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On the individual exposure from airborne hazardous releases: The effect of atmospheric turbulence

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Abstract

One of the key problems in coping with deliberate or accidental atmospheric releases is the ability to reliably predict the individual exposure during the event. Furthermore, for the implementation of countermeasures, it is essential to predict the maximum expected dosage and the exposure time within which the dosage exceeds certain health limits. Current state of the art methods, which are based on the concentration cumulative distribution function (cdf) and require the knowledge of the concentration variance and the intermittency factor, have certain limitations especially when the exposure time becomes comparable with the peak spectral time. The proposed method aims at estimating maximum dosage as a function of the exposure time, mean concentration and the turbulence integral time scale. It is much simpler than the cdf models and it poses no restrictions on the exposure time length. One of the important consequences is that it can broaden the capability of the ensemble average computational models to estimate maximum dosage for any exposure time. The method has been tested successfully utilizing the ammonia field experiments FLADIS T16 and T17.

Keywords: Individual exposure; Dosage; Concentration fluctuations; Turbulence integral time scale

1. Introduction

One of the key problems in coping with deliberate or accidental atmospheric releases is the ability to reliably predict individual exposure during the event. Due to the stochastic nature of turbulence, the instantaneous wind field at the time of the release is practically unknown. To assess consequences and countermeasures, one needs to predict or/and estimate the maximum expected dosage within a certain time interval at a particular position. To achieve this, knowledge of the behaviour of concentrations fluctuations at the point under consideration is needed.

The usual methodology to treat this problem today is to try to obtain first the knowledge of the mean concentration (C), the concentration variance (σ_c) and the intermittency factor (γ). The maximum expected concentrations with a given confidence level, are derived from the corresponding concentration cumulative distribution function (cdf) which is considered to be a function of C, σ_c and γ, by assuming a particular shape of the concentration probability density function [e.g. 1].

This theory has been developed for predicting maximum instantaneous concentration. However, it can be applied for time averaged concentrations, provided that the time interval falls well within the inertial range of concentration spectra [2]. It must be noted that in practice, for consequence assessment, one is interested in estimating the maximum dosage, i.e., the concentration integrated over a limited time interval. In addition, the maximum dosage dependence on the integration time interval is also required. In practical terms, the averaging time intervals are of the order of a few seconds sufficiently far from the source. This approach can be utilized to estimate dosages at the inhalation time level which is of the order of 3 s or more. However, in order to estimate dosages near the source or/and for longer time intervals there is a need to have some knowledge of the turbulence time scales which are directly related to the peak spectra time. The observation data have shown that as the integration time interval approaches the peak spectra time, there is a distortion in the cumulative distribution function. More specifically, the probability of concentrations higher than the mean

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concentration is significantly reduced. There is also an increase of the intermittency factor indicated by the reduced cumulative probability at low concentrations [e.g. 2].

From the above discussion it is clear that the application of the above-mentioned methodology to calculate exposure times above which the received dosage exceeds the imposed health limits presents certain problems. The method proposed in this paper aims at removing these limitations.

2. Methodology

In the present paper a new methodology is presented which is much simpler than the cdf models and in the same time more flexible with respect to the dose time interval. The fundamental problem to be solved is that there is a hazardous substance release and we want to estimate the maximum expected dosage $[D(\Delta\tau)]_{\max}$ at a particular receptor over an exposure time $\Delta\tau$:

$$[D(\Delta\tau)]_{\max} = \left[\int_{\Delta\tau} C(t)\,\mathrm{d}t\right]_{\max} = C_{\max}(\Delta\tau)\Delta\tau \tag{1}$$

The maximum average concentration $C_{\max}(\Delta\tau)$ is defined as

$$C_{\max}(\Delta\tau) = \frac{1}{\Delta\tau}\left[\int_{\Delta\tau} C(t)\,\mathrm{d}t\right]_{\max} \tag{2}$$

and $C(t)$ is the instantaneous concentration at the receptor point. Thus, the problem is transferred to the estimation the maximum time averaged concentration $C_{\max}(\Delta\tau)$.

