

# Indoor deposition reducing the effect of toxic gas clouds in ordinary buildings

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## Abstract

A theoretical model is presented showing the effects of indoor deposition following the leakage of gas from outside a building. The model is used to study toxic gas clouds from accidents or attacks with chemical warfare agents. The influence of deposition on indoor pollutant concentration, total exposure, and maximum concentration is studied. The effect of deposition in ventilation channels, cracks and insulation is also treated, as well as effects of internal filtration. Available values of indoor deposition velocities for some toxic gases are reviewed. The model shows that reduced air exchange will increase the protection against gases, which have indoor deposition velocities larger than zero. Internal filtration provided by human respiration and increased total indoor surface also contribute to reducing exposure. Available observations and experiments, at low to moderate concentrations, for  $\text{NH}_3$ ,  $\text{Cl}_2$ ,  $\text{SO}_2$ , and a simulant for the nerve agent VX, show indoor deposition velocities in the range  $3 \times 10^{-5}$ – $3 \times 10^{-4} \text{ m s}^{-1}$ . However,  $\text{NH}_3$  and the simulant for VX, showed a saturation effect with decreasing deposition velocities with time.

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## 1. Introduction

In many models treating the indoor pollutant concentration following accidental releases of toxic gases, or after attacks with chemical warfare agents, it is assumed that the air exchange is the only process affecting the indoor concentration [1–3]. Despite giving lower maximum indoor than outdoor concentrations, such models will give the result that the total indoor exposure ( $\int_0^\infty c_i dt$ ) equals the total outdoor exposure ( $\int_0^\infty c_o dt$ ) if the air exchange is constant with time. However, Van Leeuwen [4] takes into account the influence of deposition in a model describing the indoor protection following releases of hazardous materials. Van Leeuwen found that indoor protection can be improved by decreasing the ventilation, by increasing the deposition, and by shortening the ventilation delay after the cloud has passed outdoor.

Measurements of continuously released reactive pollutants in cities also suggest that indoor sinks exist which reduce the concentration. Yocom [5] summarized

existing air quality data showing that the mean ratio of the indoor to outdoor concentration of  $\text{SO}_2$  and  $\text{NO}_2$  is 0.5, whereas the ratio for  $\text{O}_3$  is 0.2. For CO (a nonreactive gas) indoor to outdoor concentration ratio is normally 1.0. These ratios tend to be lower at higher pollutant concentrations. Measured dilution rates which decreased exponentially, in the absence of air exchange, revealed for  $\text{NO}_2$  30–50 min half life times and for  $\text{O}_3$  half lives of 2–12 min [6–9]. Laboratory studies of  $\text{NO}_2$  have shown obvious deposition on some typical indoor materials, but also reduction of the deposition velocity onto these materials following repeated exposure [10]. The processes reducing pollutant concentration are explained by, e.g. deposition to, or decomposition on, indoor surfaces [5, 11] and may proceed at rates comparable to or even greater than the leakage of pollutant into the building. Accordingly it is reasonable to expect indoor deposition after outdoor accidental releases of other reactive industrial chemicals, e.g.  $\text{Cl}_2$ ,  $\text{NH}_3$ . There may however also be differences in behaviour of these gases because of higher outdoor concentration levels. Observations of outdoor deposition of some gases including  $\text{SO}_2$  and  $\text{Cl}_2$  are summarized by McMahon and Denison [12].

Nonreactive toxic gases may also deposit on indoor surfaces, e.g. nerve agents, which are known to deposit outdoors and are easily adsorbed by lungs and skin. Based on the deposition process Zhuqing [13] have proposed an unvented poison-gas adsorption passage for shelters. Nonreactive gases however may exhibit greater desorption effects than the reactive gases.

In contrast to the circumstances accompanying accidental releases, there are many more models treating the indoor deposition of reactive gases in cities, e.g.  $\text{NO}_2$ ,  $\text{SO}_2$  and  $\text{O}_3$ , which are continuously released. Shair and Heitner [14] developed and tested a one chamber model which takes into account ventilation, deposition, and indoor emission. Nazaroff and Cass [11] developed this model further into a multichamber model using chemical kinetic mechanisms employed in outdoor air quality models. In some aerosol models filtration effects caused by e.g. wall cracks are also taken into account [15]. Theoretical studies of aerosols also show that deposition processes are important, especially in sealed rooms with low air exchange [16].

In this paper a theoretical model is presented showing the indoor deposition effects following an outside release of a toxic gas cloud. The effects of deposition in ventilation channels, cracks, and insulation are treated with a filter factor. Because some countries recommend to reduce the air exchange or seal a room, in order to improve the protection, the effect of deposition in these situations is studied specially. Available values of indoor deposition velocities for some toxic gases are also reviewed. Some thoughts on desorption are discussed although there is very little data available on that process.

