Modelling the Occurrence of Large Concentration Values in Pollutant Plumes

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Abstract: Large concentration values are of particular interest in assessing hazards or nuisance associated with clouds or plumes of toxic, flammable or malodorous gases. Previously we have used statistical extreme value theory to analyse the probability distribution of large concentrations at a fixed point in space. Here we present a probabilistic model for the temporal occurrence (at a discrete set of points in space – the receptor locations) of large concentrations in a plume. We consider a fixed downwind distance, and take the plume centreline to be at one of a discrete set of crosswind positions. Changes of centreline position are modelled by a continuous time Markov chain. At each receptor location the large concentration values are taken to occur at the points of a Poisson process in time whose rate depends on the distance from the centreline. We present some results from fitting this model to data from atmospheric point source experiments. The fitted parameters for the Markov chain are not very robust or reliable. Possible reasons for this are discussed, and suggestions are made for how the results might be improved.

Keywords: Pollutant dispersion; Concentration fluctuations; Large concentrations; Markov-modulated Poisson process; Extreme value theory

1. INTRODUCTION

The hazards or nuisance associated with clouds or plumes of toxic, flammable or malodorous gases dispersing in the atmosphere can be strongly influenced by the occurrence of large concentration values. As well as the size of the concentration value, the spatio-temporal structure of the concentration field is also important. For example, for exposure to toxic gases, the damage done depends, in a nonlinear way, on the concentration history over the whole exposure interval [Ride, 1995]. With flammable gases, whether ignition occurs at a particular location and time depends on the concentration value at that point, but to determine whether burn-up of a cloud is possible it is necessary to have detailed knowledge of the space-time structure of the concentration field.

Since the dispersion takes place in a turbulent atmosphere, the concentration field is random, and we are interested in the probability distribution of large values of concentration, and of the times and locations at which they occur. We have previously considered the former [Mole et al. 1995; Anderson et al. 1997; Munro et al. 2001]; here we concentrate on the times at which large values of concentration occur. We restrict attention to the case of a source emitting at a constant rate into a stationary atmospheric flow, so the concentration is also stationary.

Anderson et al. [1997] used a switching Poisson process model for the values and the occurrence times of large concentrations, at a fixed point in space. The Poisson process in that model switched on and off, to mimic the intermittent concentration time series observed in meandering atmospheric plumes [see, for ex-

ample, Hanna 1984; Murlis 1986; Mylne and Mason 1991; Mole and Jones 1994], with large concentrations being observed when the receptor location is close to the instantaneous plume centreline.

As a first step towards a model for the space-time structure of large concentration values, we introduce a model which seeks to represent the meandering of the plume, and the feature that large concentration values are more likely to occur close to the instantaneous plume centreline than far from it. While the concentration values themselves could also be included in the model, for simplicity we model only the times of occurrence of large values at a finite, discrete set of spatial points (which will be thought of as the receptor locations where concentration measurements are made). We take large values to be those concentrations which exceed a specified high threshold.

We consider a fixed distance downwind of the source, and let the plume centreline take one of a finite, discrete set of crosswind positions. For simplicity, as a first approximation, we ignore vertical movement of the plume centreline. We assume that the movement of the plume centreline can be described by a continuous time Markov chain with constant transition rate matrix.

At a fixed crosswind location we assume that large concentration values occur as the points of a Poisson process in time. To produce the desired variation with relative position, the rate of the Poisson process needs to be a decreasing function of distance from the instantaneous plume centreline position.

In §2 we describe the details of the overall model that results from these assumptions, and derive the corresponding likelihood function. §3 deals with the fitting of the model to experimental data by maximising the likelihood, and §4 discusses the results and makes suggestions for improving them.

