Comments on the properties and uses of atmospheric dispersion datasets (*)

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Summary. — Great recent improvements in the quality and quantity of atmospheric dispersion datasets have highlighted the crucial importance of concentration fluctuations. However, this has inevitably been accompanied by the realisation that estimating the properties of concentration fluctuations accurately involves new, difficult, but interesting, research problems. Some of these problems are discussed and illustrated. The paper concludes with some recommendations about how research funding agencies (such as governments, regulatory authorities and industry) should change their present strategy in response to new knowledge.

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1. – Introduction

This brief paper considers some aspects of atmospheric dispersion datasets, by which is meant, more particularly, time series of the measured concentration of a pollutant dispersing in the atmosphere. There will first be a discussion of some important features of such time series, and the paper will conclude with some comments about the uses to which they should properly be put. Reference can be made to papers by Britter [1], Hanna [2], Mole, Chatwin and Sullivan [3], and Chatwin and Sullivan [4], where some themes of the present paper are developed in other directions.

There can be no doubt that over the last 10-15 years both the quality and the quantity of available data have increased enormously. The improvements in data quality are principally due to great progress in instrument technology resulting in

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significantly enhanced spatial and temporal resolution, and the increase in data quantity has largely arisen through the widespread use in data acquisition systems of more and more powerful computers. Both changes were stimulated, at least partly, by the overdue recognition of the scientific and practical importance of fluctuations in the concentration Γ about its mean μ.

2. – Concentration time series

2˙1. *General properties.* – All time series now show that the magnitudes of the perceived fluctuations about the mean μ are not small, and are often large. Examples from instantaneous releases in wind tunnels are discussed by Chatwin and Robinson [5], and two typical examples from statistically steady releases in field trials are shown in fig. 1, using data discussed in depth by Lewis and Chatwin [6]. Such observations have stimulated theoretical work on models for risk assessment (both toxics and flammables) that explicitly incorporate concentration fluctuations (*e.g.* Griffiths [7], Ride [8]); a firm conclusion of all such work is that models of risk assessment that are based solely on the mean concentration μ will usually be seriously wrong—and such models are regrettably still common.

However, in developing models that incorporate concentration fluctuations, it is important to ensure that measurements of concentration are as reliable as possible. Recent work has highlighted interesting questions on this topic, many of which have not, arguably, received the attention they warrant.

2˙2. *Instrument smoothing and time averaging.* – Important dynamics of the concentration field occurs at spatial scales of order 10^{-4} m (the conduction cut-off length), and even the best modern instruments are unable to resolve accurately at such scales. More mundanely, many commercially available instruments do not, apparently, even recognise that a resolution problem exists.

This is well illustrated by fig. 2. In field trials in 1990 sponsored by the European Union (EU) (Project BA; see the summary by Nielsen [9] from which the data were taken), two different sensor systems were used to obtain measurements of Γ (which was, in this case, the concentration of propane gas). One system involved the use of simultaneous measurements of "sonic" temperature (using Kaijo Denki ultrasonic anemometers) and "true" temperature (using adjacent fast response thermocouples); only the former of these is affected by the presence of propane, hence by the value of Γ. Both temperatures were recorded at 10 Hz, and two different calculation methods were used to estimate Γ from these records. These two methods gave the results shown in figs. 2(a) and (b), and these two "fast" measurements were averaged to give "slow sonic" estimates of Γ recorded at 1.25 Hz and illustrated in fig. 2(c). The purpose of this averaging was to give results that could be compared with those from the second sensor system, namely a catalytic type gas detector (collocated as closely as possible with the instruments that measured temperature) whose results were recorded at 1.25 Hz and are shown in fig. 2(d). It is clear that there are great differences between the four estimates of Γ and these differences are emphasized further by fig. 3 and in table I.

Figure 4 shows a more recent example of the same phenomenon, but with different instruments. The fast time series in fig. 4(a) is taken with a UVIC sensor (also used to obtain the data in fig. 1). Apart from the slow response, the Polytron trace in fig. 4(b)