## 2. Theoretical formulation

### 2.1. General aspects

The model can be applied to a whole building or to a single room. The treatment of the processes is an extension of the formulation of Shair and Heitner [14]. The

building or the room in question is represented by a chamber in which the air is considered to be well-mixed. This assumption is expected to be satisfied in single rooms especially if there are people generating turbulence. However between different rooms in a building there may be concentration gradients, and thus only the mean concentration, and the mean effect of deposition are studied. Air from outside may enter the building or the room through ventilation channels, through wall cracks, or through the insulation material of the walls. Total ventilation is represented by the total amount of air changes per time unit  $k$  ( $s^{-1}$ ). Each route of air exchange may be accompanied by filtration effects leading to an external filter factor  $f = (c_o - c_f)/c_o$ , where  $c_o$  is outdoor concentration and  $c_f$  is the concentration of the penetrating air. A total external filter factor  $f_o$  is then defined as a weighted arithmetical mean of the filter factors of each ventilation route.  $c_i$  represents the indoor concentration of well-mixed air. Indoor observations of  $NO_2$  and  $O_3$  without ventilation have shown an exponential decrease [6–9], such that the change of indoor concentration is proportional to the concentration itself. This is analogous to the formulations concerning the outdoor dry deposition of a gas, which is assumed to be proportional to its concentration [12]. Thus the net rate at which a pollutant is removed from room air by deposition onto floors, walls or other surfaces is assumed to be proportional to the indoor concentration, giving a sink term equal to  $-v_d A/V c_i$ , where  $v_d$  ( $m s^{-1}$ ) is the deposition velocity and  $A/V$  represents the total surface to volume ratio of the room. The value of  $v_d$  depends on the gas, on the properties of the surfaces, on the turbulence of the air in the room [11], and perhaps also on temperature. Internal filtration, e.g. through mask filters, adsorption in the lungs, or through special internal filtration equipment, will also give a sink term equal to  $-(V_{ei}/V) f_i c_i$ , where  $V_{ei}$  is the air flow ( $m^3 s^{-1}$ ) of the internal ventilation, and  $f_i$  is the internal filter factor. Internal sources, e.g. desorption, are represented by  $s$ . However, because of lack of data,  $s$  is normally assumed to be zero in presented solutions (Figs. 1–4). Other possible processes, such as deposition on aerosol particles are also excluded. The change in time  $t$  of the indoor concentration can be formulated as:

$$\frac{dc_i}{dt} = kc_o(1 - f_o) - kc_i - v_d(A/V)c_i - (V_{ei}/V)f_i c_i + s. \quad (1)$$

Eq. (1) has the general solution:

$$c_i = \exp\left(-\int_{t_0}^t (k + v_d A/V + (V_{ei}/V)f_i) d\varepsilon\right) \left(c_{i_0} + \int_{t_0}^t (kc_o(1 - f_o) + s) \times \exp\left(\int_{t_0}^t (k + v_d A/V + (V_{ei}/V)f_i) d\varepsilon\right) dt\right), \quad (2)$$

where  $c_{i_0}$  is indoor concentration at the time  $t_0$ . However in the following a solution is used when  $s$  and  $c_{i_0}$  are zero and all other parameters are constant in time. Further

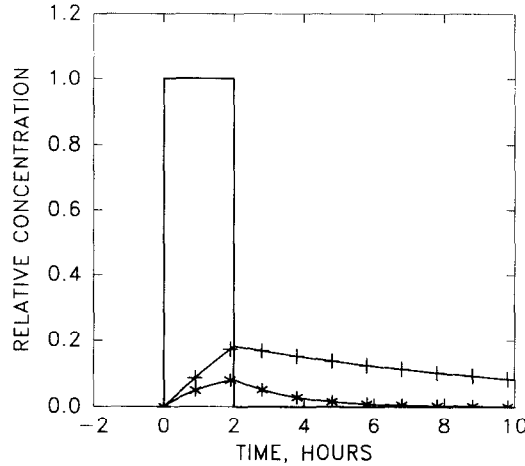


Fig. 1. The indoor concentration  $c_i$  in a sealed room according to Eq. (3). The outdoor concentration  $c_o = 1.0$  during the time period 0–2 h but otherwise zero.  $k$  is  $2.8 \times 10^{-5} \text{ s}^{-1}$  ( $0.1 \text{ h}^{-1}$ ),  $s$  and  $V_{ei}$  are assumed to be zero. (—) Outdoor concentration; (+) indoor concentration with  $f_o = 0$  and  $v_d = 0$  (no filtration/deposition); (\*) indoor concentration with  $f_o = 0.3$  and  $v_d A/V = 1.4 \times 10^{-4} \text{ s}^{-1}$  ( $0.5 \text{ h}^{-1}$ ).

$c_o$  is assumed to be constant for  $0 \leq t \leq t_B$ , and zero for  $t > t_B$ :

$$c_i = \frac{k(1-f_o)c_o}{k + v_d A/V + (V_{ei}/V)f_i} (1 - \exp(-(k + v_d A/V + (V_{ei}/V)f_i)t)),$$

$$0 \leq t \leq t_B, \quad (3a)$$

$$c_i = \frac{k(1-f_o)c_o}{k + v_d A/V + (V_{ei}/V)f_i} (1 - \exp(-(k + v_d A/V + (V_{ei}/V)f_i)t_B)) \cdot \exp(-(k + v_d A/V + (V_{ei}/V)f_i)(t - t_B)), \quad t > t_B \quad (3b)$$

Figs. 1 and 2 show  $c_i$  for a sealed room and an ordinary room with  $k$  equal to  $2.8 \times 10^{-5} \text{ s}^{-1}$  ( $0.1 \text{ h}^{-1}$ ), and  $2.8 \times 10^{-4} \text{ s}^{-1}$  ( $1 \text{ h}^{-1}$ ) respectively, for two combinations of  $f_o$  and  $v_d A/V$ . Thereby  $V_{ei}$  is also assumed to be zero.

