}^3$ , 23  $\mu\text{g}/\text{m}^3$  and 31  $\mu\text{g}/\text{m}^3$ , respectively. Due to boiling points below 60° C, these pollutants are classified as very volatile organic compounds (VVOC). Therefore, they are – with the exception of formaldehyde – not regularly recorded indoors.

As Figure 16 shows, the frequency of formaldehyde concentrations  $>100 \mu\text{g}/\text{m}^3$  seems to correlate with the ACR. In 8 of 77 category-1 rooms with ACR  $<0.2 \text{ h}^{-1}$  formaldehyde amounted to  $\geq 100 \mu\text{g}/\text{m}^3$ , i.e. the frequency of exceeding values was 10.4% (95% CI 4.6% – 19.5%). Among the category-2 und -3 rooms was only one each with exceeding formaldehyde (1.6% and 3.2% relative frequency, 95% CI 0% – 8.5% and 0% – 16.7%, respectively). According to X<sup>2</sup> testing these differences were close to be significant ( $p = 0.060$ ), but this seems unlikely due to strong overlaps of the very broad 95% CI. Our finding thus points to successful emission reduction measures through low-emission products by which pollutant concentrations can be accomplished that are within the range of the accepted limits, even in very airtight rooms.

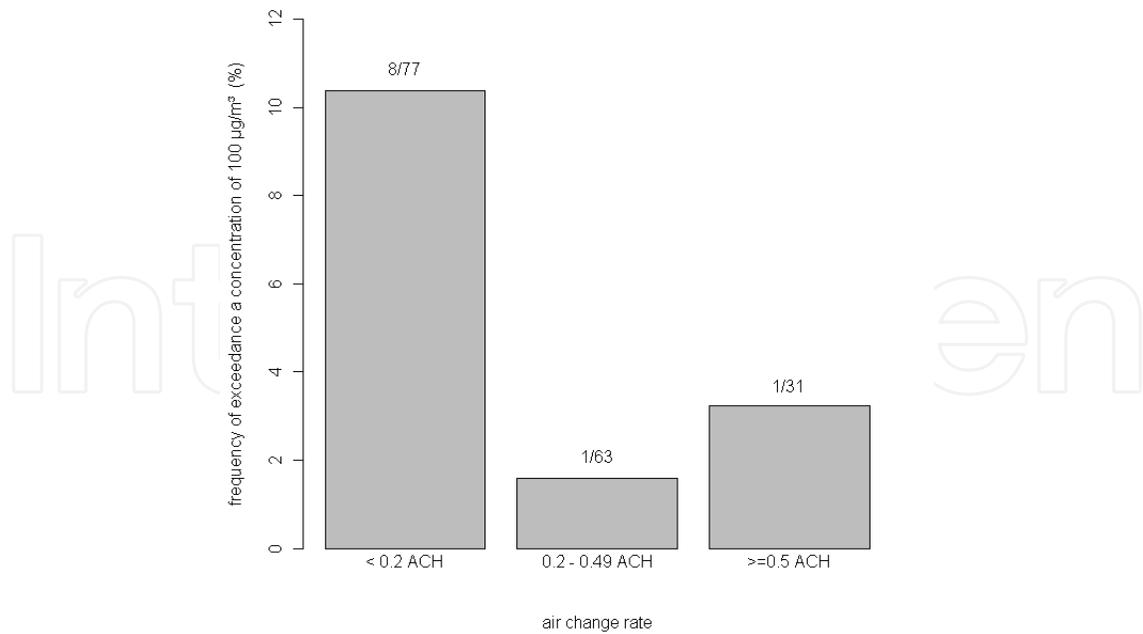


Fig. 16. Frequency of formaldehyde concentrations  $>100 \mu\text{g}/\text{m}^3$  and ACR (natural ventilation) in rooms of the City of Berlin.

