Journal of Hazardous Materials 192 (2011) 419–422

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Short communication

Analytical solutions for exposures and toxic loads in well-mixed shelters in support of shelter-in-place assessments

S.T. Parker*, C.J. Coffey[1]

Dstl, Porton Down, Salisbury, Wiltshire SP4 0JQ, United Kingdom

ARTICLE INFO

Article history:
Received 21 December 2010
Received in revised form 27 April 2011
Accepted 30 April 2011
Available online 11 May 2011

Keywords:
Indoor dispersion
Exposure
Toxic load
Shelter-in-place

ABSTRACT

Understanding the exposure and toxic load for the interior of buildings during and following the passage of an external airborne hazard can be a critical piece of information in deciding the benefit from adopting a shelter-in-place strategy. Whilst numerical methods allow the calculation of such parameters for the general case, analytical solutions allow more rapid assessments to be made and highlight the key parameters more clearly. Analytical expressions are derived for the exposure due to the acute inhalation of toxic chemicals and the associated toxic load as a function of time, external hazard duration and building air change rate assuming a top-hat outdoor concentration profile and no indoor loss mechanism. It is shown that the internal exposure tends to the external exposure at long times for any external concentration profile. Expressions are derived for toxic loads with exponents $n = m/2$ where $2 \leq m \leq 7$ is an integer to cover the range of typical values ($1 \leq n \leq 3.5$). At long times the ratio of internal to external toxic load for a top-hat outdoor concentration profile is shown to be a function of the product of the air change rate and the duration of the external hazard.

1. Introduction

In the event of a release of hazardous airborne material in the outdoor environment, sheltering within buildings or other enclosed spaces (hereafter shelters) may reduce the likelihood of harmful effects. The benefit that can be achieved relies on the fact that the concentration within the shelter will take some time to respond to a sudden increase in external concentration. The peak concentration within the shelter is therefore unlikely to reach the same value as the outdoor concentration for a transient outdoor hazard. However, for most hazardous materials the likelihood of a negative impact on health will depend on a cumulative measure of the concentration experienced as opposed to a peak value. A commonly used measure is the exposure. If the concentration within the shelter is $C(t)$, where t is the time, then the exposure can be defined as the integral of the concentration with respect to time

$$E(t) = \int_0^t C(u)\mathrm{d}u, \tag{1}$$

where u is a dummy variable of integration. Some studies [1,2] have suggested that for some materials it may be more appropriate to consider the toxic load, defined as:

$$T(t) = \int_0^t C^n(u)\mathrm{d}u, \tag{2}$$

where $n > 0$. The exponent n is dependent on the toxicological properties of the hazardous material and typically $1 \leq n \leq 3.5$, although it can take values less than 1. Exposure (1) is equivalent to the toxic load (2) when $n = 1$. It should be noted that toxicological data is typically derived using experiments designed to achieve fixed concentrations for the duration of a given experiment. However, the expressions above are applied widely [2,3].

Several other studies have considered the likely benefit of such sheltering-in-place (SIP) [2–9]. Chan et al. [2] showed that the timing of subsequent evacuation and the toxic-load exponent can be important factors in determining the benefit of SIP strategy.

This paper presents analytical expressions for exposure and toxic load in well-mixed single zone shelters for simple exterior concentration profiles. These are generally appropriate for shelters with an air change rate that is large compared to the release time of the toxic chemical and allow rapid assessment of the likely benefits of SIP for different toxic-load exponents, air change rates and evacuation timings. Whilst these calculations can be carried out numerically, analytical solutions allow more rapid assessments and highlight the importance of key parameters more clearly. In partic-

* Corresponding author. Tel.: +44 1980 613266; fax.: +44 1980 613987.
E-mail addresses: stparker@dstl.gov.uk (S.T. Parker), christopher.coffey@amec.com (C.J. Coffey).
[1] Current address: AMEC, Booths Park, Chelford Road, Knutsford, Cheshire, WA16 8QZ, United Kingdom.

0304-3894/$ – see front matter. Crown Copyright © 2011 Published by Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2011.04.107

ular, analytical expressions are presented for the exposure and toxic load for toxic load exponents $n = m/2$ where m is an integer between 2 and 7. Expressions are provided for these measures for any time from the period at the end of the external hazard and their limiting values for long times. These provide useful methods for developing rule-of-thumb estimates of the ratio between internal and external toxic loads. An important result is reinforced — that the exposure within a shelter tends to the external exposure with increasing time for any outdoor concentration profile in the absence of loss mechanisms. This is true irrespective of the air change rate or duration of the exterior hazard, although the air change rate influences the rate at which this limit is approached.