2. THE MARKOV MODULATED POISSON PROCESS MODEL

Suppose we have m receptors, at locations r_k $(k = 1, \ldots, m)$. We model the meandering of the plume by assuming the crosswind centreline position $Y(t)$ can take one of a set of values y_j $(j = 1, \ldots, n)$. We assume that $Y(t)$ is governed by a continuous time Markov chain with constant transition rate matrix $Q = (q_{ij})$. Then

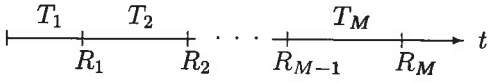

Figure 1. The times T_i between occurrences of large concentration values, and the receptor locations R_i at which they occur.

$$P[Y(t+\Delta t) = y_j | Y(t) = y_i] = \delta_{ij} + q_{ij}\Delta t + o(\Delta t)$$

where δ_{ij} is the Kronecker delta, $q_{ij} \geq 0$ for $i \neq j$, $q_{ii} \leq 0$, and

$$\sum_{j=1}^{n} q_{ij} = 0.$$

At receptor location r_k, large values of concentration are taken to occur at the points of a Poisson process of constant rate $\lambda_{i,k}$ (≥ 0) if $Y(t) = y_i$. If we let $N_k(t_1, t_2)$ be the number of large concentration values at r_k in (t_1, t_2), then this means that

$$P[N_k(t, t+\Delta t) = 0 | Y(t) = y_i] = 1 - \lambda_{i,k}\Delta t + o(\Delta t)$$
$$P[N_k(t, t+\Delta t) = 1 | Y(t) = y_i] = \lambda_{i,k}\Delta t + o(\Delta t)$$
$$P[N_k(t, t+\Delta t) > 1 | Y(t) = y_i] = o(\Delta t).$$

The resulting overall model for the occurrence of large concentration values is a homogeneous (in time) Markov-modulated Poisson process (MMPP) – see for example Fischer and Meier-Hellstern [1992].

We consider the time intervals T_i $(i = 1, \ldots, M)$ between occurrences of large concentration values, having aggregated these occurrences over all receptors. We let R_i be the receptor location at which the ith large value occurs – see Figure 1. We also let $N(t_1, t_2)$ be the number of large concentration values, over all receptors, which occur in (t_1, t_2), i.e.

$$N(t_1, t_2) = \sum_{k=1}^{m} N_k(t_1, t_2).$$

Then

$$
\begin{aligned}
&P[t < T_1 < t + \Delta t, R_1 = r_k, Y(t) = y_j | Y(0) = y_i] \\
&= P[N(0,t) = 0, N(t, t+\Delta t) = 1, R_1 = r_k, \\
&\qquad Y(t) = y_j | Y(0) = y_i] \\
&= P[N(0,t) = 0, Y(t) = y_j | Y(0) = y_i] \\
&\quad \times P[N(t, t+\Delta t) = 1, Y(t) = y_j | Y(0) = y_i] \\
&\quad \times P[R_1 = r_k, Y(t) = y_j | Y(0) = y_i] \\
&= h_{ij}(t)(\lambda_j \Delta t)\left(\frac{\lambda_{j,k}}{\lambda_j}\right) = h_{ij}(t)\lambda_{j,k}\Delta t,
\end{aligned}
$$

where

$$
h_{ij}(t) = P[N(0,t) = 0, Y(t) = y_j | Y(0) = y_i] \tag{1}
$$

$$
\lambda_i = \sum_{k=1}^{m} \lambda_{i,k},
$$

since the aggregate over all receptors of the individual Poisson processes gives a Poisson process whose rate is the sum of the individual rates.

Writing $\Lambda^{(k)} = \mathrm{diag}(\lambda_{1,k}, \ldots, \lambda_{n,k})$ and $H = (h_{ij})$ we have

$$
\begin{aligned}
&P[t < T_1 < t + \Delta t, R_1 = r_k, Y(t) = y_j | Y(0) = y_i] \\
&\quad = \left[H(t)\Lambda^{(k)}\right]_{ij} \Delta t.
\end{aligned}
$$

Similarly,

$$
\begin{aligned}
&P[t_1 < T_1 < t_1 + \Delta t_1, t_2 < T_2 < t_2 + \Delta t_2, \\
&\qquad R_1 = r_{k_1}, R_2 = r_{k_2}, Y(t_1 + t_2) = y_l | Y(0) = y_i] \\
&= \sum_{j=1}^{n} P[t_1 < T_1 < t_1 + \Delta t_1, R_1 = r_{k_1}, \\
&\qquad\qquad Y(t_1) = y_j | Y(0) = y_i] \\
&\quad \times P[t_2 < T_2 < t_2 + \Delta t_2, R_2 = r_{k_2}, \\
&\qquad\qquad Y(t_1 + t_2) = y_l | Y(t_1) = y_j] \\
&= \sum_{j=1}^{n} \left[H(t_1)\Lambda^{(k_1)}\right]_{ij} \Delta t_1 \left[H(t_2)\Lambda^{(k_2)}\right]_{jl} \Delta t_2 \\
&= \left[H(t_1)\Lambda^{(k_1)} H(t_2)\Lambda^{(k_2)}\right]_{il} \Delta t_1 \Delta t_2.
\end{aligned}
$$