The effect of only air exchange ( $v_d = 0, f_o = 0$ ) is to reduce the indoor concentration compared to the outdoor value, provided the time of the outdoor concentration is short compared to  $1/k$ . However the indoor exposure time is significantly prolonged especially if  $k$  is low. The effect of the filter term is to further reduce the concentration. The effect of the deposition term includes further reduction of the indoor concentration and shortening the exposure time.

## 2.2. Total indoor exposure

The total indoor exposure ( $\int_0^\infty c_i dt$ ) is obtained by integrating Eq. (1) from  $t = 0$  to  $t = \infty$ . By using  $c_{i0} = c_{i\infty} = 0$  the left side of the new equation will be zero. Assuming

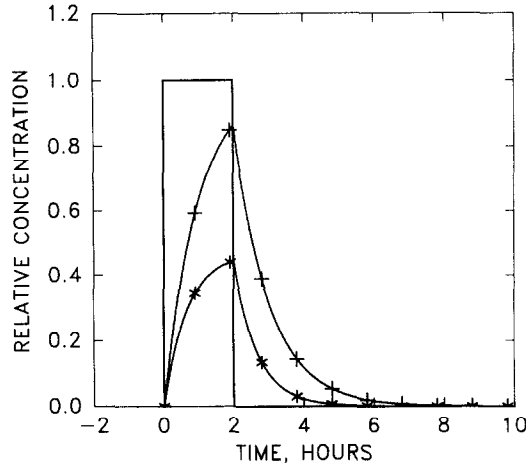


Fig. 2. The indoor concentration  $c_i$  in an ordinary room according to Eq. (3). The outdoor concentration  $c_o = 1.0$  during the time period 0–2 h but otherwise zero.  $k$  is  $2.8 \times 10^{-4} \text{ s}^{-1}$  ( $1 \text{ h}^{-1}$ ),  $s$  and  $V_{ei}$  are assumed to be zero. (—) Outdoor concentration; (+) indoor concentration with  $f_o = 0$  and  $v_d = 0$  (no filtration/deposition), (\*) indoor concentration with  $f_o = 0.3$  and  $v_d A/V = 1.4 \times 10^{-4} \text{ s}^{-1}$  ( $0.5 \text{ h}^{-1}$ ).

$s$  is zero and all other parameters are constant in time, the remaining equation is solved for  $\int_0^\infty c_i dt$ , and the ratio of indoor to outdoor total exposure will then be:

$$\frac{\int_0^\infty c_i dt}{\int_0^\infty c_o dt} = \frac{k(1 - f_o)}{k + v_d A/V + (V_{ei}/V)f_i'} \quad (4)$$

which is presented in Fig. 3 as a function of  $k$ . It can be seen from the figure that the total indoor exposure is reduced more by the deposition when the air exchange is small. From the figure it can also be concluded that internal filtration by inhalation may be important at low values of  $k$ .

### 2.3. Maximum indoor concentration

The reduced total indoor exposure, Fig. 3, is attributable to the lower value of  $c_i$  and the shorter indoor exposure time as illustrated in Figs. 1 and 2. In order to explicitly show the effect of  $v_d$ ,  $A/V$  and  $k$  on the maximum indoor concentration ( $c_{i\max}$ ),  $t$  is put equal to  $t_B$  in Eq. 3(a). The ratio of  $c_{i\max}$  to  $c_o$  is then presented in Fig. 4 for  $f_o$  equal to 0.3,  $t_B$  equal to 2 h, and  $V_{ei}$  equal to zero. The figure clearly shows that increasing  $v_d$ , increasing  $A/V$  and decreasing  $k$  will give lower values of  $c_{i\max}$ .