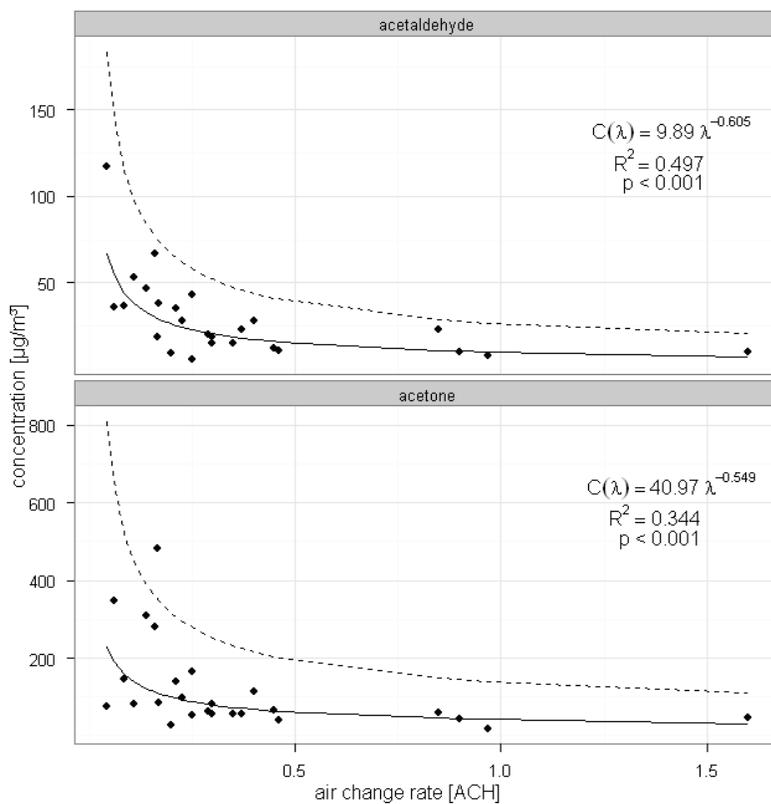


Fig. 17. a. and b. Concentrations of acetaldehyde (a) and acetone (b) and corresponding air change rates in rooms of the City of Berlin. Sampling: worst case conditions, solid line: mean concentration, dotted line: 90% prediction limit.

Unlike formaldehyde, the concentrations of acetaldehyde and acetone showed a clear dependence of the air change rate  $\lambda$ , which could be described by the power function ( $C(\lambda) = a \cdot \lambda^{-k}$ ). The independent variable air change rate could explain about 50% ( $R^2 = 0.5$ ) of the total variance of the acetaldehyde concentration values and about 34% ( $R^2 = 0.34$ ) of the acetone concentration values. The results of a curve fitting are shown in Fig. 17a and b.

According to the resulting function parameters it can be estimated that an increase of the ACR from  $0.1 \text{ h}^{-1}$  to  $0.5 \text{ h}^{-1}$  would yield a reduction of the indoor concentrations from  $40 \text{ }\mu\text{g}/\text{m}^3$  to  $15 \text{ }\mu\text{g}/\text{m}^3$  for acetaldehyde, and from  $145 \text{ }\mu\text{g}/\text{m}^3$  to  $60 \text{ }\mu\text{g}/\text{m}^3$  for acetone. This is a reduction factor of 2.4 – 2.7.

adsorbent	compound	Sample size	Min	10. P (95 % CI)	Median (95 % CI)	90. P (95 % CI)	MAX	AM (95 % CI)	GM (95 % CI)
	very volatile								
In situ detection	formaldehyde	171	< 20	< 20 (< 20)	31 (25 – 38)	70 (63 – 96)	190	41 (36 – 45)	33 (30 – 37)
DNPH-Silica	acetaldehyde	25	6	9 (n.d.)	23 (11 – 37)	53 (n.d.)	117	29 (19 – 39)	22 (16 – 30)
DNPH-Silica	acetone	25	18	39 (n.d.)	75 (53 – 128)	308 (n.d.)	481	118 (71 – 166)	84 (60 – 117)
	volatile								
Anasorb 747 / Tenax	2-ethyl-1-hexanol	148	< 1	1 (1 – 1,5)	4 (3 – 6)	12 (10 – 19)	65	7 (5 – 8)	4 (4 – 5)
Anasorb 747 / DNPH	hexanal	93	< 2	4 (3 – 6)	17 (14 – 21)	37 (30 – 45)	75	20 (17 – 22)	14 (12 – 17)
charcoal (NIOSH)	limonene	133	< 1	2 (1 – 3)	10 (8 – 13)	66 (36 – 89)	256	24 (17 – 31)	10 (8 – 13)
charcoal (NIOSH)	$\alpha$ -pinene	133	< 1	2 (2 – 5)	15 (11 – 22)	102 (57 – 143)	623	45 (28 – 62)	16 (12 – 20)
charcoal (NIOSH)	d5-siloxane	99	< 1	< 1 (<1 - 1)	3 (2 – 4)	94 (39 – 321)	736	39 (17 – 61)	4 (3 – 6)
charcoal (NIOSH)	toluene	133	2	4 (3 – 5)	14 (10 – 21)	91 (46 – 144)	396	34 (25 – 43)	16 (13 – 20)

