# Determination of the time when the concentration of an atmospheric pollutant exceeds a preset level

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Based on the probability density function obtained earlier for concentration of a pollutant spread in the atmosphere, we consider the problem of determining mathematical expectations of the frequency of occurrence of concentration to be above  a given  threshold level and the duration of such emissions. Calculations by  the deduced formulas well agree with the experimental data from Refs. 1 and 2.  It is shown that the "transient" and "equilibrium" regimes of concentration pulsation obtained by the formulas correspond to different values of pollutant concentration intermittence.

Some applied problems require the knowledge of such characteristics as mean value of the frequency  of occurrence of an atmospheric pollutant at the concentration level $C$ that exceeds a given threshold value $C_0$, the mean duration of such events, and the mean time between the events when the concentration exceeds a given level. The empirical estimates of these characteristics were obtained in Ref. 1 on the basis of pulsation measurements of a pollutant concentration, emitted from a stationary source, at a fixed point in space. It was demonstrated in Ref. 2 that one can observe two fluctuation regimes of the pollutant concentration in the ground layer of the atmosphere. These are  the "transition" and the "equilibrium" regimes.

The transition regime is characterized by the fact that, together with the part of spectral density of concentration pulsation that obeys the law $f \sim$ "$-5/3$" ($f$ is the pulsation frequency), one can observe the pulsation obeying the $f \sim$ "$-2/3$" law. The latter is valid at the dimensionless frequency greater than two, and the law of "five thirds" at the dimensionless frequency greater than seven.

According to the above-stated in Ref. 1, in determining the parameters of the processes leading to the  enhancement of  a given concentration level by an admixture, the experimental data from Ref. 1 have divided into two groups. It is well known that determining these characteristics requires to know the distribution laws of concentration and its derivative with respect to time.[3]  By the moment of Refs. 1 and 2 publication these characteristics were not known. So the results obtained in Ref. 1 are of empirical character and it is difficult to extrapolate them to the conditions of a pollutant spread different than those considered by the authors of these papers. However, at present the regularities of pollutant concentration distribution are studied quite well[4] and one can return to this problem formulated in a more general form.

Theoretical basis for determining the mean values of the pulsation time of random processes are developed in statistical radio engineering (see, for instance, Ref. 3).

For  stationary  random  processes,  mathematical expectation for the number of intersections of a given level $C_0$ by the concentration "from below" (with positive derivative), has the form

$$\lambda_+(C_0) = \int\limits_0^\infty C' f_2(C_0, C') \mathrm{d}C', \qquad (1)$$

where $f_2$ is the joint function of probability density of the pollutant concentration and its derivative with respect to time. In the stationary case, the values of the process and its derivative are statistically independent, so $f_2(C_0, C') = f_0(C)f_1(C')$, where $f_0$ is the probability density of the pollutant concentration $C$, and $f_1$ is probability density of its derivative $C'$ with respect to time. Mathematical expectation of the number of events when the concentration reaches the level $C_0$ "from above" (with negative derivative) is equal to the mean number of events  when it reaches this level "from below": $\lambda_-(C_0) = \lambda_+(C_0)$.  The total number of events is $\lambda(C_0) = 2\lambda_+(C_0)$. Mathematical expectations for the duration $\rho_+(C_0)$ and for intervals between the emissions $\rho_-(C_0)$ can be obtained by the formulas

$$\rho_+(C_0) = \frac{1 - F_0(C_0)}{\lambda_+(C_0)}; \quad \rho_-(C_0) = \frac{F_0(C_0)}{\lambda_+(C_0)}, \quad (2)$$

where $F_0$ is the distribution function of the pollutant concentration. It corresponds to the probability density $f_0$.