2. Theory

Consider a well-mixed single-zone shelter connected to the external environment. Assuming a constant internal volume (V) and a constant air flow rate (Q) into and out of the shelter, the air change rate can be defined as $\lambda = Q/V$.

2.1. Concentration

For a given external concentration profile ($C_e(t)$), the rate of change of the internal concentration ($C_i(t)$) is given by

$$\frac{\mathrm{d}C_i}{\mathrm{d}t} = \lambda(C_e - C_i). \tag{3}$$

In integral form, the internal concentration is given by the convolution integral

$$C_i(t) = \int_0^t \lambda e^{\lambda(u-t)} C_e(u)\,\mathrm{d}u. \tag{4}$$

The case where the external concentration has a top-hat (or rectangular) profile is an obvious simplification. However, for constant release rates, when release times are long compared to the time taken for the contaminant to reach the shelter, it can be a reasonable assumption. For example, Chan et al. [2] show that a constant release of 1 h duration, at a distance of 1 km in atmospheric stability condition D, presents a concentration time series that is close to a top-hat profile. Convective atmospheric conditions may introduce short term fluctuations in the external concentration time-series. However, in practice short duration intermittency in the external concentration will also be smoothed out by the volume of the building. In the case of a top-hat profile

$$C_e(t) = \begin{cases} C_0 & \text{if } t < t_p, \\ 0 & \text{if } t \geq t_p, \end{cases} \tag{5}$$

where $C_0 \geq 0$ is the peak external concentration and $t_p \geq 0$ is the duration of the pulse. The internal concentration is then given by

$$C_i(t) = \begin{cases} C_0\left(1 - e^{-\lambda t}\right) & \text{if } t < t_p, \\ C_0(1 - e^{-\lambda t_p})e^{-\lambda(t-t_p)} & \text{if } t \geq t_p. \end{cases} \tag{6}$$

Fig. 1 shows an example time series of both internal and external concentration.

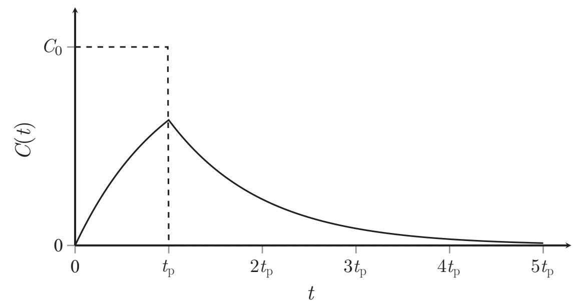

Fig. 1. A typical time series of internal concentration (—) for an external concentration with a top-hat profile with peak concentration C_0 and duration t_p (- -).

2.2. Exposure

For a general time-varying external concentration, the exposure for a static receptor in the external environment is

$$E_e(t) = \int_0^t C_e(u)\mathrm{d}u. \tag{7}$$

Similarly, for a static receptor in the internal environment, the exposure is

$$E_i(t) = \int_0^t C_i(u)\,\mathrm{d}u = \int_0^t \int_0^u \lambda e^{\lambda(v-u)} C_e(v)\mathrm{d}v\,\mathrm{d}u, \tag{8}$$

where v is a second dummy variable of integration for the inner integral. Switching the order of integration, one may show that

$$E_i(t) = \int_0^t C_e(u)\,\mathrm{d}u - \int_0^t e^{\lambda(u-t)} C_e(u)\,\mathrm{d}u, \tag{9}$$

or

$$E_i(t) = E_e(t) - \frac{C_i(t)}{\lambda}. \tag{10}$$

Thus, for large air-exchange rates, $E_i(t) \approx E_e(t)$.

Pertinent to questions of shelter-in-place is the comparison of the external and internal exposure. As such, the ratio of exposure inside the shelter to that outside ($\rho_E = E_i/E_e$) is examined. For large times, a finite duration external concentration and a greater than zero air-change rate,

$$\lim_{t\to\infty} \rho_E(t) = \lim_{t\to\infty}\left(1 - \frac{C_i(t)}{\lambda E_e(t)}\right) = 1, \tag{11}$$

since for a finite duration external concentration, $C_e \to 0$ as $t \to \infty$ and from (3), $\mathrm{d}C_i/\mathrm{d}t \to -\lambda C_i$ as $t \to \infty$ and hence $C_i \to 0$. Eq. (11) is a key result, showing that remaining within a shelter indefinitely will result in the same exposure as outdoors in the absence of other contaminant removal mechanisms.