Table 12. Concentrations [ $\mu\text{g}/\text{m}^3$ ] and 95% confidence intervals of selected VVOC and VOC in the air of rooms in the City of Berlin; n.d.: 95% confidence interval not detected (small sample size), 10. P: 10<sup>th</sup> percentile, 90. P: 90<sup>th</sup> percentile, AM: arithmetic mean, GM: geometric mean.

Similar relations were found for other volatile organic compounds (Table 12). Again, the dependence of their indoor air concentrations on the ACR could be characterized by a power function (see Figures 17c to 17h). Regression analysis revealed that an increase of the ACR from  $0.1 \text{ h}^{-1}$  to  $0.5 \text{ h}^{-1}$  decreased the concentrations of 2-ethyl-1-hexanol and hexanal (or hexanaldehyde) by a factor of  $\approx 2.3$ , since 2-ethyl-hexanol declined from  $6.2 \text{ }\mu\text{g}/\text{m}^3$  to  $2.6 \text{ }\mu\text{g}/\text{m}^3$ , and hexanal declined from  $21.2 \text{ }\mu\text{g}/\text{m}^3$  to  $9.1 \text{ }\mu\text{g}/\text{m}^3$ . For limonene and  $\alpha$ -pinene the reduction factors were 2.6 and 2.2, respectively. For these pollutants, the indoor air concentrations would be reduced from  $24 \text{ }\mu\text{g}/\text{m}^3$  and  $30 \text{ }\mu\text{g}/\text{m}^3$  to values of  $9 \text{ }\mu\text{g}/\text{m}^3$  and  $14 \text{ }\mu\text{g}/\text{m}^3$ , respectively. For decamethylcyclopentasiloxane (D5) and toluene the reduction factors amounted to 2.7 and 2.4, corresponding to a decline  $6.5 \text{ }\mu\text{g}/\text{m}^3$  to  $2.4 \text{ }\mu\text{g}/\text{m}^3$  (D5) or  $32 \text{ }\mu\text{g}/\text{m}^3$  to  $13 \text{ }\mu\text{g}/\text{m}^3$  (toluene).