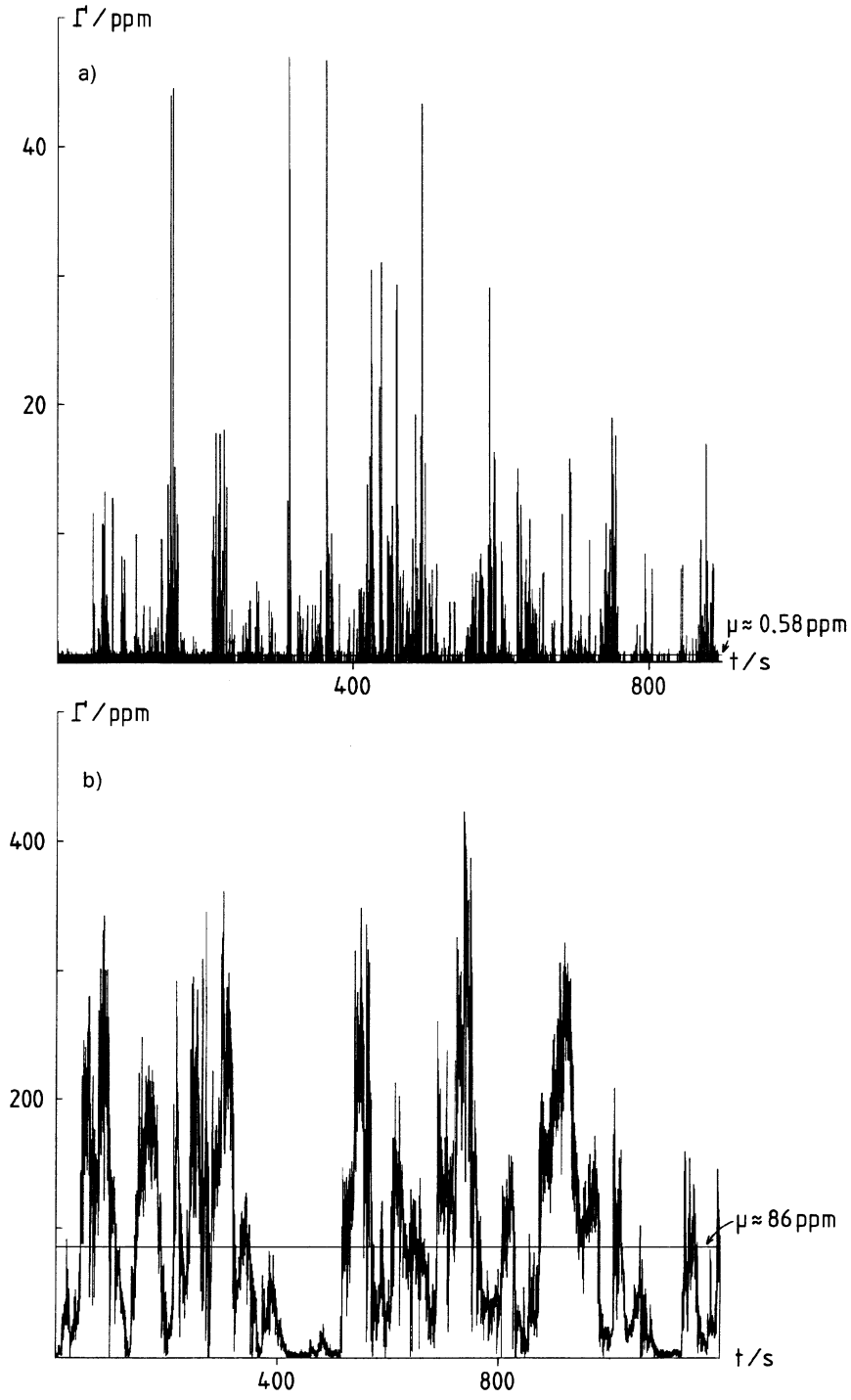

Fig. 1. – Two typical time series from statistically steady releases in field trials. a) Dugway, Utah, November 1992; b) Landskrona, Sweden, August 1993 (FLADIS project).

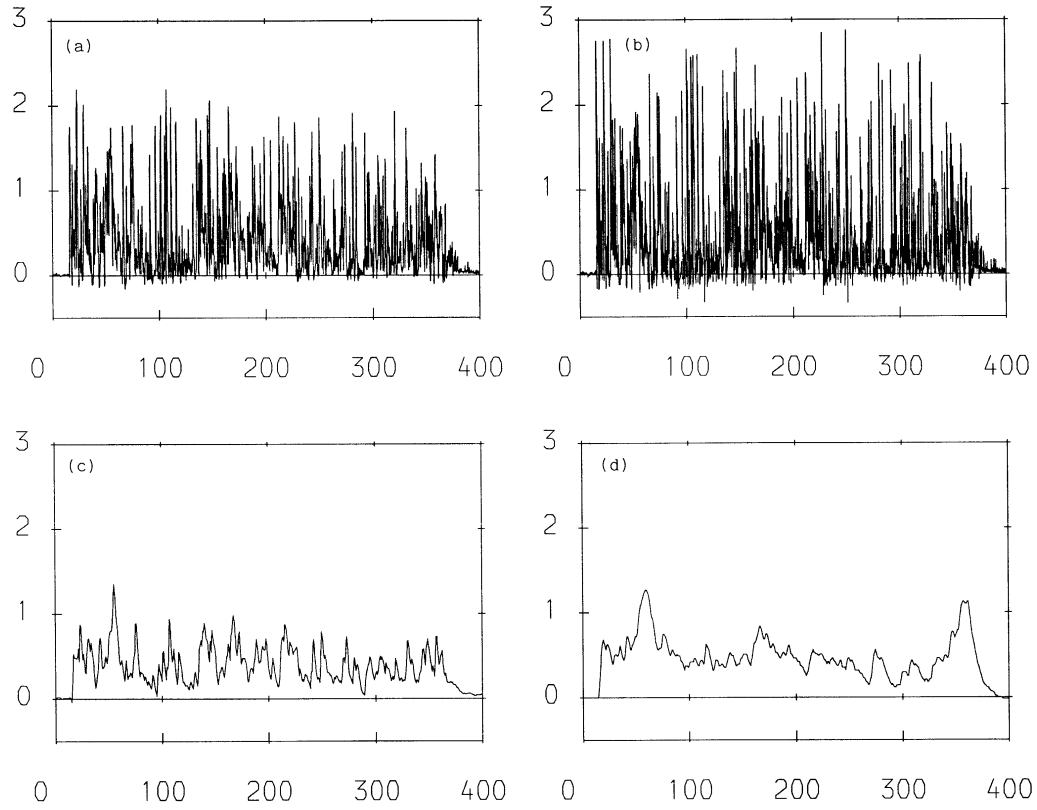

Fig. 2. – Four measurements of the time series of Γ at the same location: (a) fast sonic: method I; (b) fast sonic: method II; (c) slow sonic (1.25 Hz); (d) catalytic.

shows the further complication of baseline drift at low times. Reference may be made to Mylne and Mason [10] and Lewis and Chatwin [11] for further consideration of this phenomenon.

The time-averaged data in figs. 2(c) and 4(c) were obtained for purposes of scientific comparison, and this is also true for fig. 5, drawn in 1990 by Dr J. K. W. Davies of HSE as part of Project BA using data taken in the laboratories of the University of Hamburg by Professor M. Schatzmann. This figure shows the very significant effect of averaging period on the estimated intensity of the concentration fluctuations.