In the case of a top-hat profile (5) specific solutions can be written as follows:

$$E_e(t) = \begin{cases} C_0 t & \text{if } t < t_p, \\ C_0 t_p & \text{if } t \geq t_p; \end{cases} \tag{12}$$

$$E_i(t) = \begin{cases} C_0\left(t - \frac{1}{\lambda}\left(1 - e^{-\lambda t}\right)\right) & \text{if } t < t_p, \\ C_0\left(t_p - \frac{1}{\lambda}\left(e^{-\lambda(t-t_p)} - e^{-\lambda t}\right)\right) & \text{if } t \geq t_p; \end{cases} \tag{13}$$

and

$$\rho_E(t) = \begin{cases} 1 - \frac{1}{\lambda t}\left(1 - e^{-\lambda t}\right) & \text{if } t < t_p, \\ 1 - \frac{1}{\lambda t_p}\left(e^{-\lambda(t-t_p)} - e^{-\lambda t}\right) & \text{if } t \geq t_p. \end{cases} \tag{14}$$

Fig. 2 shows a typical exposure time series and Fig. 3 shows the ratio of exposures for the same case.

For such a top-hat concentration profile, evacuating the shelter at t_p would result in no further exposure. Therefore, the ratio of exposures at t_p

$$\rho_E(t_p) = 1 - \frac{1}{\lambda t_p}(1 - e^{-\lambda t_p}) \tag{15}$$

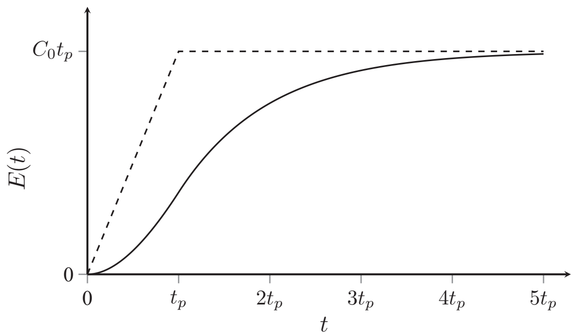

Fig. 2. A typical time series of internal exposure (—) and external exposure (- -) for an external concentration with a top-hat profile, a peak concentration C_0, and a duration t_p.

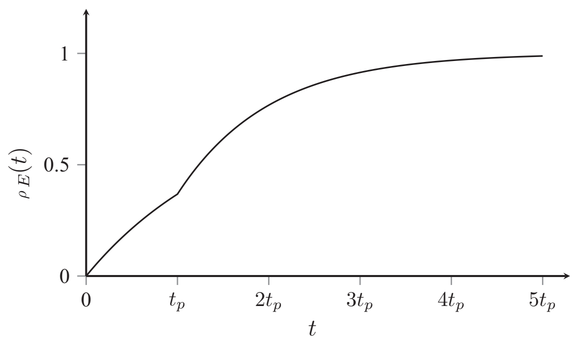

Fig. 3. Ratio of internal to external exposure for an exterior concentration with a top-hat profile, a peak concentration C_0, and a duration t_p.

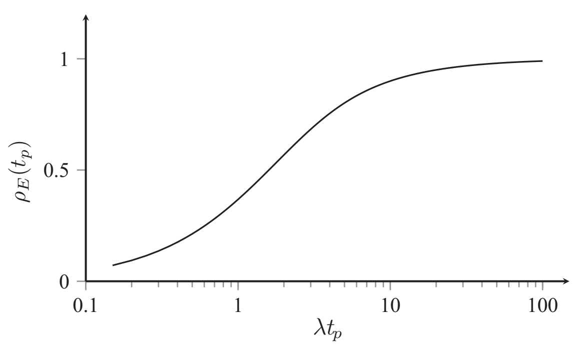

Fig. 4. The minimum exposure ratio $\rho_E(t_p)$ based on shelter evacuation immediately after t_p, for an external concentration with a top-hat profile, a peak concentration C_0, and a duration t_p.

describes the minimum exposure possible for shelter occupants relative to the outdoor exposure. Fig. 4 shows how the minimum exposure ratio varies with λt_p. For large air exchange rates (λ) or large pulse durations (t_p), the exposure of a static receptor inside the shelter is approximately equal to that of one outside.