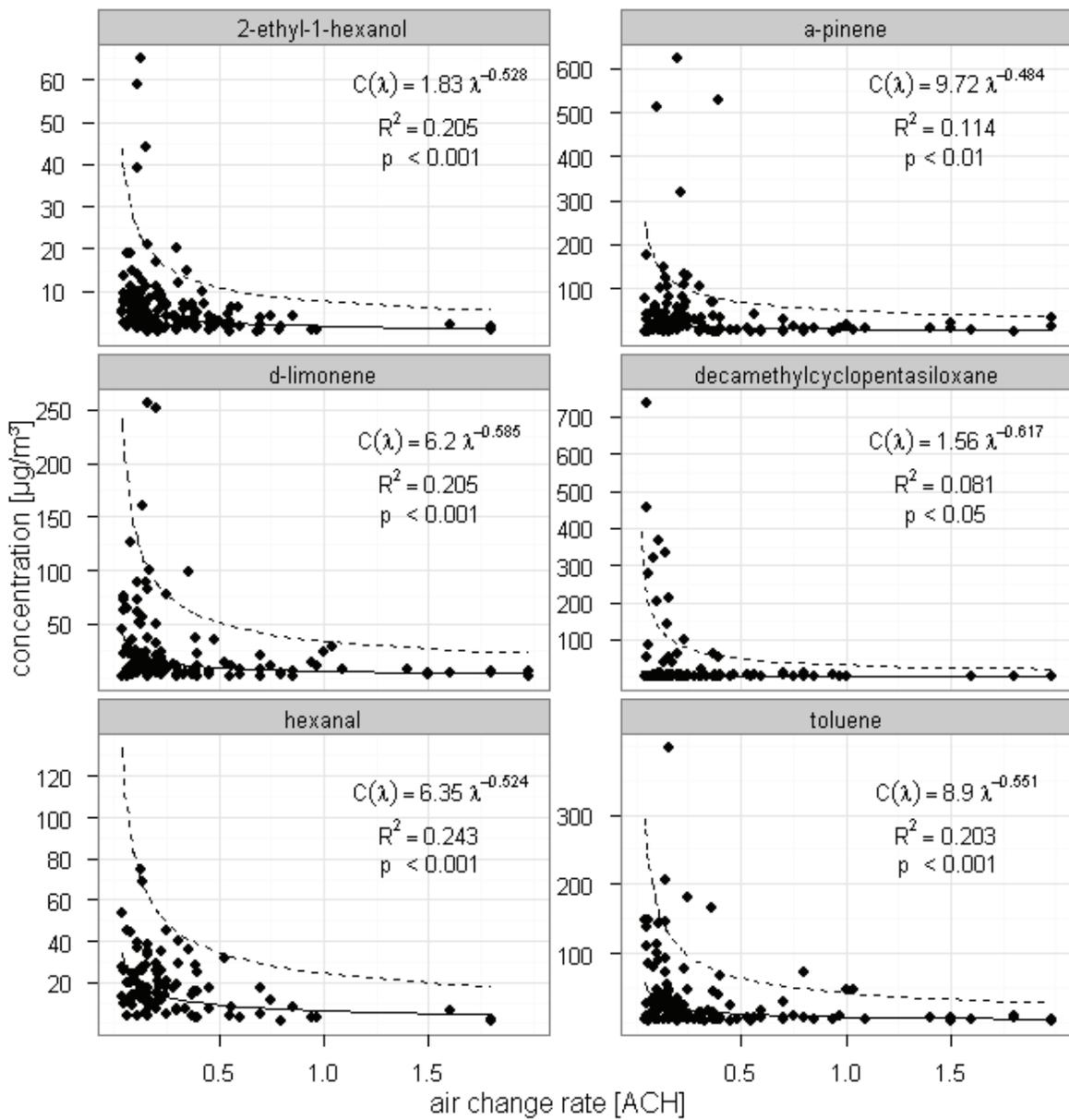


Fig. 17. c-h. Concentrations of selected volatile organic compounds (VOC) and corresponding ACR in rooms of the City of Berlin. Sampling under worst case conditions, solid line: mean concentration, dotted line: 90% prediction limit.

These results may allow to conclude that increasing the ACR to the fourfold will reduce indoor air concentration of VVOC and VOC to half. This means that the concentration is inversely proportional to the square root of the ACR, but seems to be contradictory to theoretical considerations, if one assumes a nearly constant emission rate of volatile compounds. In this case, the concentration would be inversely proportional to the ACR under equilibrium conditions, meaning that a fourfold increase in the ACR would reduce the concentration to a quarter.

One of the reasons for the deviating results is surely the fact that nowadays many buildings are constructed with such a high air-tightness that a kinetic equilibrium cannot be achieved under typical sampling conditions (i.e. time span of several hours between last window ventilation and start of the measurement; see also Figure 3b). That leads, firstly, to low ACR ( $<0.2 \text{ h}^{-1}$ ) and, secondly, to an underestimation of the VOC concentrations. The regression analysis yields a flatter curve and an exponent  $k < 1$  for the air change rate  $\lambda$ . Another reason is the different interior decoration (furniture, wallpapers, other equipment) of the spaces investigated here, and thus their differing pollution loads. Our VOC sample comprised rooms which differed greatly in respect to number and kind of possible sources. Therefore, we can assume that even in rooms with low air change either no VOC sources existed or only such sources with low emission rates. As a consequence, only low concentrations could have been measured in the air of these rooms. This fact would also contribute to the flattening of the air change-concentration curve.

The influence of ACR on the concentrations of VOC is also valid when we consider substance classes. The spectrum of individual compounds of these classes resembles that described by Schleibinger et al. (2001, 2002). As with formaldehyde, the analysis was performed with grouped data to reveal the dependency on the ACR, and categorisation was done as above. For determining of frequencies of exceeding concentrations the individual measurement values were summed up. Subsequently, the sum values were dichotomised on the basis of the medians of the empirical distributions. The empirical medians agreed quite well with the limits proposed by Schleibinger et al. (2002) – especially when taking into account the 95% confidence intervals of the percentiles (Table 13). Exceptions here are the siloxanes, for which the 90<sup>th</sup> percentile was clearly above Schleibinger's recommendation (Schleibinger, 2002). The marked deviation from previous studies (Schleibinger et al., 2001; Scholz, 1998) may indicate an increased (and still increasing) use of siloxanes as constituents of coating materials, paints, cosmetics etc., thus contributing more than ever to indoor air pollution.