Let us assume that at a certain receptor point a high time resolution concentration instrument produces one reading per $\Delta\tau$ seconds. In an atmospheric boundary layer $\Delta\tau$ could for example of the order of 1 s. Let us make the additional assumption that the instrument reading represents the time average concentration $C(\Delta\tau)$ over the resolution time $\Delta\tau$. For simplicity the concentration field is assumed stationary with a constant mean concentration $\bar{C}$ and integral time scale T_{L}. Following the concentration signal for an infinite time, we will observe at a particular time step a maximum time averaged concentration value $C_{\max}(\Delta\tau)$. The signal can be numerically processed and Eq. (2) can be used to estimate the maximum time averaged concentration $C_{\max}(\Delta\tau)$ for any time interval $\Delta\tau$ larger than the resolution time interval.

The $C_{\max}(\Delta\tau)/\bar{C}$ ratio is expected to have values much greater than unity. As an example, for $\Delta\tau = 1$ s, the ratio could reach values in the order of 100 [1]. As $\Delta\tau$ increases the ratio is expected to decrease. For large enough $\Delta\tau$, the ratio tends to become unity since practically the estimated values $C_{\max}(\Delta\tau)$ are reaching the mean value $\bar{C}$. The time interval that $C_{\max}(\Delta\tau)$ starts approaching the $\bar{C}$ value is expected to be scaled by the integral time T_{L}.

The fundamental question is how $C_{\max}(\Delta\tau)/\bar{C}$ varies with $\Delta\tau$. In fact, significant amount of work exists in the literature correlating $C_{\max}(\Delta\tau)$ at different averaging times $\Delta\tau$. More specifically, suppose that for a particular location, the maximum expected concentrations averaged over the times $\Delta\tau$ and ΔT are $C_{\max}(\Delta\tau)$ and $C_{\max}(\Delta T)$, respectively. It has been demonstrated that a proper correlation to relate these two concentrations is [3,4]:

$$\frac{C_{\max}(\Delta\tau)}{C_{\max}(\Delta T)} = \left(\frac{\Delta\tau}{\Delta T}\right)^{-n} \tag{3}$$

The derivation of the given correlation in Eq. (3) is based on past efforts to estimate maximum time averaged concentrations from Gaussian plume at different averaging times [5]. A summary of the relevant work is given in the IAEA Safery Series No. 50-SG-S3 [6]. The value of the exponent n for average times of 1 h or less seems to be affected mainly from the source height and the atmospheric stability. The n values derived from field data show a range 0.2–0.5 for ground sources and 0.12–0.7 for elevated ones.

In Eq. (3) let us select ΔT large enough so that $C_{\max}(\Delta T) \approx \bar{C}$. Based on the above-mentioned discussion ΔT is expected to be scaled by the integral time scale T_{L}. Thus, Eq. (3) can be transformed to the equation:

$$\frac{C_{\max}(\Delta\tau)}{\bar{C}} = a\left(\frac{\Delta\tau}{T_{\mathrm{L}}}\right)^{-n} \tag{4}$$

However both Eqs. (3) and (4) have an inconsistency. When $\Delta\tau$ becomes very large, $C_{\max}(\Delta\tau)$ tends to zero which is not correct. Based on the above discussion $C_{\max}(\Delta\tau)$ should tend to $\bar{C}$. To remove this inconsistency the following modification is proposed for examination:

$$\frac{C_{\max}(\Delta\tau)}{\bar{C}} = 1 + a\left(\frac{\Delta\tau}{T_{\mathrm{L}}}\right)^{-n} \tag{5}$$

It should be noticed that for $C_{\max}(\Delta\tau)/\bar{C} \gg 1$ which is the case for small $\Delta\tau$, Eqs. (4) and (5) become identical. The parameter a is a coefficient depending among others, on how many times we need to multiply T_{L} in order $C_{\max}$ to reach $\bar{C}$. Concerning additional dependency of parameter a, it is logical to expect that as the concentration spread is higher the value of a is higher. In fact experimental evidence on $C_{\max}(\Delta\tau)/\bar{C}$ behaviour suggests that the parameter a depends strongly on fluctuation intensity:

$$i^2 = \frac{\sigma_{\mathrm{C}}^2}{\bar{C}^2} \tag{6}$$

For example Lung et al. [1] analyzing their experimental data have shown by best experimental fit that

$$\frac{C_{\max}(1\,\mathrm{s})}{\bar{C}} \approx 1 + 3.66 i^2 \tag{7}$$

It should be noted that this experiment refers to a near ground radioactive source Kr^{85} and concentration signals of 1 Hz obtained by highly sensitive proportional counter tubes [1].