According to Ref. 4, the probability density of a pollutant concentration $f_0$ has the form

$$f_0(C) = (1 - \gamma)\delta(C) + \frac{1}{\sqrt{\pi}\beta} \times$$

$$\times \left\{ \exp\left[ -\frac{(C - \bar{C})^2}{\beta^2} \right] - \exp\left[ -\frac{(C + \bar{C})^2}{\beta^2} \right] \right\},$$

$$\gamma = \mathrm{erf}(\bar{C}/\beta), \qquad (3)$$

where $\gamma$ is concentration intermittence; $\overline{C}$ is mathematical expectation of the concentration; $\beta$ is the second parameter of the probability density (it is connected with the variance of the pollutant concentration $\sigma^2$ (see Ref. 4); $\delta(...)$ is the Dirac delta function; erf$(...)$ is the error function. The function (3) is the exact analytical solution of the Fokker–Planck–Kolmogorov equation; it is verified by data of experiments in a wind tunnel, data of independent field experiments, and corresponds to the classical asymptotic from the theory of turbulent combustion.[4]  It was found in Ref. 4 that Eq. (3) well describes   the   experimental   data   at    $\gamma > 0.1$.    At $\gamma < 0.1$,  the  effect  of  viscosity  at  the  boundaries between  the  spatial  domains  with  and  without  a pollutant cannot be neglected. For this reason, the term involving the delta function must, in the general case, be  represented  as  a  function  of  finite  width.  The empirical estimates of the effect have been attempted in Ref.  5.  The  theoretical  estimates  can  be  found  in Ref. 6.

The probability density of the derivative from the pollutant concentration $f_1$ can be obtained, like $f_0$, by solving analogous equation. However, here its form is verified on the basis of only general physical reasoning. The continuous part of the probability density of concentration is represented as a difference of two fundamental solutions to the heat equation. This fact demonstrates close connection existing between the considered problem and theory of normal Markovian diffusion processes.

It is evident that the difference between two fundamental solutions is associated with the fact that the pollutant concentration is a non-negative value. The derivative of the concentration with respect to time can take any value, including the negative ones. In contrast to concentration, it does not intermit. So it is quite natural to assume that the form of the probability density of the derivative of concentration is normal. In the general case, this function is defined by two parameters, namely, the mathematical expectation and variance. For a stationary case, the mathematical expectation of the derivative is zero, and variance is equal to the second derivative of the correlation function of derivative pulsation under the condition of zero  time  shift.[3]      For  the  dimensionality considerations, this value should equal to the ratio of the concentration variance to the squared characteristic time of concentration pulsation $\tau_e$, i.e., Eulerian time scale. Note that this reasoning is quite rigorous when applied to the correlation function  of  concentration pulsation  in the exponential form. According to the above-said, the sought probability density function must take the form

$$f_1(C') = \frac{\tau_e}{\sqrt{2\pi}\,\sigma} \exp\left[-\frac{(\tau_e\,C')^2}{2\sigma^2}\right]. \qquad (4)$$

The  following  density  of  spectral  power  of pulsation[3] corresponds to the exponential form of the correlation  function  of  concentration  pulsation $B(\tau) = \sigma^2 \exp\left(-|\tau|/\tau_e\right)$:

$$S(f) = 2\tau_e\sigma^2/[1 + (\tau_e f)^2].$$

In our paper "Correlation function of pulsation..." (see Atmos. Oceanic. Opt. No. 8, in press), the Figure presents the logarithm of $f\,S(f)/\tau_e\,\sigma^2$ as a function of the logarithm of the dimensionless frequency $\tau_e f$.  One can see that for $\tau_e f$ of the order of 2 the spectral density obeys the law of "two thirds" and at frequencies $\tau_e f > 7$, the law of "five thirds". Thus, the form of the correlation function for the pulsating concentration of an  atmospheric  pollutant  must  be  close  to  the exponential one what has been demonstrated by the results obtained in Ref. 2.

Now  we  have  all  values  and  relations  that  are necessary  for  making  calculations,  and  now  we  can start  to  discuss  the  results  obtained.  According  to Eqs. (1), (3), and (4), the expression for determining $\lambda_+(C_0)$ has the form

$$\lambda_+(C_0) = \frac{\sigma}{\sqrt{2\pi}\beta\tau_e} \times$$

$$\times \left\{\exp\left[-\frac{(C_0 - \overline{C})^2}{\beta^2}\right] - \exp\left[-\frac{(C_0 + \overline{C})^2}{\beta^2}\right]\right\}. \quad (5)$$