Finally in this sub-section, fig. 6 and table II show a marked effect of spatial averaging alone on σ, the standard deviation of the perceived concentration fluctuations. This work is by Sakai *et al.* [12], and shows an increase in σ as probe size decreases, and hence as the ability to resolve small scales increases. (It is noteworthy that the dimension of the smallest probe is of the same order as the conduction cut-off length.) Four different sets of measurements of σ are shown, and the plots confirm that in all cases the data satisfy the proposal by Chatwin and Sullivan [13] that

$$\sigma^2 = \beta^2 \mu(\alpha\mu_0 - \mu), \tag{1}$$

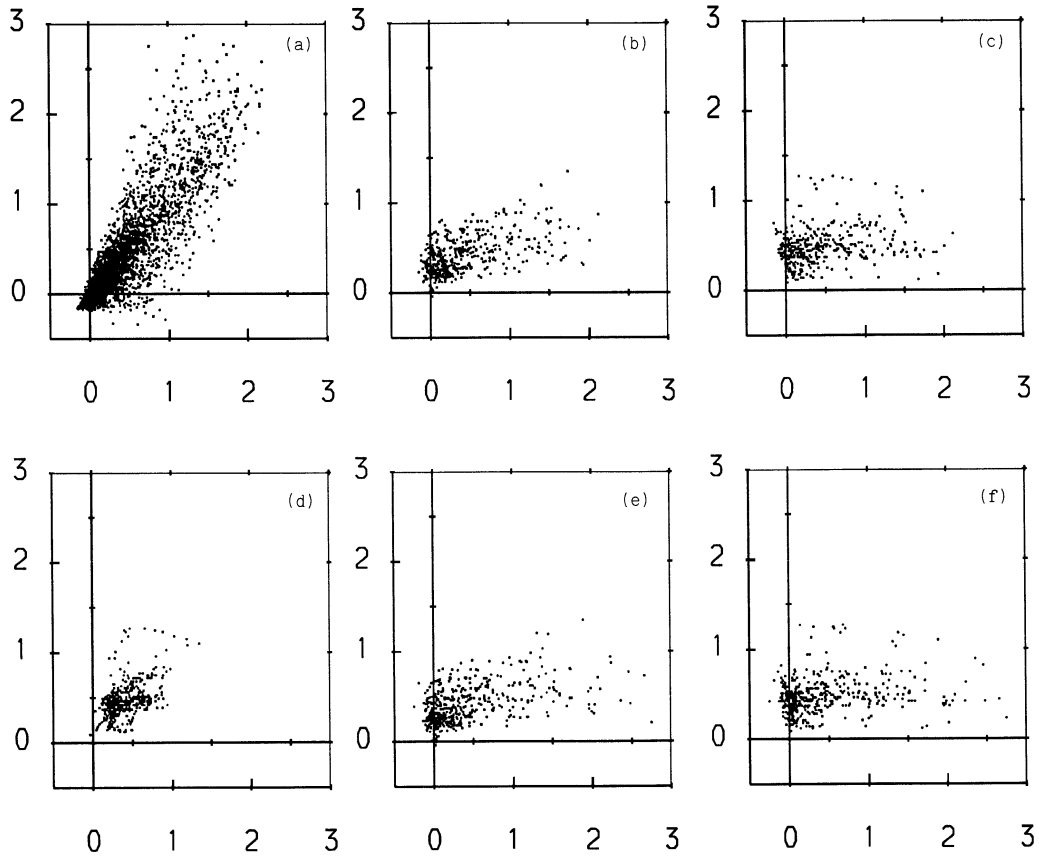

Fig. 3. – The six possible pairwise comparisons of the results in fig. 2. In detail the plots are: (a) x = fast sonic, method I, y = fast sonic, method II; (b): x = fast sonic, method I, y = slow sonic; (c) x = fast sonic, method I, y = catalytic; (d) x = slow sonic, y = catalytic; (e) x = fast sonic, method II, y = slow sonic; (f) x = fast sonic, method II, y = catalytic. If the results were identical, each of the plots would collapse onto the line $y = x$.

TABLE I. – *Some numerical results for the plots in fig.* 3. *r is the sample correlation coefficient, and* s_x, s_y *are the sample standard deviations* (rms *concentration fluctuation*). *Perfect agreement would give* $r = 1$, *and* $s_y/s_x = 1$, *in each case.*

Figure	x	y	r	s_y/s_x
3(a)	FS I	FS II	0.865	1.192
3(b)	FS I	SS	0.536	0.453
3(c)	FS I	C	0.225	0.481
3(d)	SS	C	0.452	1.063
3(e)	FS II	SS	0.461	0.375
3(f)	FS II	C	0.185	0.398

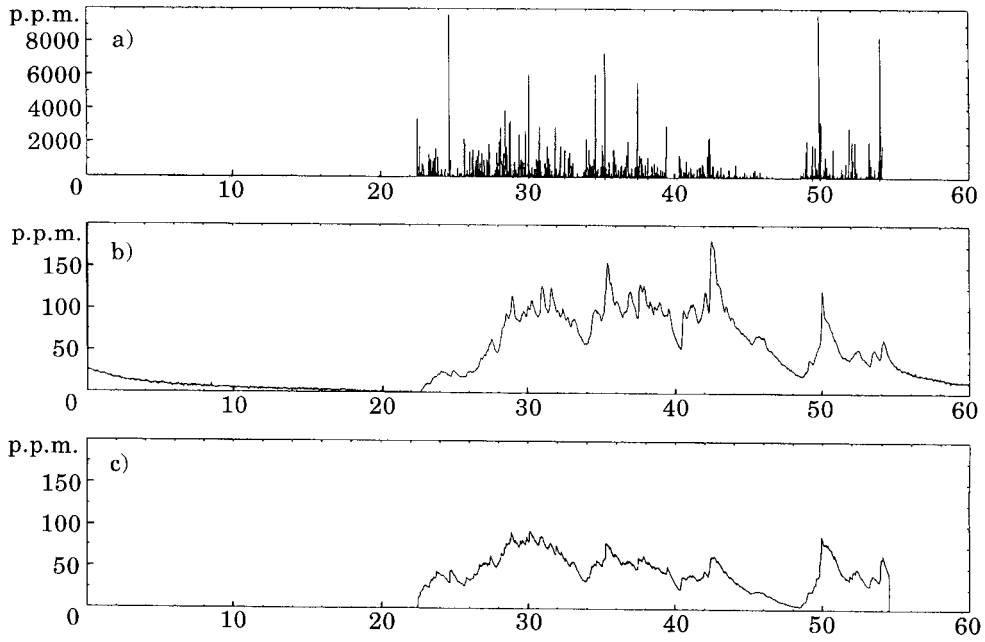

Fig. 4. – Three measurements of the time series of Γ at the same location; a) fast UVIC; b) Polytron B; c) the series in a) filtered to be comparable with 4b). The abscissa is time in minutes.