The relationship (7) suggests:

$$a = bi^2 \tag{8}$$

Thus the following correlation $C_{\max}(\Delta\tau)/\bar{C}$ is finally proposed:

$$\frac{C_{\max}(\Delta\tau)}{\bar{C}} = 1 + bi^2\left(\frac{\Delta\tau}{T_{\mathrm{L}}}\right)^{-n} \tag{9}$$

It is reminded that $C_{\max}(\Delta\tau)$ refers to a stationary signal of infinite time and therefore is expected to have a specific value. In statistical terms it represents a particular point in the pdf of $C(\Delta\tau)$ corresponding to a specific critical value. The variance of $C(\Delta\tau)$ is related to $C_{\max}(\Delta\tau)$. In the literature there are several methods to estimate this variance [11]. However, the relationship between $C_{\max}(\Delta\tau)$ and the variance of $C(\Delta\tau)$ strongly depends on the particular pdf distribution which is likely to change with respect to $\Delta\tau$.

The parameters b and n in Eq. (9) can be estimated experimentally. One should keep in mind that in reality the available concentration signals have a finite duration rather than an infinite one. One restriction that we can put on the calibration signals is that the sampling time is sufficiently long and the signals are nearly stationary. In any case the stochastic nature of turbulence combined with a finite sampling time does not allow us to give a precise value of these parameters. What we are aiming at the present work is a first estimation of their "reference" values and their variability.

Indicative values of these parameters can be given even at this stage. An indicative value of $n = 0.3$ is reasonable based on past experience especially for ground releases. Indicative value for b can be derived from the work of Lung et al. [1]. It is suggested in this work a reference value for $T_L = 20$ s. From Eq. (7) we can derive as indicative value $b = 1.5$. Summarizing, the *indicative* "reference" values for Eq. (9) constants are suggested to be

$$b = 1.5, \qquad n = 0.3 \tag{10}$$

In the remainder of the paper, the relations (9) and (10) will be tested using the data from the FLADIS T16 and T17 field experiments.

3. The FLADIS experiment

The original objective of the FLADIS field experiments was to investigate dispersion of flashing releases of ammonia in the atmosphere [7]. For the purposes of this study data from Trials 16 and 17 were utilized, which were the most successful trials. The ammonia was released horizontally as a flashing jet. The experimental characteristics are given in Table 1 [8].

The sensors were mainly arranged in three arcs across an ideal plume centreline in order to detect plume dimensions. The first arc (Arc 20) is located at approximately 20 m from the release point where the concentration field is expected to be affected by the source presence and the local stability (heavy gas). The second arc (Arc 70) was placed at approximately 70 m, where the concentration field is expected to have still some influence from the source but the stability is near neutral. The concentration field at Arc 235, which was placed at approximately 235 m, is expected to be passive gas with practically no influence from the source. Additionally several of the concentration sensors were placed on masts at various distances from the release point. The data utilized in the present study are the concentration time series with 1 s temporal resolution.

Table 1
FLADIS experimental characteristics

Parameter	T16	T17
Release duration (min)	20	25
Release rate (kg/s)	0.27	0.27
Average wind speed at 10 m (m s^{-1})	4.4	3.7
Wind direction (relative to centreline) (°)	−8	−28
Ambient temperature (°C)	16	16
Atmospheric pressure (mb)	1020	1020
Relative humidity (%)	62	63
Friction velocity (m s^{-1})	0.41	0.31
Surface roughness (m)	~0.04	~0.04
Monin-Obukhov length (m)	138	59

3.1. Data assessment

The purpose of the data assessment was to select the time series data set that are qualified for the present analysis. Every sensor signal has been visually tested for obvious instrument misbehaviour.