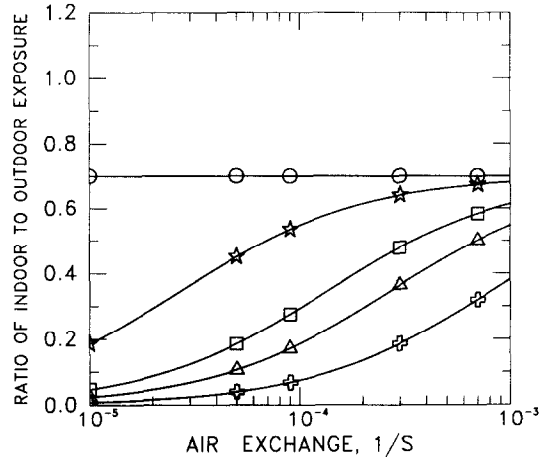


Fig. 3. Ratio of indoor to outdoor total exposure according to Eq. (4) for  $f_o = 0.3$  and  $s = V_{ei} = 0$ . Although  $V_{ei}$  is zero the effect of internal filtration can be estimated from the figure. E.g. the inhalation of four persons would give  $V_{ei} \approx 40$  litres per minute and  $(V_{ei}/V)f_i \approx 2.7 \times 10^{-5} \text{ s}^{-1}$  if  $f_i \approx 1.0$  and  $V = 25 \text{ m}^3$ . Thus the effect of internal filtration in this case is approximately comparable to the effect of  $v_d A/V = 2.8 \times 10^{-5} \text{ s}^{-1}$ .  $v_d A/V$ : (○) 0; (☆)  $2.8 \times 10^{-5} \text{ s}^{-1}$  ( $0.1 \text{ h}^{-1}$ ); (□)  $1.4 \times 10^{-4} \text{ s}^{-1}$  ( $0.5 \text{ h}^{-1}$ ); (△)  $2.8 \times 10^{-4} \text{ s}^{-1}$  ( $1.0 \text{ h}^{-1}$ ); (⊕)  $8.0 \times 10^{-4} \text{ s}^{-1}$  ( $3.0 \text{ h}^{-1}$ ).

#### 2.4. External filter factors of ordinary buildings and sealed rooms

The air is assumed to penetrate a building through three possible ways: ventilation channel (without special filter), cracks, and insulation material in the wall. It is assumed that deposition occurs during the penetration, but desorption is excluded. The air is considered to be well-mixed perpendicular to the mean flow direction. Then the pollutant mass transfer  $dm$  from the air stream to a surface ( $b dx$ ) during the time interval  $dt$  will be

$$dm = -c_f v_d b dx dt, \quad (5)$$

where  $c_f$  is the concentration of the penetrating air,  $b$  is the adsorbing surface per unit length and  $x$  is the distance from the outdoor side of the wall. Dividing by the penetrating air volume ( $V_e dt$ ) the change in concentration of the filtrated air  $dc_f$  can be written as:

$$dc_f = -\frac{c_f v_d b dx}{V_e}, \quad (6)$$

where  $V_e =$  ventilation ( $\text{m}^3 \text{ s}^{-1}$ ). If all parameters are constant the solution of Eq. (6) is

$$c_f = c_o e^{-(v_d b L / V_e)}, \quad (7)$$

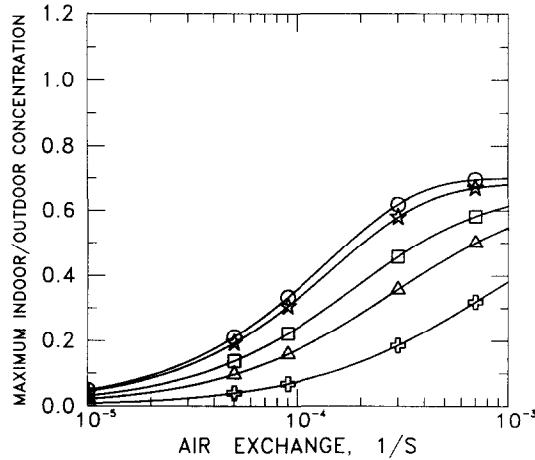


Fig. 4. The ratio of maximum indoor concentration to outdoor concentration according to Eq. 3(a) with  $t = t_B = 2$  h. The outdoor concentration  $c_o$  is assumed to be constant during the time period 0-2 h, but otherwise zero.  $f_o = 0.3$  and  $s = V_{ei} = 0$ .  $v_dA/V$ : (○) 0; (☆)  $2.8 \times 10^{-5} \text{ s}^{-1}$  (0.1  $\text{h}^{-1}$ ); (□)  $1.4 \times 10^{-4} \text{ s}^{-1}$  (0.5  $\text{h}^{-1}$ ); (△)  $2.8 \times 10^{-4} \text{ s}^{-1}$  (1.0  $\text{h}^{-1}$ ); (⊕)  $8.0 \times 10^{-4} \text{ s}^{-1}$  (3.0  $\text{h}^{-1}$ ).

Table 1

Calculated external filter factors ( $f$ ) for different air routes according to Eq. (8) for an assumed value of  $v_d$  equal to  $10^{-4} \text{ m s}^{-1}$

Ventilation flow ( $\text{m}^3 \text{ s}^{-1}$ )	Filter factor $f$							
	Ventilation channel length $L = 5$ m diameter = 0.1 m	Cracks length $L = 0.1$ m; periphery $b$			Insulation of glass fibres Volume ( $\text{m}^3$ )			
		10 m	30 m	100 m	0.001	0.01	0.1	1.0
$5.6 \times 10^{-3}$ (= $20 \text{ m}^3 \text{ h}^{-1}$ )	0.03	0.02	0.05	0.2	0.09	0.6	0.9999	1.0
$1 \times 10^{-3}$ (= $3.6 \text{ m}^3 \text{ h}^{-1}$ )	0.15	0.10	0.3	0.6	0.4	0.995	1.0	1.0

where  $c_o$  = outdoor concentration and  $L$  = the length of the ventilation channel or the thickness of the wall. The filtration factor ( $f = 1 - c_i/c_o$ ) can now be written as