Extending this argument to all the occurrences of large concentrations gives

$$
\begin{aligned}
&P[t_j < T_j < t_j + \Delta t_j; R_j = r_{k_j} \text{ for } j = 1, \ldots, M; \\
&\qquad Y\left(\sum_{j=1}^{M} t_j\right) = y_l \,|\, Y(0) = y_i] \\
&\quad = \left[\prod_{j=1}^{M} H(t_j)\Lambda^{(k_j)} \Delta t_j\right]_{il}.
\end{aligned}
$$

Considering all possible centreline positions y_l, we have

$$
\begin{aligned}
&P[t_j < T_j < t_j + \Delta t_j; R_j = r_{k_j} \\
&\qquad \text{for } j = 1, \ldots, M | Y(0) = y_i] \\
&= \sum_{l=1}^{n} \left[\prod_{j=1}^{M} H(t_j)\Lambda^{(k_j)} \Delta t_j\right]_{il} \\
&= \left\{\left[\prod_{j=1}^{M} H(t_j)\Lambda^{(k_j)} \Delta t_j\right] u\right\}_i,
\end{aligned}
$$

where $u = (1, 1, \ldots, 1)^T$. If we assume that the centreline position Y is initially in equilibrium (with stationary state π satisfying $\pi Q = 0$ and $\pi u = 1$) then

$$
\begin{aligned}
&P[t_j < T_j < t_j + \Delta t_j; R_j = r_{k_j} \text{ for } j = 1, \ldots, M] \\
&\quad = \sum_{i=1}^{n} \pi_i \left\{\left[\prod_{j=1}^{M} H(t_j)\Lambda^{(k_j)} \Delta t_j\right] u\right\}_i.
\end{aligned}
$$

Thus, given the observations $\{T_j = t_j\}$ and $\{R_j = r_{k_j}\}$, the likelihood $\mathfrak{L}$ is given by

$$
\mathfrak{L} = \pi \left\{\prod_{j=1}^{M} H(t_j)\Lambda^{(k_j)}\right\} u. \tag{2}
$$

To be able to fit a model to experimental data by maximising $\mathfrak{L}$, we need to specify the transition rate matrix Q for the plume centreline position (which also determines π), and the Poisson process rates $\lambda_{i,k}$ for the occurrence of large concentration values at the receptors. H is fully determined by these, as shown below.

2.1 The Matrix H

From the definition (1),

$$
\begin{aligned}
&h_{ij}(t + \Delta t) = P[N(0, t+\Delta t) = 0, \\
&\qquad Y(t + \Delta t) = y_j | Y(0) = y_i] \\
&= \sum_{l=1}^{n} P[N(0,t) = 0, Y(t) = y_l | Y(0) = y_i] \\
&\qquad \times P[N(t, t+\Delta t) = 0 | Y(t) = y_l] \\
&\qquad \times P[Y(t + \Delta t) = y_j | Y(t) = y_l] \\
&= h_{ij}(t)(1 - \lambda_j \Delta t)(1 + q_{jj}\Delta t) \\
&\quad + \sum_{l \neq j} h_{il}(t)(1 - \lambda_l \Delta t) q_{lj} \Delta t + o(\Delta t),
\end{aligned}
$$

which implies that

$$
\begin{aligned}
\frac{d}{dt} h_{ij}(t) &= h_{ij}(t)(q_{jj} - \lambda_j) + \sum_{l \neq j} h_{il}(t) q_{lj} \\
&= \sum_{l=1}^{n} h_{il}(t)(q_{lj} - \Lambda_{lj}),
\end{aligned}
$$

where

$$\Lambda = \sum_{k=1}^{m} \Lambda^{(k)} = \mathrm{diag}(\lambda_1, \ldots, \lambda_n).$$

