

LETTERS TO THE EDITOR

Comments on an article of Slinn

By R. JAENICKE, *Institut für Meteorologie, Universität Mainz, FRG*

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The discussion of the relationship between concentration fluctuations and residence times is certainly highly welcomed, because it can spread the news of how a simple analytical equation eases the often needed estimate of residence times for trace substances, gases and aerosols in size ranges. However, the treatment by Slinn (1988a) might confuse potential users.

To ease the following clarification, I will use Slinn's notation with C being a concentration and \bar{C} the mean concentration, whether averaged over the exponentially decaying concentration in time or in distance downwind of a finite-area source.

I assume that the definition of the coefficient of variation as given in the upper portion of eq. (3a) is a misprint. The second definition given in eq. (3a) states

$$F_c^2 + 1 = \frac{(\bar{C}^2)}{(\bar{C})^2} \text{ leading to } F_c = \frac{(\bar{C}^2 - \bar{C}^2)^{1/2}}{\bar{C}}.$$

A third way of presenting the coefficient of deviation can be seen on the ordinate of Fig. 3:

$$F_c = \frac{\sigma_c}{\bar{C}},$$

with σ_c = standard deviation, which Slinn introduces as Σ in the insert of Fig. 2 to increase the confusion even more.

In the rest of the discussion, Slinn uses $F_c = (\bar{C}^2 - \bar{C}^2)^{1/2}/\bar{C}$, as what he believes to be a peculiar definition of the coefficient of deviation, and claims that I have not noticed that in Junge's (1974) paper.

I believe we agree in the definition of the empirical variance of a sample

$$\sigma_c^2 = \frac{1}{n} \sum_{i=1}^n (C_i - \bar{C})^2,$$

with

n = number of elements of the sample,

C_i = individual concentrations,

\bar{C} = mean concentration as

$$\bar{C} = \frac{1}{n} \sum_{i=1}^n C_i.$$

From this equation, a second one develops easily. There is no difference in the result, if we carry out this discussion in terms of sum or terms of integral. Notice the distinction between \bar{C}^2 and \bar{C}^2 ,

$$\sigma_c^2 = \frac{1}{n} \sum_{i=1}^n (C_i^2 - 2C_i\bar{C} + \bar{C}^2).$$

As we have defined \bar{C} , we define

$$\bar{C}^2 = \frac{1}{n} \sum_{i=1}^n C_i^2$$

and it follows that $\sigma_c^2 = \bar{C}^2 - \bar{C}^2$ as variance, and $\sigma_c = (\bar{C}^2 - \bar{C}^2)^{1/2}$ as standard deviation. As coefficients of deviation, we have $F_c = \sigma_c/\bar{C}$ and consequently $F_c = (\bar{C}^2 - \bar{C}^2)^{1/2}/\bar{C}$ with nothing peculiar about it.

Just to remove the confusion, Slinn's eq. (3b) must read correctly

$$F_c^2 + 1 = \frac{\Delta s}{2T_r} \frac{1 - \exp(-2\Delta s/T_r)}{(1 - \exp(-\Delta s/T_r))^2}.$$

Slinn has interpreted two "explanations" in my paper (Jaenicke, 1982) as unnecessary, restrictive assumptions.

(i) The introduction of an upper time limit t_{\max} of the averaging period (what he believes because of a minimum detection limit of the sampling instrument).

(ii) Observations during this period are of equal probability.

The t_{\max} is nothing other than the Δs used by Slinn, except t_{\max} covers the time period beginning at the source. If we would permit $\Delta s \rightarrow \infty$, the result would be $\bar{C} \rightarrow 0$ and $\sigma_c \rightarrow 0$, just indicating that the mean and the standard deviation of the decaying process contain no information about the subject, because the substance under investigation has faded away. However, both values approach 0 in a different way, resulting in $F_c \rightarrow \infty$ for $\Delta s \rightarrow \infty$. So, a t_{\max} had to be introduced and is not restrictive for any practical purpose. Slinn stresses correctly the fact of Δs covering any time period during the decay process. However, in practice, the time period between 0 and x_1 cannot be excluded with certainty from calculating the moments. The problem with t_{\max} is that it has to be specified and needs verification from observations.

The second point is just what Slinn terms (eq. (2a)): "... averaged [integrated] over a ... range of sampled times". This is not an assumption, it is a direction for use. As in any integration, it is not permitted to have different densities (probabilities) in the integration range.

I agree with Slinn that in a stationary process, the coefficient of deviation should be independent of the sampling time in contrast to the equation derived from the model. This is valid, if we consider the coefficient of deviation for a sample of measurements at a single point. The modest agreement of the model with coefficients of deviation obtained from measurements (Fig. 3, Jaenicke, 1982) might indicate that the measurements are not so much influenced by stationary processes as by exponential decays.

The explanation might be that some of the trace substances have their sources confined to certain geographical areas, like the continents for the condensation nuclei of the aerosol or finite-area sources for some trace gases. Aerosol measurements over the oceans with increasing or decreasing distance from the continents are made in the decaying process. Incidentally, this is exactly how Junge (1974) designed and evaluated his numerical model. On the other hand, if we have sources randomly distributed around a recording station, the variation of the wind direction and speed with time brings this station into varying time distances to these point sources, just the way the model assumes it. As could be shown (Fig. 2, Jaenicke, 1982), the various strengths of the sources are of minor importance if the time distances downwind are a few days.

As a reminder, Hamrud's (1983) results should be used with caution to demonstrate the validity of the above model. Hamrud uses a grid of 10° or 1111 km, so air is sampled roughly every 3 or 4 days during the decay process, depending on the average wind speed. This way, the coefficient of deviation is underestimated for shorter residence times compared to the analytically derived value. A reviewer reminds us that because of this, Hamrud's (1983) model is not applied for residence times shorter than ~ 1 month. This is pointed out in that paper. Furthermore, the curve presented by Slinn from Hamrud (1983) cannot be found in the publication.

Including the ideas presented in this comment, one should reconsider the treatment of explaining the empirical data relating the concentration fluctuations to pollutant residence time in the atmosphere (Slinn, 1988b).

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