2.3. Toxic load

Whilst exposure can be a useful measure Ten Berge et al. [1] argued that for many materials the concept of a toxic load (2) may be more appropriate. For a general time-varying external concentration the external toxic load is

$$T_e(t) = \int_0^t C_e(u)^n \,\mathrm{d}u, \tag{16}$$

and the internal toxic load is

$$T_i(t) = \int_0^t C_i(u)^n \,\mathrm{d}u = \int_0^t \left(\int_0^u \lambda e^{\lambda(v-u)} C_e(v)\,\mathrm{d}v \right)^n \mathrm{d}u. \tag{17}$$

For certain values of n, analytical expressions can be derived for the toxic load inside the shelter for a top-hat concentration profile. As an example, the case when $n = 2$ is considered.

$$T_e(t) = \begin{cases} C_0^2 t & \text{if } t < t_p, \\ C_0^2 t_p & \text{if } t \geq t_p; \end{cases} \tag{18}$$

$$T_i(t) = \begin{cases} C_0^2 \left(t - \dfrac{1}{2\lambda}\left(3 - e^{-\lambda t}\right)\left(1 - e^{-\lambda t}\right) \right) & \text{if } t < t_p, \\ C_0^2 \left(t_p - \dfrac{1}{\lambda}\left((1 - e^{-\lambda t_p}) + \dfrac{1}{2}(e^{-\lambda(t-t_p)} - e^{-\lambda t})^2 \right) \right) & \text{if } t \geq t_p. \end{cases} \tag{19}$$

The ratio of toxic load indoors to outdoors (ρ_T) is given by

$$\rho_T(t) = \begin{cases} 1 - \dfrac{1}{2\lambda t}\left(3 - e^{-\lambda t}\right)\left(1 - e^{-\lambda t}\right) & \text{if } t < t_p, \\ 1 - \dfrac{1}{\lambda t_p}\left((1 - e^{-\lambda t_p}) + \dfrac{1}{2}(e^{-\lambda(t-t_p)} - e^{-\lambda t})^2 \right) & \text{if } t \geq t_p. \end{cases} \tag{20}$$

Unlike the ratio of exposures, as t increases the ratio of toxic loads is generally less than one. The ratio for large t is given by

$$\lim_{t \to \infty} \rho_T(t) = 1 - \frac{1}{\lambda t_p}(1 - e^{-\lambda t_p}). \tag{21}$$

Similarly, analytical results can be derived for the cases when $n = 3/2$, 5/2, 3 and 7/2. Table 1 presents these results together with the cases when $n = 1$ (exposure) and 2. In the case of $n = 1$, we obtain the well-known result that the indoor exposure approaches that outdoors. The dependence of the toxic load ratio for large times on the product of λ and t_p is shown in Fig. 5.

These expressions can be compared with previously published values of dose reduction factors for sheltering indoors [9] for integer values of n which additionally considered absorption to building

Table 1

Analytical expressions for toxic load (left-hand column) within a shelter for $t \geq t_p$ and toxic load ratio (right-hand column) at large times for $n = 1$, 3/2, 2, 5/2, 3 and 7/2, where $a = \left(e^{-\lambda(t-t_p)} - e^{-\lambda t}\right)$ and $b = (1 - e^{-\lambda t_p})$.