A further screening of the visually accepted data has been performed following the procedure shown in Fig. 1. Initially all sensors containing less than 750 data points were discarded. Looking at the data more closely, this number was considered as a minimum threshold above which all the predominant features of each sensor time series are assumed to be present. The remaining sensors were examined for stationarity, discarding those that were not stationary. The concept of stationarity is important in the present analysis as it provides a sound indication of no systematic change in the series mean and the variance. The formal method for testing the stationarity of a time series is the "unit root" test.

In order to examine the stationarity characteristics of each sensor time series, initially the series sample autocorrelation function (ACF) up to 50 lags was estimated. A visual exami-

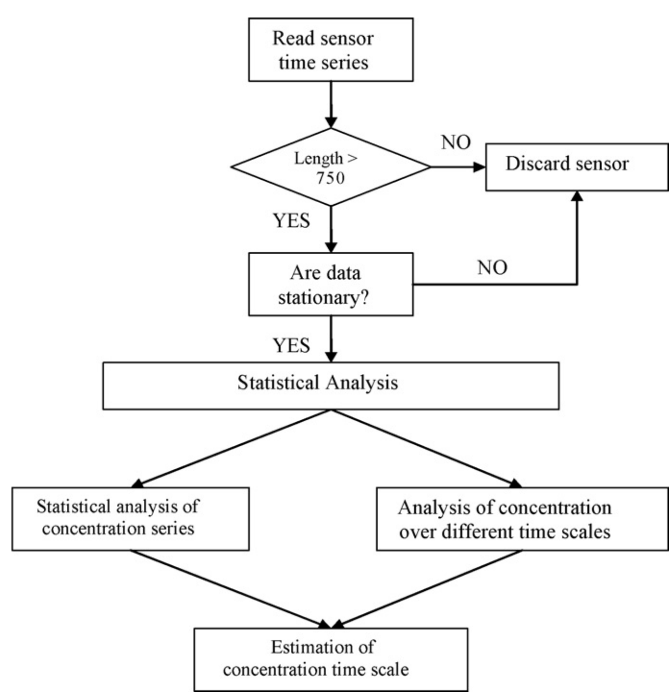

Fig. 1. Flowchart for data screening.

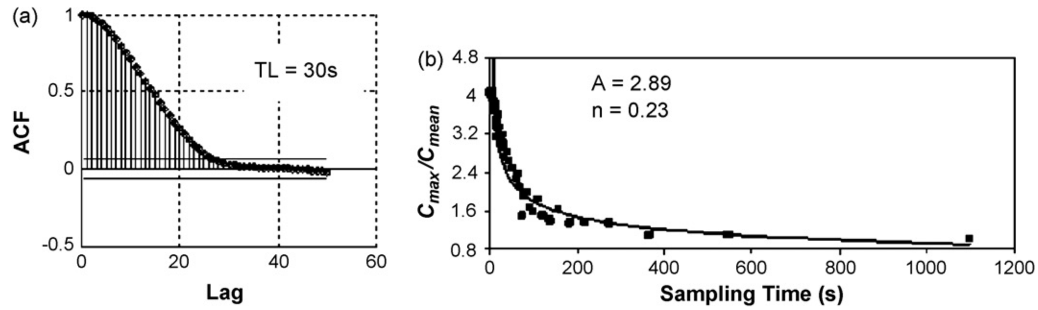

Fig. 2. Statistical properties of the concentration time series at ($X = 235$, $Y = 0$, $Z = 9$). The turbulence integral time scale is calculated by an exponential law to the ACF, whereas the parameters A and n by a power-law fit to Eq. (12) ($R^2 = 0.8562$).

nation was performed rejecting all sensors that exhibited slow drop in the ACF values, exceeding the 95% statistical significance level, defined as $\sigma = \pm 1.96/\sqrt{N}$, where N is the number of data.