$$f = 1 - e^{-(v_d b L / V e)}, \tag{8}$$

where  $(bL)$  is the total adsorbing surface. For a ventilation channel  $b$  is the periphery of the channel. For wall cracks  $b$  is the sum of the peripheries of all cracks, which is about twice the total length of all cracks. For insulation materials  $(bL)$  is the total surface of all fibres. It can be shown that for glass fibre insulation with diameter  $6 \mu\text{m}$  [23],  $(bL)$  is about  $7500 \text{ m}^2$  per  $\text{m}^3$  of insulation material. Using Eq. (8) some values of filter factors for different air ways are presented in Table 1 for an assumed value of  $v_d$  equal to  $10^{-4} \text{ m s}^{-1}$ , which is near values found for  $\text{Cl}_2$  and  $\text{SO}_2$  (see Table 2).

Table 2  
Data on indoor deposition velocities

Gas	$v_d = 1/(r_a + r_1 + r_s)$ ( $\text{m s}^{-1}$ )	Notes
NO <sub>2</sub>	$1.4 \times 10^{-4}$	Residential rooms, air quality data $c_o/c_i = 2$ , assumed $A/V = 2 \text{ m}^{-1}$ [5]
	$1 \times 10^{-4} - 2 \times 10^{-4}$	Decrease rate measurements in residential rooms, assumed $A/V = 2 \text{ m}^{-1}$ , $k$ measured with CO [6, 7]
	$0.6 \times 10^{-4}$	Summary from literature on air quality [11]
	$0.1 \times 10^{-4} - 8 \times 10^{-4}$	Decrease rate measurements in a box with a fan, various indoor materials [10]
SO <sub>2</sub>	$1.4 \times 10^{-4}$	Residential rooms, air quality data $c_o/c_i = 2$ , assumed $A/V = 2 \text{ m}^{-1}$ [5]
O <sub>3</sub>	$6 \times 10^{-4}$	Residential rooms, air quality data $c_o/c_i = 5$ , assumed $A/V = 2 \text{ m}^{-1}$ [5]
	$3.6 \times 10^{-4}$	Summary from literature on air quality [11]
	$3 \times 10^{-3} - 5 \times 10^{-4}$	Decrease rate measurements in residential rooms, assumed $A/V = 2 \text{ m}^{-1}$ , $k$ assumed $\approx 0$ due to low ventilation [8, 9]
HNO <sub>3</sub> , NO <sub>3</sub>	$7 \times 10^{-4}$	Assumed $v_d = v_{d,\text{max}}$ [11]
NO, CO	0	[11]
Cl <sub>2</sub>	$0.9 \times 10^{-4} - 1.0 \times 10^{-4}$	Decrease rate measurements in test rooms <sup>a</sup> , fan to mix the air, initial concentration 15 ppm (50 mg/m <sup>3</sup> ), $A/V = 2.1 \text{ m}^{-1}$ , $k$ measured with N <sub>2</sub> O [21]
NH <sub>3</sub>	$3 \times 10^{-5} - 5 \times 10^{-5}$	Decrease rate measurements in test rooms, fan to mix the air, initial concentration 75 ppm (50 mg/m <sup>3</sup> ), $A/V = 3.2 \text{ m}^{-1}$ . After three hours $v_d$ was $\approx 0$ , $k$ measured with N <sub>2</sub> O [21]
Trialkylphosphonoacetate	$2.6 \times 10^{-4} \pm 1.6 \times 10^{-4}$	Decrease rate measurements in test rooms <sup>a</sup> , with simulant of the nerve agent VX, fan to mix the air. Released amount planned to given initial concentration 50 $\mu\text{g}/\text{m}^3$ in case of $v_d = 0$ , $A/V = 2.2 - 3.5 \text{ m}^{-1}$ , $v_d \approx 2.6 \times 10^{-4}$ during one hour, then a tendency to desorption, $k$ measured with N <sub>2</sub> O [22]

<sup>a</sup> With painted ceiling and walls, and plastic carpet.

It is seen in Table 1 that insulation material of fibres may give a very good filter effect if  $v_d$  is  $10^{-4} \text{ m s}^{-1}$ , provided the penetrated volume is larger than  $0.01 \text{ m}^3$ . Cracks and ventilation channels may give some filtration especially for the low ventilation attained in sealed rooms. However no detailed calculations of the total filter factor  $f_o$  can be made because normally there is no information of the fraction of air passing different air ways. Only rough estimates are possible. For example in a sealed room with reduced ventilation the fraction of air passing through an insulation volume  $\geq 0.01 \text{ m}^3$  may roughly be estimated to be somewhere between 0 and 30%. If the remaining fraction is supposed to pass through a ventilation channel of 5 m length or through cracks with periphery 30 m the resulting total filter factor  $f_o$  will then be between 0.15 and 0.4 when  $v_d$  is  $10^{-4} \text{ m s}^{-1}$ .