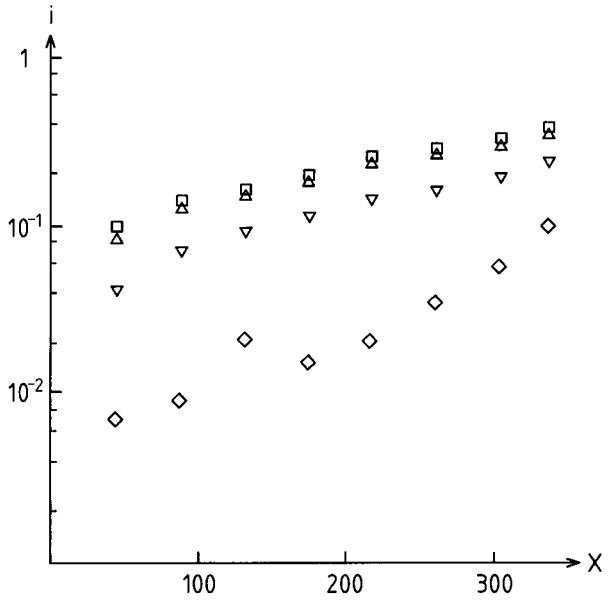

Fig. 5. – The effect of imposed averaging time T on i, the estimated intensity of concentration fluctuations (i is estimated σ/estimated μ); X is distance downwind from a steady source. □ $T = 0.07$ s; △ $T = 0.7$ s; ▽ $T = 7$ s; ◇ $T = 70$ s.

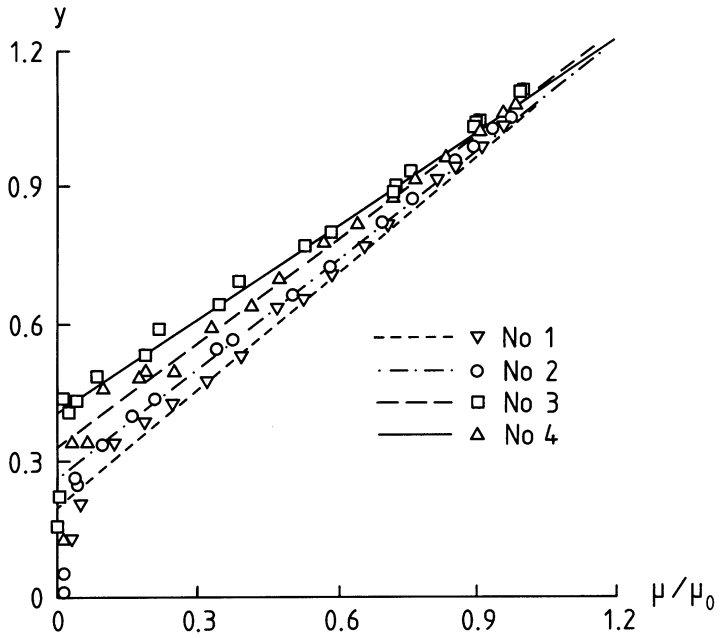

Fig. 6. – Data from Sakai *et al* [12]. Here $y = (\sigma^2 + \mu^2)/(\mu\mu_0)$, and rearrangement of eq. (1) gives the straight line: $y = \alpha\beta^2 + (1 - \beta^2)(\mu/\mu_0)$.

TABLE II. – *Values of α and β for the plots in fig. 6, using the formula in eq. (1). The constant d is the cube root of the sample volume measured by the probe.*

Probe No.	*d*/mm	α	β
1	0.54	1.31	0.40
2	0.30	1.25	0.47
3	0.13	1.33	0.50
4	0.10	1.25	0.57

where μ_0 is a scale for μ, and α and β are constants. For present purposes, the significant finding is that β increases as probe size *d* decreases (consistent with a suggestion in [13]). Further details are given by Nakamura, Sakai and Miyata [14] and by Chatwin and Sullivan [15].

2·3. *Effects of noise and thresholding.* – All time series of concentration are affected by inevitable random noise. The most striking consequence of this is that some recorded concentrations are negative; for example, this is evident in figs. 2(a) and (b). Many concentration datasets are dominated by frequent occurrences of very low concentrations, when this consequence of noise will be particularly significant.

A severe example is shown in fig. 7(a), where a concentration of zero corresponds to 0.2 V. This example is discussed in Lewis and Chatwin [11] to which reference should be made for full details of the summary that follows. After pre-processing (whose purpose was to reduce substantially the effect of instrument smoothing discussed