Usually,  the  frequency  of  the  events  when concentration exceeds a given level is calculated by the "modified" Rice formula.[7] By the modification we mean substitution  of  the  lognormal  probability  density  of concentration into the formula obtained by Rice.  As a result we obtain, taking into account the exponential form  of  the  correlation  function  of  concentration pulsation, the following expression

$$\lambda_+(C_0) = \frac{I\exp\left\{-\dfrac{\ln^2\left[C_0\sqrt{1+I^2}/\ln(1+I^2)C_m\right]}{2\ln(1+I^2)}\right\}}{\tau_e 2\pi\sqrt{1+I^2}\,\ln(1+I^2)}, \quad (6)$$

where $I = \sigma/\overline{C}$ is intensity of concentration pulsation of the atmospheric pollutant; $C_m = \overline{C}/\sqrt{1+I^2}$ is the median pollutant concentration. Figure 1 presents mean frequencies  of  occurrence  of  the  events  when concentration exceeds a given level depending on the threshold $C_0$. The    frequencies  are  calculated  by Eqs. (5), (6) and normalized to $\lambda_+(C_0/\overline{C} = 1)$; the threshold is normalized to the mathematical expectation of concentration $\overline{C}$.   The  Figure 1 presents also the experimental  data  obtained  in  Ref. 1.  The upper curves are  constructed  for   $I = 0.125$   ($\gamma = 0.14$),  and  the experimental  points  correspond  to  the  equilibrium regime  discussed  above;  the  lower  curves  are constructed for $I = 1$ ($\gamma = 0.84$), and the experimental points correspond to the transient regime. We see that the modified Rice formula (6) does not agree with the experimental     data.     This     demonstrates

that the lognormal distribution describes pulsation of atmospheric pollutant concentration rather roughly. At the same time, formula (5) quite well agrees with the experimental data.

Thus, one can state that the transient regime corresponds to pollutant concentration intermittence close to 1; the equilibrium regime corresponds to the intermittence tending to zero. Calculations by the formula (5) and their comparison with the experimental data[1] demonstrate that the transient regime is observed for $\gamma > 0.5$, and equilibrium regime for $\gamma < 0.5$. This well agrees with the assumption[2] that the transient regime must be observed for short duration of diffusion and equilibrium regime for the long one.
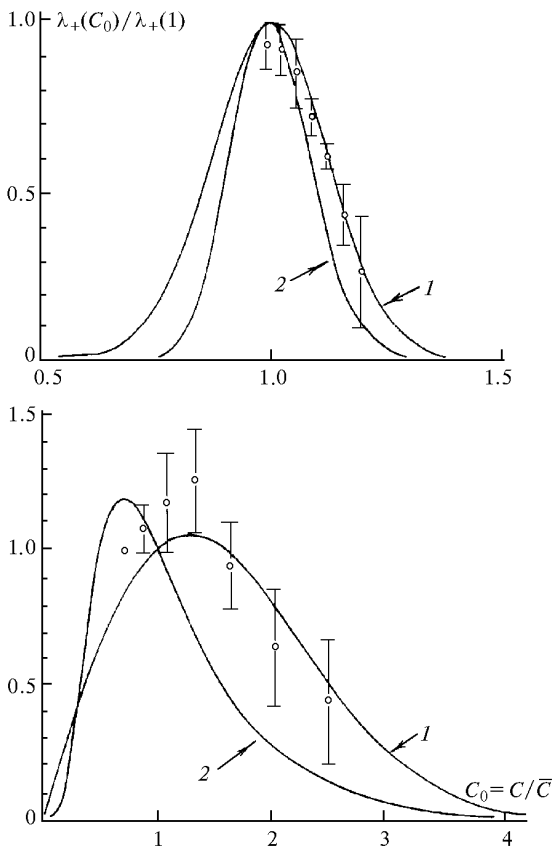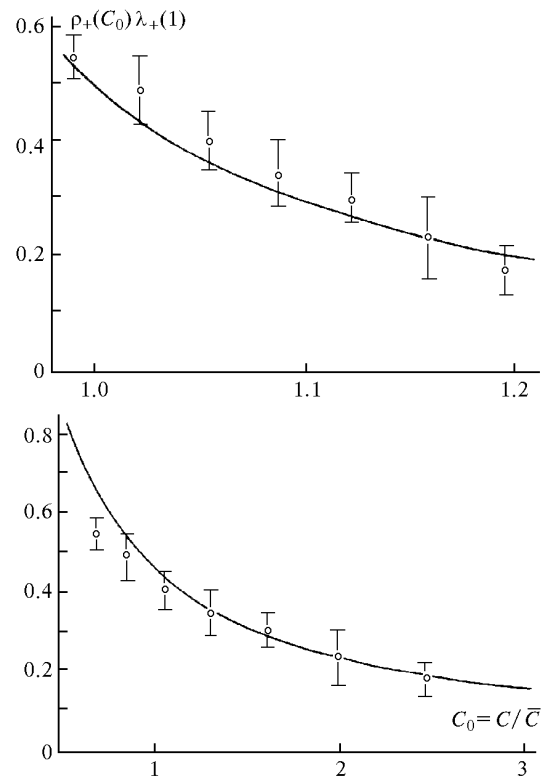