### 3. Values of indoor deposition velocities

#### 3.1. Maximum deposition velocity

According to Zannetti [17] the deposition velocity can be formulated as  $v_d = 1/(r_a + r_1 + r_s)$ , where  $r_s$  is the surface resistance,  $r_a$  is the aerodynamic resistance depending on turbulent diffusion, and  $r_1$  is the resistance in the viscous sub-layer. The largest possible deposition velocity is achieved when  $r_s$  is zero:

$$v_{d,\max} = \frac{1}{r_a + r_1}. \quad (9)$$

In Eq. (9)  $r_a = \int_{\delta}^{z_1} (1/K_t) dz$ , and  $r_1 = \int_0^{\delta} (1/D) dz$ , where  $\delta$  is the thickness of the viscous sub layer,  $K_t$  is the turbulent coefficient of diffusion,  $z$  is distance from the surface,  $z_1$  is the height to the centre of the room, and  $D$  is the molecular diffusivity. By using theoretical and experimental data on heat diffusion in a room Nazaroff and Cass [11] estimated  $v_{d,\max}$  to be  $7 \times 10^{-4} \text{ m s}^{-1}$ . Using  $K_t = 0.32qz$ , where  $q$  is a representative turbulent velocity [18, 19], gives another possibility to estimate  $v_{d,\max}$ :

$$v_{d,\max} = \frac{1}{\frac{\ln(z_1/\delta)}{0.32q} + \frac{\delta}{D}}. \quad (10)$$

By using an equation for the turbulent kinetic energy [18], Karlsson et al. [19] have calculated typical values of  $q$  to be of order  $0.01 \text{ m s}^{-1}$  in a room with reduced ventilation, no temperature gradients, no heat sources, and no people present. Using the same equation for turbulent kinetic energy it can be shown that the human heat release (about 50 W per person) will give buoyancy generated turbulence and  $q > 0.01 \text{ m s}^{-1}$ . As  $D$  may be in the range  $5 \times 10^{-6} - 2 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ ,  $\delta$  is of order  $10^{-3} \text{ m}$  and  $z_1$  is about 1.5 m, Eq. (10) will then give  $v_{d,\max}$  equal to about  $4 \times 10^{-4} \text{ m s}^{-1}$  when no people or other heat sources are present, and  $> 4 \times 10^{-4} \text{ m s}^{-1}$  when people are present, which support the value,  $7 \times 10^{-4} \text{ m s}^{-1}$ , used by Nazaroff and Cass [11].

#### 3.2. Data on indoor deposition velocities

Measurements of air quality in cities have shown that the mean of  $c_o/c_i > 1$  for  $\text{SO}_2$ ,  $\text{NO}_2$  and  $\text{O}_3$  [5], in absence of indoor sources. This can be used for calculating  $v_d$  by assuming that the long time mean values of  $dc_i/dt \approx 0$  in Eq. (1), which gives

$$v_d = \frac{k}{A/V} \left( \frac{(1-f_o)c_o}{c_i} - 1 \right) - \frac{V_{ei}f_i}{A} + \frac{s}{A/Vc_i}. \quad (11)$$

In normal residential rooms typical values of  $A/V$  is  $2 \text{ m}^{-1}$ . In normal residential houses with closed windows and doors, the values of  $k$  often vary between 0.5 and 1.5 air changes per hour [20], but in energy-efficient houses or in sealed rooms  $k$  may be  $0.1-0.5 \text{ h}^{-1}$  [5]. At normal ventilation  $f_o$  is probably small (see 2.4) why  $f_o$  here is

assumed to be zero. Assuming a mean of one person breathing  $10 \text{ l min}^{-1}$  in each room, and the total room area  $A = 40 \text{ m}^2$  will result in  $V_{ei}/A = 4 \times 10^{-6} \text{ m s}^{-1}$ . Thus by using  $s$  and  $f_o$  equal to 0,  $f_i$  equal to 1,  $A/V$  equal to  $2 \text{ m}^{-1}$  and  $k$  equal to  $1 \text{ h}^{-1}$  ( $2.8 \times 10^{-4} \text{ s}^{-1}$ ), Eq. (11) will give an approximate value of  $v_d$ .

Measurements of decrease rates after release of an agent in a room with low or no ventilation, or with simultaneous measurement of  $k$  with an inert tracer gas, can also be used to calculate  $v_d$  from Eq. 1. It is likely that  $v_d$  varies for different materials, which also experiments with  $\text{NO}_2$  in test boxes have shown [10]. Thus measurements in a residential room will give a mean value of  $v_d$  for all surfaces in the room. Available data on indoor deposition velocities are summarized in Table 2.

Table 2 shows that decrease experiments with  $\text{Cl}_2$  at  $50 \text{ mg m}^{-3}$  gave similar value for  $v_d$  as calculations for  $\text{SO}_2$  based on air quality data at lower concentration levels. Thus available data points to similar values for  $\text{SO}_2$  and  $\text{Cl}_2$  equal to about  $10^{-4} \text{ m s}^{-1}$ . Decrease rate experiments of  $\text{NH}_3$  gave a lower value ( $\approx 4 \times 10^{-5} \text{ m s}^{-1}$ ) which after three hours showed a saturation effect with  $v_d \approx 0$ . Decrease rate experiments with trialkylphosphonoacetate, a simulant for the nerve agent VX, showed a fast reduction of the concentration in the air, leading to a large initial value of  $v_d$ , but then a tendency to desorption.