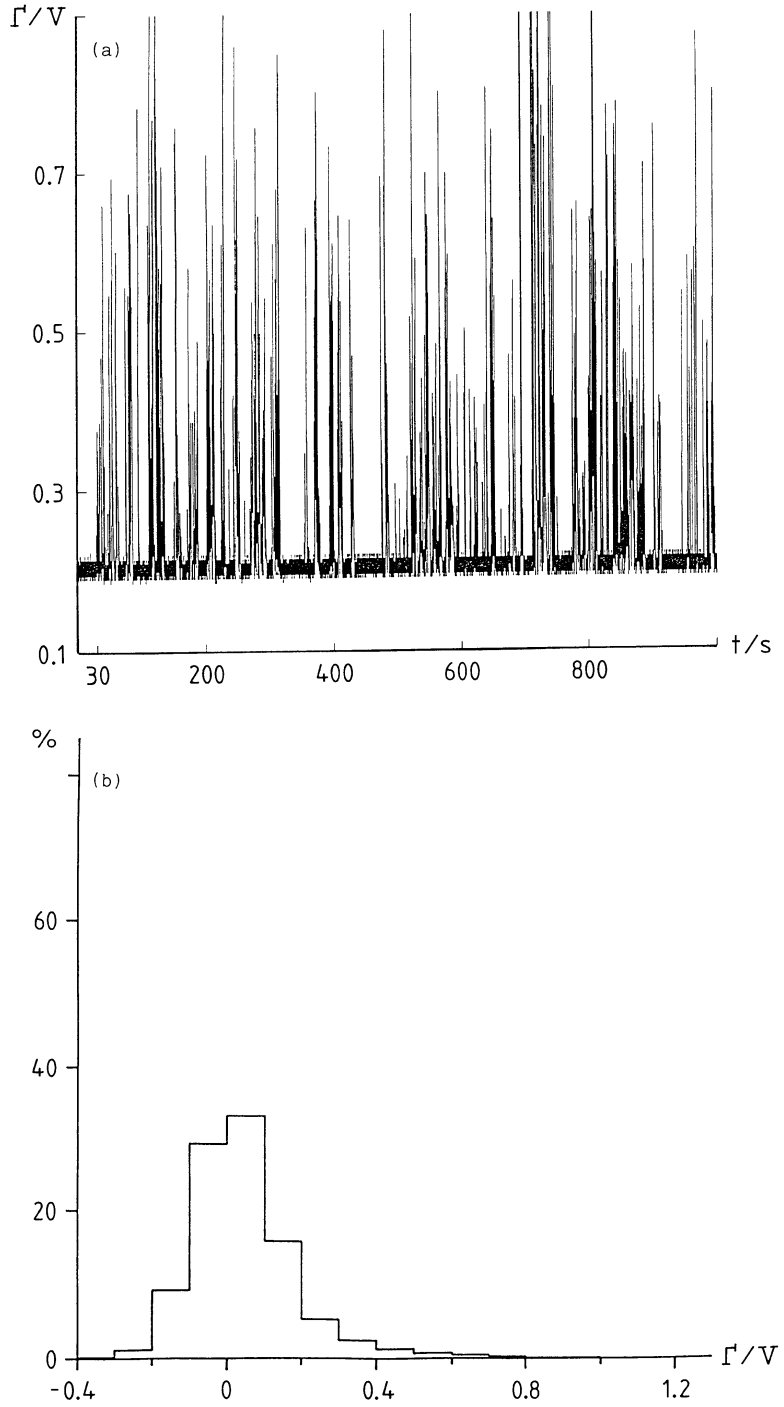

Fig. 7. – (a) Data taken by Dr C. D. Jones at Dugway, Utah with a UVIC Mark I sensor. The first 30 s of this record were used to estimate noise properties. (b) The histogram of the data in (a) after pre-processing.

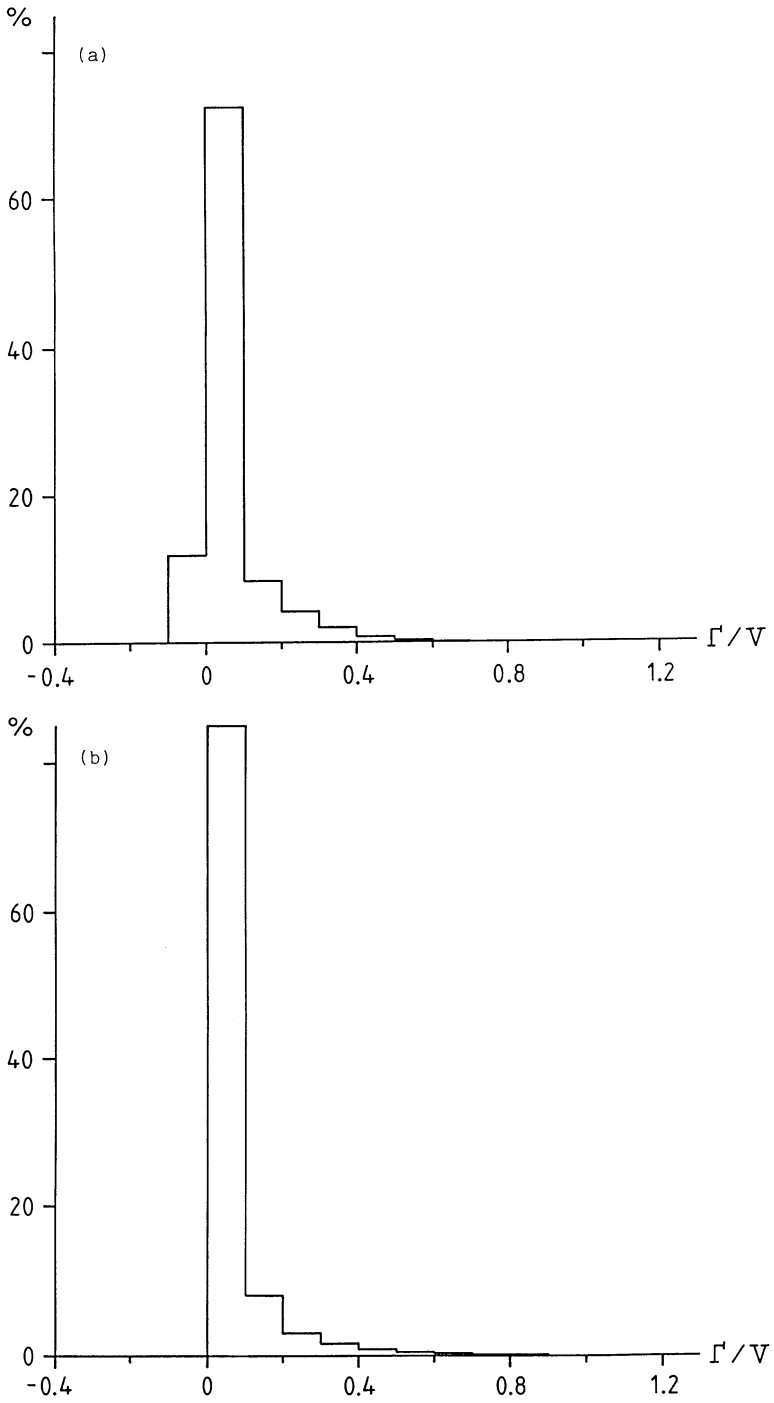

Fig. 8. – Estimated "true" histograms of Γ for the data shown in fig. 7 after applying two different noise deconvolution methods. (a) Wiener filter; (b) MEI.

above—see Mole [16]), the histogram derived from fig. 7(a) is shown in fig. 7(b); due to noise almost 40% of the perceived concentrations are negative!