**Fig. 1.** The dependence of $\lambda_+(C_0)/\lambda_+(C_0 = \overline{C})$ on $C_0 = C/\overline{C}$. Curve *1* corresponds to calculations by Eq. (5), curve *2* to calculations by Eq. (6). The circles and vertical bars present the data from Ref. 4. The upper curves are constructed for $I = 0.125$, the lower ones for $I = 1$.

The formula for determining mathematical expectation of the emission duration $\rho_+(C_0)$ has the form

$$\rho_+(C_0) =$$

$$= \frac{\pi\beta\tau_e \left\{ \mathrm{erf}\left[\frac{(C_0 + \overline{C})^2}{\beta^2}\right] - \mathrm{erf}\left[\frac{(C_0 - \overline{C})^2}{\beta^2}\right] \right\}}{\sqrt{2}\sigma \left\{ \exp\left[-\frac{(C_0 - \overline{C})^2}{\beta^2}\right] - \exp\left[-\frac{(C_0 + \overline{C})^2}{\beta^2}\right] \right\}} . \quad (7)$$

Figure 2 presents mathematical expectations of the emission duration as functions of the threshold values $C_0$. These have been calculated by Eq. (7) and normalized to $1/\lambda_+(C_0/\overline{C} = 1)$. The threshold is normalized to the mathematical expectation of concentration $\overline{C}$. The points and vertical bars present the experimental data from Ref. 1 and their scatter. The upper curve is constructed for $I = 0.125$ ($\gamma = 0.14$) and corresponds to the equilibrium regime discussed above, while the lower curve for $I = 1$ ($\gamma = 0.84$) that corresponds to the transient regime. Thus, quite a satisfactory coincidence between the experimental results and calculated by Eq. (7) is obtained.



**Fig. 2.** The dependence of $\rho_+(C_0)\lambda_+(C_0 = \overline{C})$ on $C_0 = C/\overline{C}$ calculated by Eq. (7). The circles and vertical bars present the data from Ref. 4. The upper curve corresponds to $I = 0.125$, the lower one to $I = 1$.

Note that the function $f_1(C')$, which was used above without any rigorous grounds, describes the distribution of the concentration derivative with respect to time quite satisfactorily, because calculations by the formulas (5) and (7) agree well with the experimental data. It is clear that $\lambda_+(C_0)$ and $\rho_+(C_0)$ can be obtained if and only if one has the values of mathematical expectation of a pollutant concentration, its variance, and the Eulerian time scale of pulsation process. The above-mentioned condition that $\gamma > 0.1$ (Ref. 4) is one of the restrictions upon the use of the obtained relations.

We also would like to note that the probability density (3) used in this paper was obtained in Ref. 4 under the assumption that the process of pollutant spread is ergodic. In this case, we can change the procedure of averaging over an ensemble by averaging over time. Although the atmospheric processes are not stationary one can, however, if the period of pollutant concentration averaging is much longer than a characteristic scale of concentration pulsation and much less than the total time of pollutant spread, assume the ergodicity condition to be fulfilled sufficiently accurate for a practical use. In the theory of random processes, this procedure is called "sliding average".[3] The characteristic time of concentration pulsation in the atmosphere (Eulerian temporal scale) is about 10 s, the characteristic time of atmospheric pollutant spread is several hours. So, in this problem, the interval of concentration averaging should be equal to approximately 10 min.[4] In the case when the correlation function is close to the exponential form,

the reasoning remains valid for the derivative of the concentration as well.

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