Compound class	Sample size	Median (95% CI)	90. P (95% CI)	MAX	AM (95% CI)	GM (95% CI)	Target value	Reference value
aldehydes (C4 – C10)	93	49 (36 – 59)	88 (76 – 127)	203	51 (43 – 58)	37 (30 – 45)	50	120
aromatics	133	39 (30 – 51)	206 (149 – 248)	1140	82 (60 – 103)	45 (37 – 54)	50	200
siloxanes (D3 – D5)	99	4 (2 – 6)	125 (51 – 338)	741	43 (21 – 65)	5 (4 – 8)	5	10
terpenes und sesquiterpenes	133	51 (36 – 61)	230 (150 – 297)	1008	103 (73 – 133)	47 (37 – 58)	40	150

Table 13. Indoor air concentrations and 95% confidence intervals [ $\mu\text{g}/\text{m}^3$ ] for selected classes of chemical compounds. Results of a survey performed in the city of Berlin. Published target and reference values by Schleibinger et al. (2002) are included. 90. P: 90<sup>th</sup> percentile, AM: arithmetic mean, GM: geometric mean.

The relationship between ambient air concentrations of selected substance classes and the ACR the following picture can be summarised as follows (Fig. 18): Depending on the substance class, between 50% and 70% of rooms with ACR  $<0.2 \text{ h}^{-1}$  (sample size of this subgroup varied from  $n = 41$  to  $n = 53$ ) showed concentrations which were above the median of the respective substance class. For ACR between  $0.2 \text{ h}^{-1}$  and  $0.5 \text{ h}^{-1}$  (sample size between  $n = 37$  and  $n = 52$ ) this was true for only 40 - 50% of the rooms. This proportion was further reduced to 10 - 20% when the ACR were equal to or greater than  $0.5 \text{ h}^{-1}$  ( $n = 15-27$ ). This trend was statistically significant for all substance classes studied here ( $\chi^2$  test for trend: aldehydes  $p < 0.01$ , aromatics  $p < 0.001$ , terpenes  $p < 0.001$ ), Even for siloxanes the trend was significant, although not very pronounced ( $p < 0.05$ ).

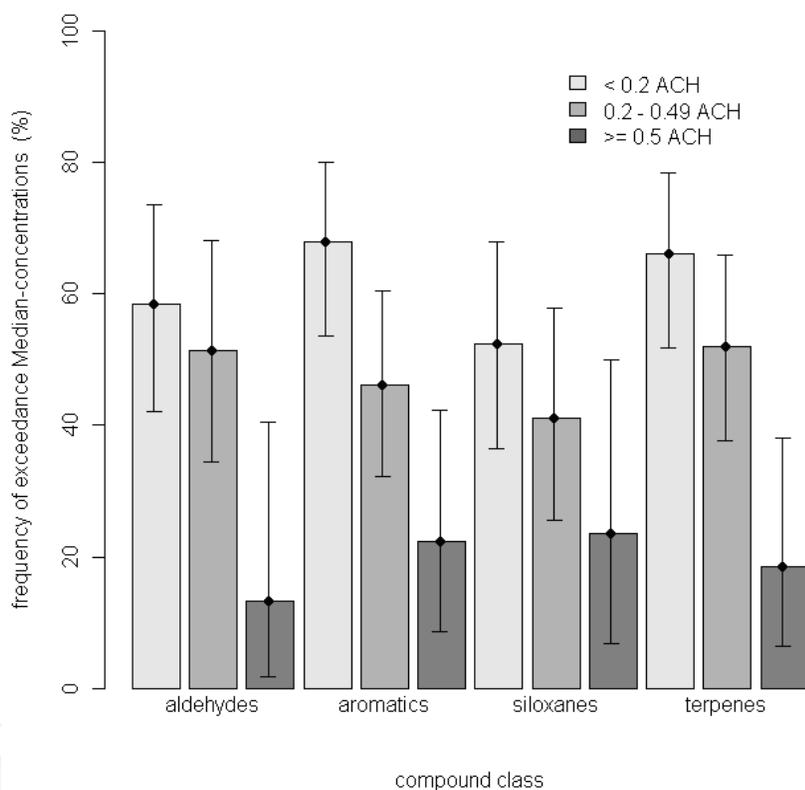


Fig. 18. Frequency of concentrations above median (50<sup>th</sup> percentile), given for selected compound classes and stratified by ACR. Active air sampling under worst case conditions, error bars: 95% confidence intervals.