Clearly such data should not be used as they stand to analyse concentration fluctuations, and it is appropriate to draw attention to some recent work on this problem. Mole [16] discusses methods of removing noise in general, with emphasis on Wiener filter methods. The Wiener filter has the important theoretical advantage of minimising the mean-square difference between the recorded and true concentrations, but does not ensure that negative estimates of concentration do not remain after its use. An application of a Wiener filter method to the data shown in fig. 7 results in the histogram shown in fig. 8(a). Although the percentage of estimated concentrations that are negative has been substantially reduced from nearly 40% to about 12%, the latter figure is still uncomfortably high. A different technique which does ensure that all estimated concentrations are non-negative is MEI (Maximum Entropy Inversion); see Lewis and Chatwin [11]. In brief, this technique maximizes an entropy function to obtain estimates of the real concentrations, where there are elementary constraints applied that are based on characteristics of the data. The entropy function contains a logarithmic term that guarantees the recovery of positive estimates of concentrations from the data. Applied to the data in fig. 7, one version of MEI gives the histogram shown in fig. 8(b). Note that both techniques are applied so as to preserve the measured value of μ, the mean concentration, but do give different estimates for the value of σ.

These methods involve choice about, for example, how to estimate the noise spectrum and, for any one dataset, there is no independent criterion for assessing whether either is acceptable or, assuming both are, which is the better. Therefore, the techniques were applied to some time series generated artificially, and then corrupted by random noise. Table III, adapted from results given in Lewis and Chatwin [11],

TABLE III. – *Comparison of three methods for removing/reducing noise applied to two artificial datasets corrupted by the addition of random noise. See* [11]. *Upper part*: *raw data from gamma distribution. Signal-to-noise ratio* 10.12. *Lower part*: *raw data from lognormal distribution. Signal-to-noise ratio* 3.42. (*The signal-to-noise ratio is here defined as the variance of the corrupted signal divided by the variance of the noise.*)

Properties of datasets	Raw	Corrupted (raw plus noise)	Wiener	MEI	Thresholding
mean μ	1.00	1.00	1.00	1.00	1.01
S.D. σ	0.76	0.80	0.74	0.75	0.79
skewness	1.94	1.67	1.93	1.84	1.73
kurtosis	8.69	7.66	8.75	8.34	7.82
% of negative concentrations	0	3.55	0.73	0	0
mean μ	0.50	0.50	0.50	0.50	0.59
S.D. σ	0.78	0.93	0.75	0.76	0.86
skewness	6.83	4.03	5.60	6.30	5.29
kurtosis	96.46	48.38	62.80	89.14	67.22
% of negative concentrations	0	26.25	8.30	0	0

summarizes some results of applying the techniques discussed above. In brief, the results of the MEI method appear to be more robust. However, for the data in table III, upper part, with the larger signal-to-noise ratio, the Wiener filter method gives somewhat better estimates of the moments than MEI, but this is offset by the fact that about 0.73% of the estimates are negative.

Also shown in table III are results obtained by thresholding, an old (but non-scientific) technique in which all measured values of concentration Γ below an arbitrarily chosen and subjective threshold value θ_T are set equal to zero. For the data in table III, this method is obviously inferior to either of the other two methods, and some even more striking examples are given by Robinson, Lewis and Chatwin [17].

(It should be noted that neither of the time series shown above in fig. 1 displays negative values of Γ because the MEI method has already been applied.)

2·4. *Intermittency factor.* – In some work on atmospheric dispersion, it has been traditional to use the concept of the intermittency factor $p = p(\mathbf{x}, t)$, where $\mathbf{x}$ and t denote position and time, respectively. The conventional definition is

$$p = \text{prob}\{\Gamma(\mathbf{x}, t) > 0\}, \tag{2}$$

but see below. Use of this definition is however complicated by the presence of noise (and incidentally by binning) and, in practice, (2) has usually been replaced by

$$p = \text{prob}\{\Gamma(\mathbf{x}, t) > \theta_T\}, \tag{3}$$

where θ_T is a positive threshold value of concentration with the disadvantages stated above. It is therefore not surprising that a dedicated examination shows that use of