Then, the Phillips and Perron test [9] for the presence of unit root which is a nonparametric method of checking for higher-order serial correlation in a series was applied. The test regression for the Phillips–Perron (PP) test is the first order autoregressive, AR(1), process:

$$\Delta y_t = \mu + \gamma y_{t-1} + \varepsilon_t \quad (11)$$

Here μ and γ are model coefficients, and ε is assumed to be white noise. The test examines the null hypothesis H_0: $\gamma = 0$ against the alternative H_1: $\gamma < 0$. The t-statistic under the null hypothesis of a unit root does not have the conventional t-distribution. The PP test makes a correction to the t-statistic of the γ coefficient from the above regression to account for the serial correlation in ε. The correction is nonparametric since we use an estimate of the spectrum of ε at frequency zero that is robust to heteroskedasticity and autocorrelation of unknown form. MacKinnon [10], implemented a much larger set of simulations showing the distribution under the null hypothesis is non-standard, and simulated the critical values for selected sample sizes. The test statistic and the critical levels at the 1 and 5% confidence intervals are estimated. If the hypothesis is accepted at the 5% level, then this is evidence of weak stationarity.

After performing the above-mentioned checking procedure, the number of signals that have been selected for the present analysis were 24 for T16 and 18 for T17.

The most complete dataset, T16, is selected for the model constant refinement whereas the T17 data are used for the model validation.

Table 2
FLADIS T16: estimated parameters for stationary sensors (the source is located at position ($X = 0$, $Y = 0$, $Z = 1.5$))

Sensor ID	X_c (m)	Y_c (m)	Z_c (m)	Station.[a]	i^2	A	n	T_L (s)
1	18.5	−8.9	0.1	S	8.71	27.71	0.34	16.70
2	18.5	−8.9	1.5	S	4.85	22.67	0.37	15.56
3	19.5	−5.9	0.1	S	2.49	12.55	0.28	24.80
4	19.5	−5.9	1.5	S	2.70	14.86	0.31	18.79
5	20	−2.9	0.1	S	0.54	2.83	0.28	21.35
6	20	−2.9	1.5	S	1.02	6.16	0.36	15.67
7	20	0	0.1	S	0.26	1.70	0.27	30.38
8	20	0	0.75	S	0.30	1.64	0.24	34.43
9	20	0	1.5	S	0.39	2.38	0.24	22.67
10	20	0	3	S	0.64	6.30	0.38	19.00
11	20	3.1	0.1	S	0.24	1.30	0.24	26.54
12	20	3.1	1.5	S	0.31	3.16	0.37	13.22
13	19.5	6.1	0.1	W	0.46	2.14	0.21	40.78
14	19.5	6.1	1.5	S	0.70	5.45	0.35	18.37
15	18.5	9.1	0.1	S	1.53	6.72	0.28	19.09
16	18.5	9.1	1.5	S	1.52	10.79	0.37	15.50
17	70	−10	0.5	S	0.87	4.77	0.29	20.07
18	70	0	0.1	S	0.48	3.06	0.25	34.02
19	70	0	2	S	0.48	2.89	0.25	31.72
20	70	0	4	S	0.49	4.76	0.29	38.45
21	70	10	0.5	S	0.30	2.61	0.28	25.11
22	68	20	0.5	S	1.02	5.94	0.27	50.12
23	231.5	−30	1.5	W	2.52	13.63	0.29	31.28
24	235.5	0	9	S	0.41	4.10	0.32	30.74

[a] S = stationary series, W = weak stationarity.

3.2. Data analysis

Firstly, the effort has been concentrated on T16 accepted data set in order to obtain the experimental values of b and n constants introduced in the proposed model Eq. (9) and to compare these values with the indicative ones given in the relation (10).

The exponent n is estimated from the relationship equivalent to model Eq. (9):

$$\frac{C_{\max}(\Delta\tau)}{\bar{C}} - 1 = A\Delta\tau^{-n} \tag{12}$$

applying least squares fitting techniques.

The time intervals $\Delta\tau$ selected in the analysis for each receptor are multiples of the time resolution (1 s) provided they do not exceed the half value of the signal duration.

The constant b is obtained from the parameter A via the relationship:

$$b = \frac{A}{i^2 T_L^n} \tag{13}$$

The integral time scale T_L, is estimated from signal autocorrelation function (ACF), using the exponential law [11]:

$$\mathrm{ACF}(\tau) = e^{-\tau/T_L} \tag{14}$$

and applying least squares fitting.