Air quality data on  $\text{SO}_2$  points to an increased deposition velocity for high polluted areas ( $1600 \text{ } \mu\text{g m}^{-3}$ ) compared to low concentration ( $80 \text{ } \mu\text{g m}^{-3}$ ), [5]. Compared to outdoor deposition velocities, the indoor values of  $\text{Cl}_2$ ,  $\text{SO}_2$ ,  $\text{NO}_2$  and  $\text{O}_3$  seem to, be one to two order of magnitudes lower [12].

## 4. Discussion

Shair and Heitner [14] tested their model with air quality measurements of  $\text{O}_3$  and found good agreement between theory and experiment. Thus it seems reasonable to expect that the present model is valid when applied to accidents or attacks with chemical warfare agents, provided that there exists data on indoor deposition velocities for the gases and surfaces considered. However the assumptions of the model are further discussed in the following, including the behaviour of the model.

### 4.1. Assumptions

Experiments with  $\text{NO}_2$ ,  $\text{NH}_3$  and trialkylphosphonoacetate have shown deposition followed by saturation effects, resulting in lower values of  $v_d$  and for trialkylphosphonoacetate also desorption [10, 21, 22]. Thus desorption, written as  $s$  in Eq. (1), should be included. The desorption effect is similar to a filter breakthrough and can be assumed to exist to some extent for most gases. Desorption is likely to depend on the amount of deposited agent, the temperature, and on the concentration of the agent in the air. Thus there is a requirement for an additional equation describing the change of the amount of agent on the surface. This new equation could be derived, e.g. from the evaporation theories discussed by Horst and Slinn [24] for outdoor conditions.

Alternatively desorption could be treated similar to filter breakthrough [25]. However available measurements and experimental data are made at low to moderate concentrations and need to be completed with experiments at high concentrations in order to give calibration data for a desorption model.

The assumption of only one chamber in Eq. (1) may give an underestimation of the indoor deposition effect, if the air passes other chambers where deposition can occur, before the air leaks into the room under consideration. Thus other rooms in a building may provide a filter effect for an interior room, and a multi-chamber model should then be used to calculate the full effect. The formulation of Eq. (1) and the assumption of only one chamber also implies that the outdoor concentration  $c_o$  is homogeneous.  $c_o$  however may vary, and the highest concentration is often found closest to the ground if there is no plume rise. Thus the best protection against an agent may be achieved at higher floors if they can be well sealed.

The air exchange  $k$  in residential rooms is in general much lower than the estimated values of  $v_{d,max} A/V$  (which represent turbulent and molecular diffusion) and thus the assumption of well-mixed condition in a single room is obtained as regarded in Eq. (1). In general  $v_{d,max}$  is also much larger than the observed values of  $v_d$  and thus deposition do not contradict the assumption of well-mixed conditions in a room. In case of only molecular diffusion perpendicular to the ventilation flow the maximum deposition velocity ( $v_{d,max}$ ) is  $D/z_1$  (see 3.1), where  $z_1$  is the distance from the centre of the flow to the adsorbing surface, and  $D \geq 5 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ . For a ventilation channel  $z_1$  may be 0.05 m giving  $v_{d,max} \geq 10^{-4} \text{ m s}^{-1}$ . However the flow in ventilation channels probably is turbulent giving  $v_{d,max}$  even larger. For cracks  $z_1 \approx 10^{-3} \text{ m}$  giving  $v_{d,max} \geq 5 \times 10^{-3} \text{ m s}^{-1}$ , and for insulation of glass fibres  $z_1 \approx 6 \times 10^{-5} \text{ m}^{-1}$  giving  $v_{d,max} \geq 8 \times 10^{-2} \text{ m s}^{-1}$ . Thus in general  $v_d \ll v_{d,max}$  and deposition do not contradict the assumption of well-mixed conditions perpendicular to the ventilation flow in the sub-model for external filter factor.

Deposition onto natural aerosol particles (excluded in Eq. (1)) may reduce the gas phase concentration, but although adsorbed it may possibly keep its toxic effect. Thus it seems more appropriate to exclude this aerosol related effect until more information becomes available on the interaction between gases and aerosols. However this effect may give an overestimation of  $v_d$  in measurements and experiments (see Table 2), since the effect as observed may partly depend upon deposition onto aerosol particles. The urban aerosol surface distribution frequency often peaks at a diameter of about  $2 \times 10^{-7} \text{ m}$  with a number  $< 10^{11} \text{ m}^{-3}$ , [26], giving an aerosol surface to room volume ratio of  $< 2 \times 10^{-2} \text{ m}^{-1}$ , which is  $\leq 1\%$  of normal values of  $A/V$  in residential rooms. Thus the error in measured  $v_d$  from this effect is assumed to be  $\leq 1\%$ . For hygroscopic agents like  $\text{NH}_3$ , HF and HCl, part of the gas may form an aerosol when mixing with humid air [27, 28]. Because aerosol deposition is significant for aerosol size larger than about  $1 \mu\text{m}$  [29], the size distribution of such gas-aerosol systems should need to be clarified.