n	$T_i(t)$	$\lim_{t\to\infty} \rho_T(t)$
1	$C_0\left(t_p - \frac{a}{\lambda}\right)$	1
3/2	$C_0^{3/2}\left(t_p - \frac{1}{\lambda}\left(2b^{1/2} - 2\ln(1 + b^{1/2}) + \frac{2}{3}a^{3/2}\right)\right)$	$1 - \frac{1}{\lambda t_p}\left(2b^{1/2} - 2\ln(1 + b^{1/2})\right)$
2	$C_0^2\left(t_p - \frac{1}{\lambda}\left(b + \frac{1}{2}a^2\right)\right)$	$1 - \frac{b}{\lambda t_p}$
5/2	$C_0^{5/2}\left(t_p - \frac{1}{\lambda}\left(2b^{1/2} + \frac{2}{3}b^{3/2} - 2\ln(1 + b^{1/2}) + \frac{2}{5}a^{5/2}\right)\right)$	$1 - \frac{1}{\lambda t_p}\left(2b^{1/2} + \frac{2}{3}b^{3/2} - 2\ln(1 + b^{1/2})\right)$
3	$C_0^3\left(t_p - \frac{1}{\lambda}\left(\frac{1}{2}b(2 + b) + \frac{1}{3}a^3\right)\right)$	$1 - \frac{1}{\lambda t_p}\left(\frac{1}{2}b(2 + b)\right)$
7/2	$C_0^{7/2}\left(t_p - \frac{1}{\lambda}\left(2b^{1/2} + \frac{2}{3}b^{3/2} + \frac{2}{5}b^{5/2} - 2\ln(1 + b^{1/2}) + \frac{2}{7}a^{7/2}\right)\right)$	$1 - \frac{1}{\lambda t_p}\left(2b^{1/2} + \frac{2}{3}b^{3/2} + \frac{2}{5}b^{5/2} - 2\ln(1 + b^{1/2})\right)$

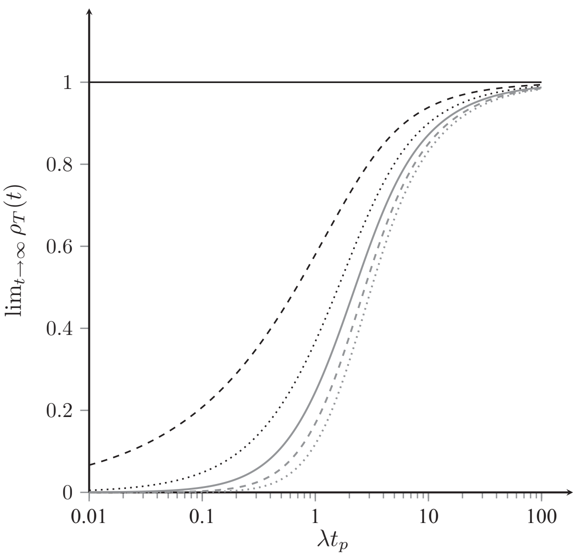

Fig. 5. The ratio of the interior to exterior toxic load as a function of the product λt_p as $t \to \infty$ for $n = 1$ (—), $n = 3/2$ (- -), $n = 2$ (. . .), $n = 5/2$ (▬▬), $n = 3$ (▬ ▬) and $n = 7/2$ (∗ ∗ ∗).

interior surfaces. When the absorption rate is zero and n is an integer, the formulae in Table 1 can be rearranged as expressions for dose reduction factors and become identical to those presented in Ref. [9].

3. Discussion

The expressions derived provide a rapid assessment of the ratio of exposure and toxic load for those within shelters compared to those outside and how this limit depends on the air change rate and duration of the external concentration profile. However, the above analysis considers an idealised shelter. For real shelters a number of features are likely to influence the internal concentration time series including surface interactions, non well-mixed shelters and modified ventilation rates. These are addressed in turn below.

Many chemical vapours will interact with interior shelter surfaces due to adsorption, absorption and desorption [10,11] and this will act to reduce the internal concentration in the short term and may increase the concentration later. Montoya et al. [8] provide equations for the evolution of concentration within a single zone for three difference sorption models. For fully reversible sorption as modelled by Montoya et al. [8] and where the toxic exponent is greater than one this will act to reduce the ratio of internal to external toxic load [5]. van Leeuwen [9] shows that in the case of irreversible absorption this ratio will also be reduced when $n = 1$. It should be noted that most sorption is in general non-negligible for most gaseous contaminants of concern.

The air within real shelters may not be well-mixed and in that case concentration time series experienced in different parts of the shelter may differ. However, when considering shelter-in-place options it may well be the case that detailed descriptions of shelter interiors and their ventilation properties are not known. In such cases, treating a shelter as a single well-mixed zone allows first-order estimates of the possible benefit to be made.

The expressions above all assume that the shelter occupants are in the shelter from the beginning of the release. Delays in entering the shelter would of course affect the exposure or toxic load. For the ratios of exposure and toxic load between inside and the exterior, the point of reference is continuous exposure outside close to the shelter for the same duration. In a real scenario on a small scale, it may be more likely that the immediate area would be rapidly evacuated reducing the exterior exposure and toxic load.