Thus

$$\frac{dH}{dt} = H(Q - \Lambda),$$

with $H(0) = I$, so

$$H = e^{(Q-\Lambda)t}. \tag{3}$$

2.2 Plume Centreline Movement

We choose $n = 5$, so we have 5 possible positions of the plume centreline. We take these positions to be equally spaced, with separation Δy, and assume that the middle position coincides with the mean centreline position. Furthermore, we assume that the plume movement is symmetric about the mean position, and that the centreline can only move to positions adjacent to its current position. This implies that Q must take the form

$$Q = \beta \begin{pmatrix} -\gamma & \gamma & 0 & 0 & 0 \\ \alpha & -1 & 1-\alpha & 0 & 0 \\ 0 & \frac{1}{2} & -1 & \frac{1}{2} & 0 \\ 0 & 0 & 1-\alpha & -1 & \alpha \\ 0 & 0 & 0 & \gamma & -\gamma \end{pmatrix},$$

where α, β and γ are constants, with $\beta > 0$, $\gamma > 0$ and $0 < \alpha < 1$. β is the rate of transitions from the mean position, and $\gamma\beta$ is the rate of transitions from each of the extreme positions. This allows for the possibility that, in different cases, the plume spends different proportions of time in the extremities and in the mean position. The equilibrium distribution π, satisfying $\pi Q = 0$ and $\pi u = 1$, is

$$\pi = (a, b, c, b, a),$$

where

$$a = \frac{\alpha}{2\{\alpha + \gamma(2-\alpha)\}},$$

$$b = \frac{\gamma}{2\{\alpha + \gamma(2-\alpha)\}},$$

$$c = \frac{(1-\alpha)\gamma}{\alpha + \gamma(2-\alpha)}.$$

We would expect that the plume centreline would be more likely to be found near its mean position than in the extremities, in which case $a < b < c$, implying that

$$\alpha < \mathrm{Min}\left\{\frac{1}{2}, \gamma\right\}. \tag{4}$$

2.3 Occurrence of Large Concentrations at Receptors

We expect that the closer a receptor is to the plume centreline, the greater the number of large concentration values it will experience. To represent this behaviour we let the rate of occurrence of large concentration values at receptor location r_k be a Gaussian function of its distance from the plume centreline. Thus

$$\lambda_{i,k} = \rho \exp\left\{-\frac{(r_k - y_i)^2}{2\sigma^2}\right\},$$

where σ and ρ are positive constants. σ is a measure of the instantaneous plume width, and ρ is the rate at which large concentrations occur at the plume centreline.

3. FITTING THE MODEL TO EXPERIMENTAL DATA

The experimental data used were from the point source releases described by Mole and Jones [1994]. The model was fitted to the cases with 4 receptors in a crosswind line, namely experiments 8, 9, 10 and 11. These were all conducted under unstable atmospheric conditions, over flat terrain. The distance between the extreme receptors was 6m, and the downwind distance x from the source varied from 7.5m to 15m in the different experiments. The duration of these experiments was roughly 40–50 minutes each, with a sampling frequency of 10Hz.

The possible plume centreline positions $y_1, \ldots, y_5$ (with constant separation Δy) were chosen by reference to the number of exceedances, at the receptors, of a high concentration threshold. The aim was for y_3 to be close to the mean centreline, and for y_1 and y_5 to be towards the extremes of the plume meandering.

The concentration exceedances were "declustered" to remove dependence between them, using the method described in Mole et al. [1995]. The number of observed exceedances M, their times $\{t_j\}$ and receptor locations $\{r_{k_j}\}$ were

Table 1. Fitted values of α, β, γ, ρ and σ. x is the distance downwind of the source, and Δy is the chosen distance between possible plume centreline directions.

x (m)	Experiment	Δy (m)	α	β (Hz)	γ	ρ (Hz)	σ (m)
7.5	11	3.00	0.00405	528	0.00282	0.138	0.694
10	10	3.03	0.00000	1.27	3.50	0.0994	2.55
12.5	8	2.73	0.385	0.0203	0.371	0.0386	2.43
15	9	2.97	0.971	1.54	0.331	0.0694	1.29

Table 2. Fitted values of α, β, γ, ρ and σ in Experiment 10 (10m downwind of the source), for different choices of the distance between possible plume centreline directions, Δy.