Only rough estimates of the external filter factor  $f_o$  is possible because the fraction of air passing different air ways are not known. However Nylund [30] and Bergström [31] have presented methods which allow calculating the fraction of air penetrating through ventilation channels, walls and roof structures. If these methods could be

improved to estimate the penetrated insulation volume and total crack periphery more detailed calculations could be made.

#### 4.2. Protection capacity of deposition

At normal ventilation the protective effect of deposition is in general moderate. However, at low values of air exchange, deposition can balance leakage into a room thereby resulting in low concentrations and short exposure time. Thus closing ventilation, doors, windows, and sealing cracks, will reduce the effect of chemicals for which the cumulative dose  $\int c \, dt$  is the determinant. An even larger reduction of the effect is achieved if  $\int c^n \, dt$  determines the injuries and  $n > 1$ , which seems to be the case for e.g.  $\text{NH}_3$  and the nerve agent soman [32–34]. A larger reduction of the effects is also probable if the concentration has to exceed a threshold value in order to give injuries. A low air exchange also reduces the risk of saturating the adsorbing surface. However even if deposition should be followed by a later total desorption, the maximum concentration should be lower, which result in reduced effect if  $n > 1$ , or if there is a threshold effect. An air exchange ( $k$ ) differing from the constant value which was assumed in Eq. (4) should result in other ratios of indoor to outdoor total exposure. For example, if it is possible to decide when the gas cloud has passed outdoor, and then ventilate or leave the room, the ratios would be lower. Then the effect of deposition may also be limited. Practical methods for sealing rooms are given by Yeshua and Adler [35].

At very low values of air exchange one must be aware of expired  $\text{CO}_2$ , because concentration above 8–10% may be lethal. Thus values above 3% is not recommended. E.g. in a room of  $40 \text{ m}^3$ , without any ventilation ( $k = 0$ ) with 4 persons at light work breathing  $10\text{--}20 \text{ l min}^{-1}$ , 3% will be reached after 6–12 h.

Large values of the surface to volume ( $A/V$ ) ratio of a room will increase the effect of deposition. Thus curtains and other similar surfaces giving large  $A$  will provide additional protection. Choosing a very small room in order to increase  $A/V$  is however not advisable because of the  $\text{CO}_2$  risk. Respiration of a toxic gas is of course harmful, but gas that is adsorbed by humans is removed from the room air and cannot give repeated injuries. Thus internal filtration caused by human respiration can be important at low ventilation and weak deposition.

#### 4.3. Deposition velocities

Different surfaces will in general give different deposition velocities for a certain agent, and high air humidity may increase  $v_d$  for some agents, e.g. nerve agents [36]. The simulant trialkylphosphonoacetate is chemically similar to the nerve agent VX [37], and it is likely that their deposition velocities are similar. Observations of  $\text{NO}_2$  and  $\text{O}_3$  have shown low deposition on glass surfaces [8, 10]. In contrast nerve agents have been observed to decompose on glass [36]. However glass fibre insulation is often impregnated with resins, which may change the fibre surface properties. Thus there is a need for experiments to determine  $v_d$  at different humidity for the most common surfaces, including insulation.

Outdoor observed deposition velocities ( $10^{-3}$ – $10^{-2}$  m s<sup>-1</sup>) [12], are much larger than observed indoors (see Table 2). One could expect that lower indoor turbulence should explain this difference, but it does not because estimated values of  $v_{d,max}$ , which represent indoor turbulent and molecular transports, are larger than the observed indoor deposition velocities (Table 2). Probably the cause is different surface types with larger indoor surface resistance than outdoors, but also that outdoor values of  $v_d$  often include a relative vegetation surface compared to a completely flat one.

## 5. Conclusions

An extension of the air quality indoor model by Shair and Heitner [14] is used to study toxic gas clouds from accidents or attacks with chemical warfare agents. The input data is outdoor concentration, air exchange, external and internal filter factors, indoor deposition velocity and internal filtration flow. Internal sources are assumed to be zero. A sub model to estimate the external filter factor is also developed.

The model shows that reduced air exchange will increase the protection against gases, which have indoor deposition velocities larger than zero. E.g.  $v_d = 10^{-4}$  m s<sup>-1</sup> and  $k = 0.1$  h<sup>-1</sup> (sealed room) will result in a ratio of indoor to outdoor total exposure (Eq. (4)) which is about 0.1, if there is no external or internal filtration, whereas normal ventilation ( $k = 0.5$  h<sup>-1</sup>) will give the ratio equal to about 0.4. Increased total indoor surface and internal filtration provided by human respiration also contribute to an increased protection.

Available observations and experiments, at low to moderate concentrations, for NH<sub>3</sub>, Cl<sub>2</sub>, SO<sub>2</sub>, and a simulant for the nerve agent VX, show indoor deposition velocities in the range  $3 \times 10^{-5}$ – $3 \times 10^{-4}$  m s<sup>-1</sup>. However NH<sub>3</sub> and the simulant of VX show a saturation effect and the latter also a tendency to desorption. Thus there is a need to include desorption in the model equations.

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