Some advice for sheltering in place recommends taking steps to reduce the air exchange between the interior and exterior environment. This would act to reduce the value of λ. If this was done before the contaminant reached the shelter it would reduce the toxic load ratios. However, if this was done during or after the passage of the contaminant past the shelter, the effect could be to increase these ratios.

This study assumes that the definition of toxic load is valid for time-varying concentrations. Hilderman et al. [12] explore this issue in more detail. A further assumption is that the values of the toxic load exponent n are known for the compound of interest and that they are derived from a robust set of studies. However, the experimental data from which n is derived are not available for every toxic compound and a given chemical may have a number of distinct toxic load exponents relating to different health endpoints [13]. In addition, the use of the toxic load to extrapolate to times much longer than those measured experimentally will introduce additional uncertainty. Care should therefore be taken when using this approach to remain within the limitations of the available data. In some cases additional experimental data may be required.

4. Conclusions

This brief study has considered the predicted time varying exposure and toxic load for building occupants during the passage of hazardous airborne material external to the building using a simplified model. It has been shown that the ratio of the in-building exposure to the external exposure varies as a function of the time, duration of the external hazard and the building air change rate. Importantly, it has been shown that for long times the same ratio tends to one for any external concentration profile in the absence of removal mechanisms.

A number of analytical expressions have been derived for the case of a single well-mixed zone and a top-hat external concentration profile. These expressions can be used to rapidly calculate the toxic load at any time for given toxic load exponents (where $n = m/2$ and m is an integer) for different external hazard durations, air change rates and times.

References

[1] W.F. Ten Berge, A. Zwart, L.M. Appelman, Concentration–time mortality response relationship of irritant and systemically acting vapours and gases, J. Hazard. Mater. 13 (1986) 301–309.
[2] W.R. Chan, W.W. Nazaroff, P.N. Price, A.J. Gadgil, Effectiveness of urban shelter-in-place I: idealized conditions, Atmos. Environ. 41 (2007) 4962–4976.
[3] H. Kaplan, A model for the toxic dose under time-varying concentration, J. Hazard. Mater. 167 (2009) 351–356.
[4] J.H. Sorensen, B.L. Shumpert, B.M. Vogt, Planning for protective action decision making: evacuate or shelter-in-place, J. Hazard. Mater. A109 (2004) 1–11.
[5] W.R. Chan, W.W. Nazaroff, P.N. Price, A.J. Gadgil, Effectiveness of urban shelter-in-place II: residential districts, Atmos. Environ. 41 (2007) 7082–7095.
[6] W.R. Chan, W.W. Nazaroff, P.N. Price, A.J. Gadgil, Effectiveness of urban shelter-in-place III: commercial districts, Build. Simul. 1 (2008) 144–157.
[7] M.S. Mannan, D.L. Kilpatrick, The pros and cons of shelter-in-place, Process Saf. Prog. 19 (2000) 210–218.
[8] M.I. Montoya, E. Planas, J. Casal, A comparative analysis of mathematical models for relating indoor and outdoor toxic gas concentrations in accidental releases, J. Loss Prevent. Proc. 22 (2009) 381–391.
[9] D. van Leeuwen, Protection against toxic substances by remaining indoors, in: Methods for the Determination of Possible Damage to People and Objects Resulting from Releases of Hazardous Materials CPR 16E, Green Book, Voorburg, The Netherlands, 1992 (Chapter 6).
[10] B.C. Singer, K.L. Revzan, T. Hotchi, A.T. Hodgson, N.J. Brown, Sorption of organic gases in a furnished room, Atmos. Environ. 38 (2004) 2483–2494.
[11] B.C. Singer, A.T. Hodgson, H. Destaillats, T. Hotchi, K.L. Revzan, R.G. Sextro, Indoor sorption of surrogates for sarin and related nerve agents, Environ. Sci. Technol. 39 (2005) 3203–3214.
[12] T.L. Hilderman, S.E. Hrudey, D.J. Wilson, A model for effective toxic load from fluctuating gas concentrations, J. Hazard. Mater. 64 (1999) 115–134.
[13] G.W. Hoyle, W. Chang, J. Chen, C.F. Schlueter, R.J. Rando, Deviations from Haber's law for multiple measures of acute lung injury in chlorine-exposed mice, Toxicol. Sci. 118 (2010) 696–703.