An example of the outcome of such analysis, illustrated in a graphical form, is given in Fig. 2, with respect to the sensor ($X = 235$, $Y = 0$, $Z = 9$). In Fig. 2(a), we can observe that the ACF values exhibit an exponential drop, and after the 27th lag the values are assumed to be statistically not different from zero. In Fig. 2(b), it can be observed that for different exposure times $\Delta\tau$, the ratio $C_{\max}(\Delta\tau)/\bar{C}$ is well correlated with the power law proposed by Eq. (12).

3.3. The results

3.3.1. The FLADIS T16 experiment

Table 2 summarizes the results of the above-mentioned analysis for FLADIS T16 experiment. It is noted that for the Arc-20 sensors the vast majority of those exceeding the threshold number (750 data points) have strong stationary characteristics. This is explained from the short distance from the source. All the Arc-70 sensors with a sufficient number of samples are stationary, with an almost equal split of strong and weak stationary characteristics. For the Arc-235 sensors the majority exhibit non-stationary behaviour and thus excluded from further analysis, whilst a significant number has weak stationary characteristics.

Table 3
FLADIS T16: arc-wise analysis of n

Location	n			T_L		
	Minimum	Mean	Maximum	Minimum	Mean	Maximum
Arc 20	0.21	0.31	0.38	13.22	22.06	40.79
Arc 70	0.25	0.28	0.30	20.08	33.26	50.12
Arc 235	0.29	0.31	0.32	30.74	31.02	31.29

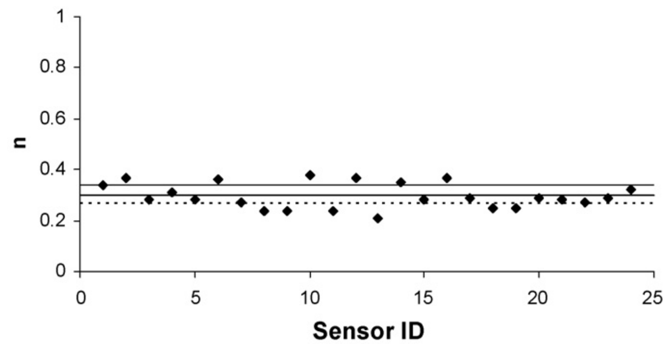

Fig. 3. Estimate of n with respect to sensor ID for T16. The data mean is 0.302 and the standard deviation 0.036.

Table 3 shows the mean values of n and T_L and their spread for each one of the three arcs.

The exponent n varies from 0.21 to 0.38 (both in Arc 20). No obvious trend is observed to the mean values of n with respect to the distance from the source, especially if one takes into account the statistical spread of the values. The T_L values vary from 13 s (Arc 20) to 50 s (Arc 70). The T_L variation can be explained from the source proximity, local stability, ambient turbulence properties, instrument errors, etc. The highest spread in the values for n and T_L is observed on Arc 20 which is characterized by the source proximity and the heavy gas effects.

In Fig. 3 the variation of the exponent n along the various sensors is shown. The mean value n and its variance can be derived as

$$n = 0.302, \qquad \sigma_n = 0.036 \tag{15}$$

It is important to note that the mean value of n is practically the same with the indicative one given in (10). If one takes into consideration that the FLADIS and the Kr^{85} experiments are totally different, this particular result adds an important element to the present model validity.

In Fig. 4, the parameter A given in Table 2, is plotted against the quantity $i^2 T_L^n$. A linear fit without a constant term returns the slope value, b, equal to 1.507 ($\sigma_b = 0.088$) and $R^2 = 0.9271$. This value is very near to the indicative value given in relation (10). It is clear from the above discussion that FLADIS T16 data