Overall, it can be deduced that the probability of exceeding pollutant concentrations not only depends on the number of indoor sources and their emitting power, but also on the airtightness of rooms. However, no exceeding concentrations can occur without the presence of indoor sources. For this reason, the use of low-emission products should be given priority in order to avoid preventable future exposure to air pollutants. Whether a given indoor air concentration is above the usual burden and appropriate remedial measures are necessary,

can be clearly evaluated by the ACR. It would therefore be desirable to define air-change-related reference values for common and tolerable concentrations of volatile indoor air pollutants.

#### 4. Conclusion

Tracer gas measurements facilitate the determination of air change between indoor and outdoor environment under different ventilation conditions. In this paper methods are described which can be used to determine the air change rate with tracer gas measurements. Special emphasis was given to carbon dioxide, since this tracer gas is often used to assess indoor air quality and air tightness of rooms. We have shown ways to determine air change rates with carbon dioxide and compared the results with those obtained by other tracer gases.

The dependence of the natural air change from the prevailing weather conditions, such as the current wind and temperature conditions could be demonstrated by own investigations. Weather effects in conjunction with different window types were leading to considerable day-to-day variability, thereby limiting the reproducibility of measurements to a large extent. However, focusing on the relationship between air change and window ventilation we could show that ventilation with wide open windows or cross-ventilation are by far the cheapest ways to remove indoor pollutants as quickly as possible from indoor air. The effectiveness of window ventilation strongly depends on the temperature difference between room air and outside air, whereas the influence of wind seems to be less important.

Studies on the air quality in Berlin's housing stock revealed a very high proportion of rooms with low air change rates. About 80% of the rooms examined by us showed air change rates below  $0.5 \text{ h}^{-1}$ , half of which were even below  $0.2 \text{ h}^{-1}$ , when determined under so-called "worst case" conditions. Especially in rooms that had newly installed windows with insulated glass and peripheral sealing strip, very low air change rates were frequently determined. Of these, about 50% (median) had air change rates below  $0.16 \text{ h}^{-1}$ . This was in significant contrast to rooms in older buildings equipped with wooden framed double box windows or composite windows, which displayed mean air change rates of  $0.47 \text{ h}^{-1}$ .

Low air change rates often contribute to elevated VOC concentrations, in rooms with insulated windows, unless emissions are not limited by appropriate low-emission products. This relationship could be confirmed by our investigations. Improvement of ventilation i.e. increasing the air change rate is an efficient measure to additionally reduce the pollutant load in indoor spaces. Therefore, to achieve limitation of unhealthy indoor-borne pollutants a combination of both measures can strongly be recommended.

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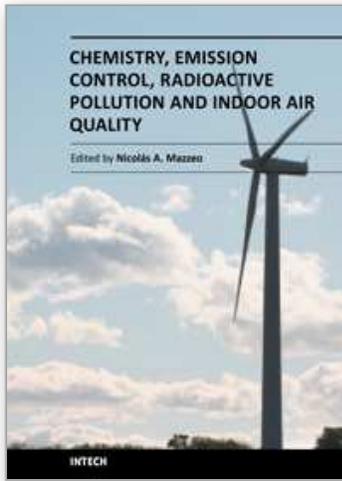
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## **Chemistry, Emission Control, Radioactive Pollution and Indoor Air Quality**

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The atmosphere may be our most precious resource. Accordingly, the balance between its use and protection is a high priority for our civilization. While many of us would consider air pollution to be an issue that the modern world has resolved to a greater extent, it still appears to have considerable influence on the global environment. In many countries with ambitious economic growth targets the acceptable levels of air pollution have been transgressed. Serious respiratory disease related problems have been identified with both indoor and outdoor pollution throughout the world. The 25 chapters of this book deal with several air pollution issues grouped into the following sections: a) air pollution chemistry; b) air pollutant emission control; c) radioactive pollution and d) indoor air quality.

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