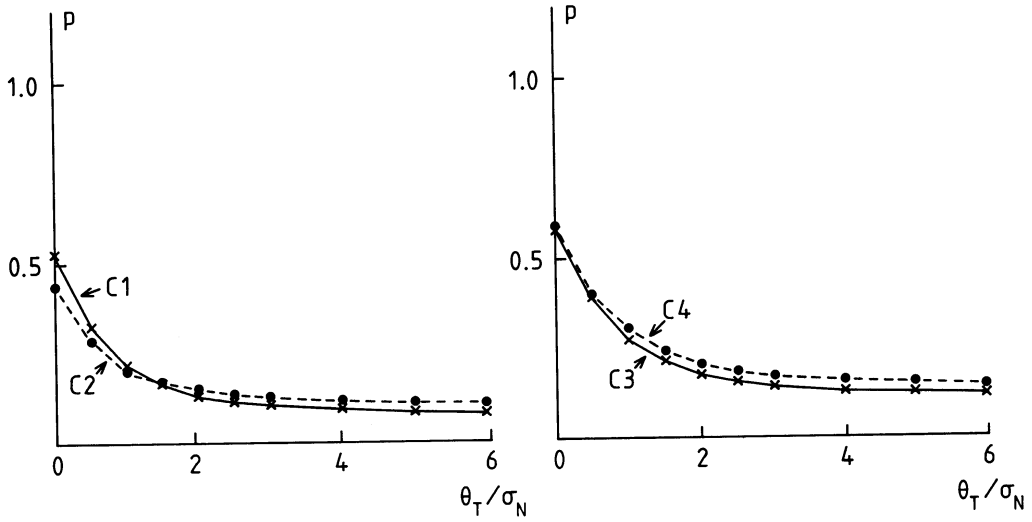

Fig. 9. – Variation of the intermittency factor p, defined in eq. (3), with threshold concentration θ_T; σ_N is the standard deviation of the noise. The experiment (ASL 10) was part of a series conducted by Dr C. D. Jones in White Sands, New Mexico in 1989 using an ion collector sensor with a steady source. C1-C4 are the separate results from 4 ion collectors.

(3) gives values of p sensitively dependent on θ_T This is illustrated in fig. 9, taken from Mole and Chatwin [18], where many similar examples can be found.

The conclusion once again is that the use of a threshold concentration is unacceptable, essentially because it has no connection with real concentrations.

It must be noted that there are also, however, fundamental objections to the definition (2), and these are discussed by Chatwin and Sullivan [19] where an alternative definition is proposed.

2·5. *Stationarity and length of record.* – Although the factors mentioned in this sub-section have hardly received study, they are included because such study would certainly appear to be merited. Since atmospheric turbulence is subject to systematic trends because the weather is changing (on scales of hours at most), so also will the statistical properties of Γ change with t on the same scale, even when the pollutant is being released at a steady rate. This factor is usually ignored, *i.e.* a time series from a steady source is regarded as a realisation of a stationary process. However the graphs in fig. 10, also taken from Mole and Chatwin [18], show that estimates of some elementary statistical properties made on this assumption show significant trends over a period of 50 minutes. Anderson, Mole and Nadarajah [20] also find significant trends

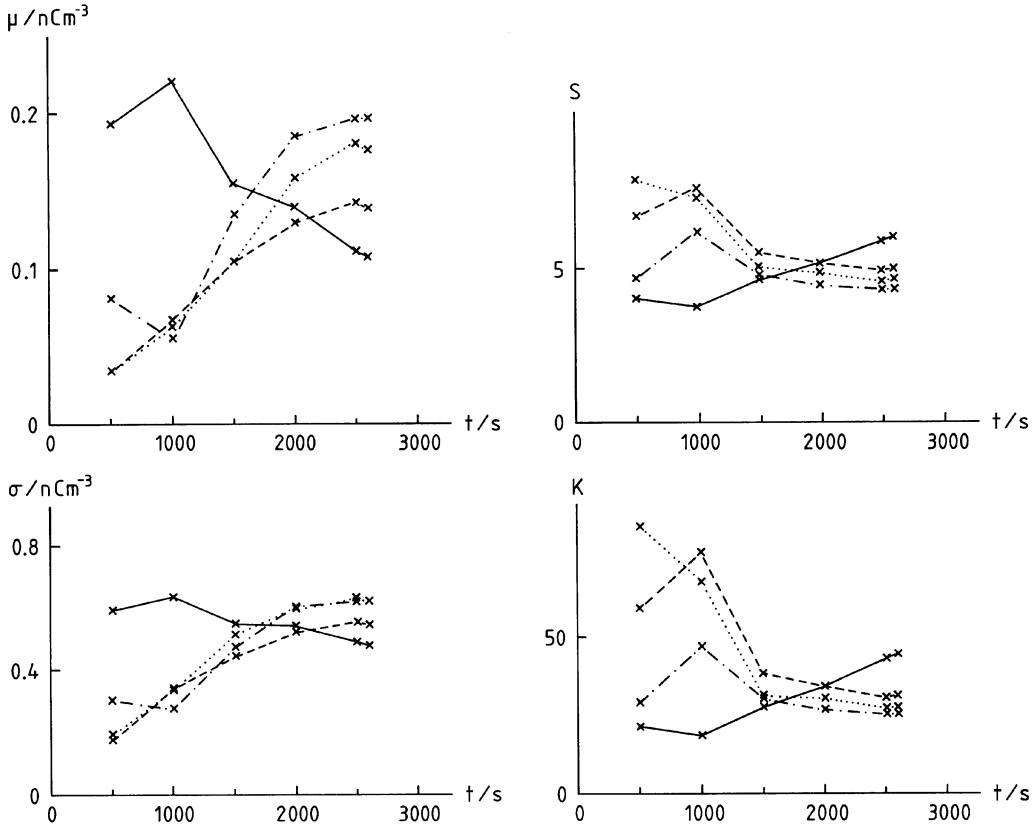

Fig. 10. – Estimates of μ, σ, skewness S and kurtosis K as a function of *accumulated* time t for ASL 10 (see caption to fig. 9). —— C1; - - - - - C2; ····· C3; –·–·– C4.

in the parameters of a model to estimate the frequencies of occurrence of large concentrations.

But, by contrast, a sufficient length of record (depending on the digitisation rate) is needed even in strictly stationary conditions to obtain stable estimates of such statistical properties. (The term "stable" means that the inevitable statistical noise is acceptably small.) Thus quantitative work is needed on these conflicting problems.

For instantaneous releases, considered in Chatwin and Robinson [5] but not elsewhere in this paper, the requirement of length of record is no longer relevant but is replaced by the need for an adequate number of replications.