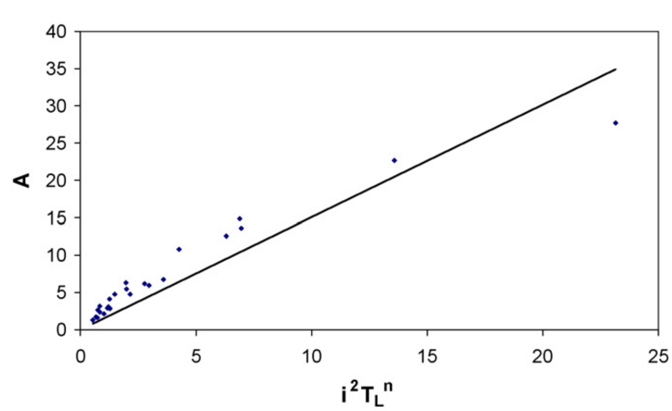

Fig. 4. Correlation of A to $i^2 T_L^n$. The data follow a linear relationship with slope 1.507 ($\sigma = 0.088$) and $R^2 = 0.9271$.

Table 4
FLADIS T17: the experimental parameters

X (m)	Y (m)	Z (m)	Station.[a]	C_{mean} (% vol)	σ_c (% vol)	T_L (s)
20	−2.9	0.1	S	0.083678	0.18689	11.29
20	−2.9	1.5	S	0.027304	0.05764	10.19
20	0	0.1	S	0.50254	0.54822	17.24
20	0	0.75	S	0.21721	0.22543	19.40
20	0	1.5	S	0.083201	0.096948	13.98
20	0	3	S	0.011964	0.013768	13.30
20	3.1	0.1	S	1.3262	0.70044	22.55
20	3.1	1.5	S	0.1837	0.12108	11.62
19.5	6.1	0.1	S	1.5406	0.47474	37.85
19.5	6.1	1.5	S	0.28422	0.14979	8.87
18.5	9.1	0.1	S	1.3723	0.43882	20.16
18.5	9.1	1.5	S	0.23472	0.10734	18.16
17	12.1	0.1	W	0.47995	0.34627	13.56
17	12.1	1.5	S	0.11674	0.09874	10.78
70	10	0.5	S	0.047832	0.044237	29.48
68	20	0.5	S	0.13749	0.065861	25.01
65	30	0.5	S	0.10894	0.052211	20.01
227.5	50	9	W	7.6606×10^{-4}	12.472×10^{-4}	70.12

[a] S = stationary series, W = weak stationarity.

analysis strengthens considerably the validity of the model Eq. (9) with b and n nominal values the ones given in (10).

3.3.2. The FLADIS T17 experiment

In Table 4 the FLADIS T17 experimental data set, that are characterized as stationary, are used to validate the proposed model.

More specifically the $\bar{C}, \sigma_c$ and T_L parameters given in Table 4 have been utilized in Eqs. (9) and (10) to predict the $C_{max}(\Delta\tau)$ values. Fig. 5(a)–(d) presents a comparison between the predicted and modelled values for the time averaged maximum concentrations $C_{max}(\Delta\tau)$ obtained with averaging time intervals $\Delta\tau = 1$, 2, 5 and 10 s, respectively. The model parameter spread confines the $C_{max}(\Delta\tau)$ error eventually within a factor of