Δy (m)	α	β (Hz)	γ	ρ (Hz)	σ (m)
4.00	0.00000	1.27	1.84	0.0928	3.30
3.03	0.00000	1.27	3.50	0.0994	2.55
3.00	0.489	0.0480	0.0959	0.0450	2.77
2.50	0.455	0.0596	0.0627	0.0400	2.94
2.00	0.438	0.0844	0.0325	0.0350	3.22
1.50	0.148	0.510	0.00236	0.0313	3.65

then used in the likelihood $\mathfrak{L}$ given by (2), and the model parameters α, β, γ, ρ and σ were estimated by maximising $\mathfrak{L}$. Some details of the numerical calculation of $\mathfrak{L}$ are given in the appendix.

4. RESULTS AND DISCUSSION

Table 1 shows the results of fitting the model to the experimental data. The plume width σ was of order 1m in all cases, consistent with physical expectation. The rate of occurrence of large concentrations on the centreline, ρ, was of order 0.1Hz or slightly less. The largest frequencies of observed exceedances over all the receptors were about 0.02–0.03Hz. This is consistent with the fitted values of ρ, since a fixed receptor will not always be close to the instantaneous plume centreline.

The fitted values of α, β and γ, which govern the plume meandering in the model, seem less satisfactory, since they show large variations between the experiments. The values of α, β and γ were also very sensitive to the chosen value of Δy, as is illustrated in Table 2 (see especially the change from $\Delta y = 3.03$m to $\Delta y = 3.00$m). The larger values of β in Tables 1 and 2 correspond to unrealistically large translation speeds for the plume centreline. Furthermore, in the cases when the values of β seem realistic, α and γ do not satisfy the expected constraint (4).

Part of the problem with the results for α, β and γ seems to arise because the likelihood surface has too many peaks of statistically insignificant height difference. This may well be because the datasets do not have enough receptor locations to resolve plume meandering with much confidence. This could be remedied by using experimental datasets obtained by Lidar [see, for example, Jørgensen et al. 1997; Nielsen et al. 1997; Bennett and Doocey 2001], in which case there can be several hundred receptor locations. A disadvantage of using Lidar data would be that the spatial resolution is only of order 1m in the crosswind direction (compared with of order 1cm for the data used in this paper), so that large concentration values are much less well resolved.

A major drawback to the part of the model dealing with plume meandering is that we used only $n = 5$ possible positions for the plume centreline. This is unlikely to be sufficient to model plume movement successfully. However, calculating the likelihood $\mathfrak{L}$ involves evaluating the exponential of an $n \times n$ matrix, which quickly

becomes computationally prohibitive as n is increased. An alternative which could make larger values of n feasible is to use Markov chain Monte Carlo (MCMC) methods [Gilks et al., 1996]. With larger values of n the model for the matrix Q would also have to be designed carefully to avoid proliferation of parameters. A different approach which could avoid this difficulty would be to model the plume movement with a process which has a continuous state space of possible positions, for example a diffusion process.

5. ACKNOWLEDGMENTS

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7. APPENDIX

Two aspects of the numerical calculation of the likelihood $\mathfrak{L}$ are worth commenting on:

Firstly, in calculating the matrix product in (2), we used the following algorithm to avoid overflow or underflow:

$$p = 0, \qquad V = \pi$$

for $j = 1$ to M

$$V = VH(t_j)\Lambda^{(k_j)}$$

$$v = \sum_{i=1}^{n} V_i$$

$$V = v^{-1}V$$

$$p = p + \ln v$$

end

$$\mathfrak{L} = e^{p}Vu.$$

Secondly, the exponential matrix $e^{(Q-\Lambda)t}$ in (3) was calculated by utilising the fact that $(Q-\Lambda)t$ is tridiagonal. MAPLE was used to produce FORTRAN code to calculate the exponential of a tridiagonal matrix, and this code was then used in the calculation of the likelihood $\mathfrak{L}$.