3. – Conclusions

An overall conclusion of the previous section must be that data analysis techniques have not yet adjusted appropriately to the recent improvements in data quality and quantity. In what was written above there was emphasis on quality, but the increase in quantity has also brought problems. To the authors' knowledge, there are large and potentially valuable datasets available which have not been (and may never be) analysed. The principal reason is that the resources needed (above all staff time) were not adequately funded at the time the experiments themselves were funded. Ultimately this is wasteful. A necessary condition for improvement is that:

> *Experiments, especially large experiments, must now be planned with data analysis and modellers as full members of all teams from the outset.*

Although it is now indisputable that proper consideration of concentration fluctuations is essential for the satisfactory evaluation of risk associated with atmospheric dispersion (often following accidental release), recognition of this fact by governments and regulatory authorities (such as the EU) is still largely absent. It is certainly overdue. Therefore

> *Those scientists working in atmospheric dispersion should, at every opportunity, emphasize the crucial importance of concentration fluctuations in models of associated risk assessment. Since no-one other than scientists can do this, it could be regarded as a duty.*

Unfortunately governments and regulatory authorities have responded to the advent of vastly increased computing power by funding almost exclusively research in modelling that develops old and out-of-date models, making them more sophisticated and complicated. Thus they ignore the abundant evidence that

> *Many, if not most, models of atmospheric dispersion and associated risk assessment now used by governments and regulatory authorities are, quite simply, wrong, both scientifically and practically, since they place undue emphasis on mean concentrations. Little or no recognition is given to fluctuations.*

A different objection to making mathematical models more complicated is technical and less fundamental, but important nonetheless. It is made clear in quotes from two papers referred to earlier, namely *a*) "In a broad sense, there is generally an

over-confidence in the ability of mathematical models to provide accurate prediction. In particular, there is significant over-confidence in the ability of very large numerical models to provide accurate prediction" (Britter [1]), and *b*) The performance of a model is "not related to its cost or complexity" and, in general, "it is very difficult to demonstrate improved model performance as enhancements in model physics are added" (Hanna [2]).

Consistent with all that has been written above, the correct response to improvements in data quality and quantity, and the consequent new knowledge should be different. In summary

> *Research and development funding strategy should now be devoted to developing new, scientifically correct, models, and to testing these against good data. Such models must be probabilistic, and should be simple.*

4. – Further information

Further details of the two multinational EU projects referred to extensively in this paper are given in Chatwin and Goodall [21] (Project BA; First Framework Programme, Major Technological Hazards), and in Chatwin *et al.* [22] (Project FLADIS; Second Framework Programme, Major Industrial Hazards), as well as in many of the other papers cited above. See also Chatwin, Lewis and Mole [23].

* * *

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REFERENCES

[1] BRITTER R. E., *The evaluation of technical models used for major-accident hazard installations,* Report to Commission of the European Communities (DG X11), August 1991.
[2] HANNA S. R., *Boundary-Layer Meteorol.*, **62** (1993) 3.
[3] MOLE N., CHATWIN P. C. and SULLIVAN P. J., in *Modelling Change in Environmental Systems*, edited by A. J. JAKEMAN, M. B. BECK and M. J. MCALEER (John Wiley & Sons Ltd., Chichester) 1993, pp. 317-340.
[4] CHATWIN P. C. and SULLIVAN P. J., in *Mixing and Transport in the Environment*, edited by K. J. BEVEN, P. C. CHATWIN and J. H. MILLBANK (John Wiley & Sons Ltd., Chichester) 1994, pp. 353-369.
[5] CHATWIN P. C. and ROBINSON C., this issue, p. 361.
[6] LEWIS D. M. and CHATWIN P. C., *Environmetrics*, **6** (1995) 583.
[7] GRIFFITHS R. F., *J. Loss Prevention Industries,* **4** (1991) 49.
[8] RIDE D. J., *Environmetrics*, **6** (1995) 643.
[9] NIELSEN M., *Preliminary treatment of meteorological data from Project BA dense gas experiments*, Report No. Risø-M-2882, Risø National Laboratory, Roskilde, Denmark, July 1990.
[10] MYLNE K. R. and MASON P. J., *Q. J. R. Meteorol. Soc.,* **117** (1991) 177.

[11] LEWIS D. M. and CHATWIN P. C., *Boundary-Layer Meteorol.*, **72** (1995) 53.
[12] SAKAI Y., NAKAMURA I., TSUNODA H. and SHENGIAN L., Private communication and lecture presented at Euromech 253, Brunel University, Uxbridge, UK, August 1989.
[13] CHATWIN P. C. and SULLIVAN P. J., *J. Fluid Mech.*, **212** (1990) 533.
[14] NAKAMURA I., SAKAI Y. and MIYATA M., *J. Fluid Mech.*, **178** (1987) 379.
[15] CHATWIN P. C. and SULLIVAN P. J., *Boundary-Layer Meteorol.*, **62** (1993) 269.
[16] MOLE N., *Environmetrics*, **1** (1990) 179.
[17] ROBINSON C., LEWIS D. M. and CHATWIN P. C., *Boundary-Layer Meteorol.*, **73** (1995) 183.
[18] MOLE N. and CHATWIN P. C., *Fluctuations in atmospheric contaminants*, Technical Report No. TR/16/90, Department of Mathematics and Statistics, Brunel University, Uxbridge, UK, October 1990.
[19] CHATWIN P. C. and SULLIVAN P. J., *Phys. Fluids A*, **1** (1989) 761.
[20] ANDERSON C. W., MOLE N. and NADARAJAH S., *Atmos. Environ.*, **31** (1997) 813.
[21] CHATWIN P. C. and GOODALL G. W., *Research on continuous and instantaneous heavy gas clouds*, Technical Report No. TR/02/91, Department of Mathematics and Statistics, Brunel University, Uxbridge, UK, February 1991.
[22] CHATWIN P. C., LEWIS D. M., ROBINSON C., SWEATMAN W. L. and ZIMMERMAN W. B., *Natural variability in atmospheric dispersion: theory and data comparison*, Report No. TR/03/94, Applied Mathematics Section, SoMaS, University of Sheffield, UK, December 1994.
[23] CHATWIN P. C., LEWIS D. M. and MOLE N., *Adv. Comp. Maths.*, **6** (1996) 227. (John Crank 80th Anniversary issue.)