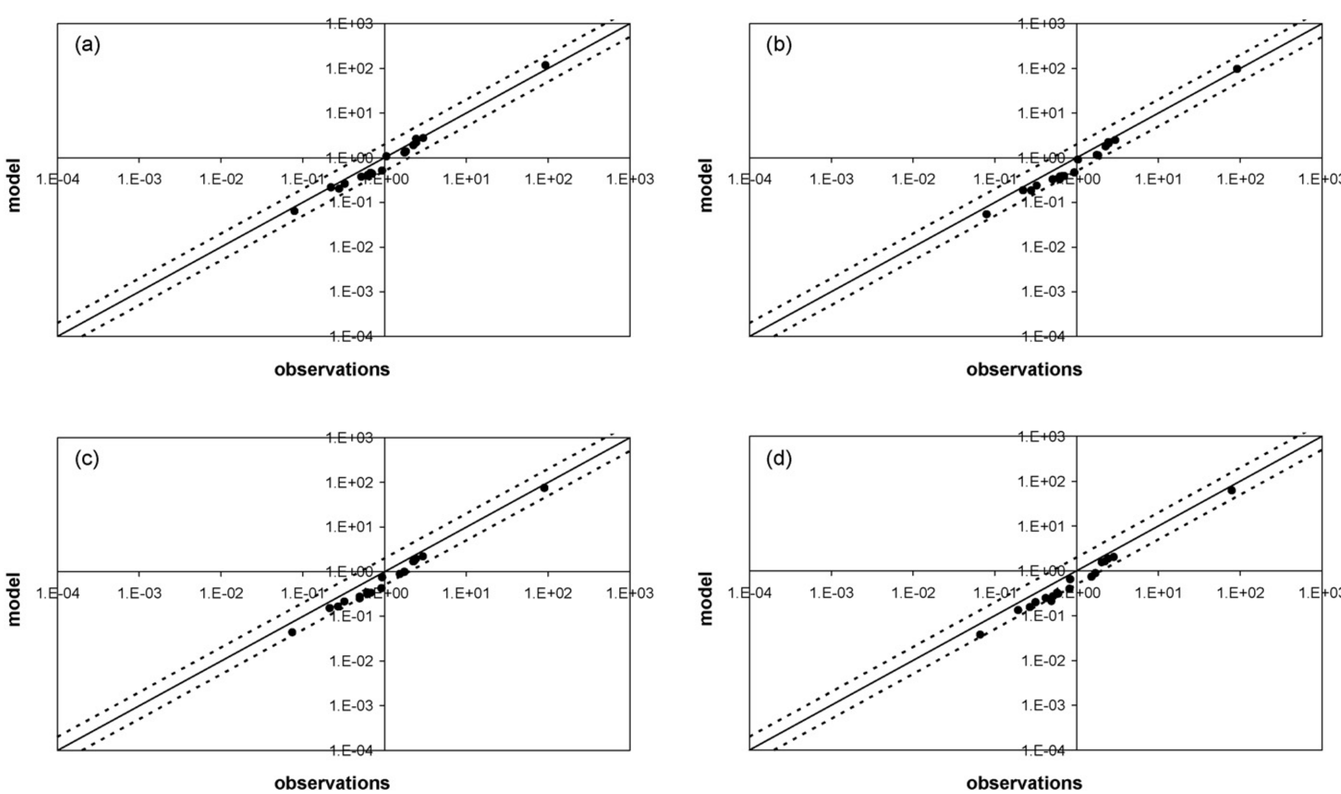

Fig. 5. Comparison of experimentally observed and model estimated C_{max} for (a) $\Delta\tau = 1$ s, (b) $\Delta\tau = 2$ s, (c) $\Delta\tau = 5$ s and (d) $\Delta\tau = 10$ s. The majority of the data are well within a factor of 2.

2. Such an error can be considered quite satisfactory especially for field data where the ambient turbulence properties cannot be clearly defined.

4. Conclusions

The present work addresses the fundamental problem of reliable prediction of individual exposure in case of deliberate or accidental atmospheric releases of hazardous substances. Based on the above discussion the following conclusions can be drawn:

(1) The usual methods, based on the concentration cumulative distribution function (cdf) and requiring additional knowledge of the concentration variance and the intermittency rate, have limitations especially when the exposure time is large enough to be comparable with the peak spectral time.
(2) The present model is much simpler than the cdf models and at the same time more flexible with respect to the exposure time length. The main idea is to correlate maximum time averaged concentration and dosage with the mean concentration, the concentration variance and the integral time scale. The whole approach has been restricted at this stage only to stationary or nearly stationary conditions.
(3) The model constants given in Eq. (10) have at this stage the character of indicative reference values. These values will need to be further refined in the future by utilizing additional existing and new data. This refinement will include not only the reference values but also their variability.
(4) The new model has been applied successfully to the ammonia field experiments FLADIS T16 and T17.
(5) The FLADIS T16 data analysis has also shown that the source proximity and the local stability are factors that seem to affect more the spread of the model constants rather than their mean value. This spread creates an error in the model predicted maximum averaged concentrations, which however is confined to within a factor of 2.
(6) The new model can broaden the capability of the ensemble average computational models such as RANS-CFD to estimate dosage at any exposure time provided that are able to reliably predict concentrations, concentration fluctuations and integral time scales.

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