

# Representative sampling of atmospheric pollutants

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When monitoring atmospheric pollutants, air is sampled during a finite time. The measured pollutant concentrations are averaged over the sampling period. The spread of atmospheric pollutants occurs in a turbulent medium, therefore, the measured concentration values are random. This work considers the problem of a choice of the sampling time to provide for pollutant concentration values with preset errors.

The air sampling is one of the primary techniques in atmospheric monitoring. Since it takes some time  $T > 0$ , the measured concentration values  $C_T$  are averaged over the observation period  $T$ :

$$C_T = \frac{1}{T} \int_0^T C(t) dt, \quad (1)$$

where  $C(t)$  is the pollutant concentration at some moment  $t$ . Atmospheric pollution propagates through a turbulent medium. Therefore,  $C$  and  $C_T$  are random quantities. In this connection, to make sampling reliable and statistically distributed is of importance. In this paper, we consider the problem of a choice of some appropriate sampling time providing for obtaining pollutant concentrations at a given error.

Consider a stationary process of the pollutant transport. In this case, the pollutant concentration expectation is independent of time:  $\bar{C} = \text{const}$ . Applying the averaging over the statistical ensemble to Eq. (1), we have

$$\bar{C}_T = \frac{1}{T} \int_0^T \bar{C}(t) dt. \quad (2)$$

The estimate of the expectation of the measured pollutant concentration (1) is unbiased,  $\bar{C}_T = \bar{C}$ , and its variance is determined by the relation<sup>1</sup>

$$\sigma_T^2 = \frac{2}{T} \int_0^T (1 - \xi/T) B(\xi) d\xi, \quad (3)$$

where  $B(\xi)$  is the correlation function of fluctuations of atmospheric pollutant concentration, the expression for which was derived earlier<sup>2</sup> based on the exact analytical solution of the Fokker–Planck–Kolmogorov equation for the transitional concentration probability density:

$$B(\xi) = \sigma^2 \exp\left(-\frac{|\xi|}{\tau}\right), \quad (4)$$

where  $\sigma^2$  is the pollutant concentration variance;  $\tau$  is the Eulerian time scale of the concentration variance.

According to Eq. (4), the expression for the measured concentration variance is the following:

$$\sigma_T^2 = \frac{2\tau^2}{T^2} \sigma^2 \left[ \frac{T}{\tau} - 1 + \exp\left(-\frac{T}{\tau}\right) \right]. \quad (5)$$

It was shown in Ref. 3 that the best estimates of the measured concentration expectation  $\bar{C}_{T_{\text{opt}}}$  and variances  $\sigma_{T_{\text{opt}}}^2$  are

$$\bar{C}_{T_{\text{opt}}} = \frac{\tau}{2\tau + T} \left[ C(0) + C(T) + \frac{1}{T} \int_0^T C(t) dt \right]; \quad (6)$$

$$\sigma_{T_{\text{opt}}}^2 = \frac{2\tau}{2\tau + T} \sigma^2. \quad (7)$$

At a sufficiently large sampling time ( $T \geq \tau$ ), estimates (5) and (7) coincide. In particular, with this requirement being fulfilled, the variance of the measured concentration of  $C_T$  can be found as

$$\sigma_T^2 = 2\sigma^2 \frac{\tau}{T}. \quad (8)$$

Table 1 gives some values of  $\sigma_T^2$  versus  $T$  calculated by Eq. (8).

**Table 1. Variance of measured pollutant concentrations versus sampling time**

$\sigma_T^2/\sigma^2$	0.1	0.05	0.01
$T/\tau$	20	40	200

There is one more circumstance to be taken into account when choosing the sampling time. It is also determined by the statistical nature of the pollution propagation process and is connected with the effect of the concentration alternation, i.e., the probability of observation of its zero values. Thus, prior to sampling, we must estimate the probability of the zero concentration. The concentration distribution function for a propagating pollutant has the form<sup>4</sup>:

$$F(C) = 1 + \frac{1}{2} \left[ \text{erf}\left(\frac{C - \bar{C}}{\beta}\right) - \text{erf}\left(\frac{C + \bar{C}}{\beta}\right) \right], \quad (9)$$

where erf stands for the probability integral;  $\beta$  is the second parameter of the distribution function. The expression for  $C_T$  coincides with Eq. (9) accurately to designations.<sup>4</sup> The function is the exact analytical solution of the Fokker–Plank–Kolmogorov equation and is derived in the framework of assumptions on the semiempirical approach to the description of atmospheric pollution propagation. Function (9) is justified by a cycle of laboratory experiments with a wind tunnel and corresponds to classical properties of asymptotics of the concentration distribution functions in terms of the turbulent combustion theory.<sup>4</sup> It follows from Eq. (9) that the probability of zero concentration for a pollutant is

$$F(0) = 1 - \text{erf}\left(\frac{\bar{C}}{\beta}\right). \quad (10)$$

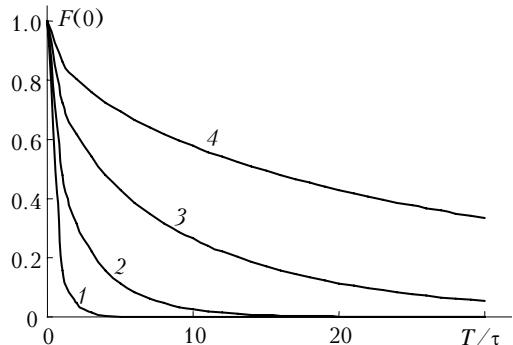
To find  $\beta$ , it is convenient to use the relation<sup>4</sup>

$$\frac{\sigma^2}{\bar{C}^2} = \text{erf}\left(\frac{\bar{C}}{\beta}\right)\left(1 + \frac{\beta^2}{2\bar{C}^2}\right) - 1 + \frac{\beta}{\sqrt{\pi}\bar{C}} \exp\left(-\frac{\bar{C}^2}{\beta^2}\right). \quad (11)$$

When  $T \geq \tau$ , from Eqs. (8), (10), and (11) the following expression is derived for the probability of zero concentration:

$$F(0) = 1 - \text{erf}\left(\frac{\bar{C}}{4\sigma}\sqrt{\frac{T}{\tau}}\right). \quad (12)$$

Figure 1 presents  $F(0)$  as the function of  $T/\tau$  for some intensity values of the pollutant concentration fluctuations  $I = \sigma/\bar{C}$ .



**Fig. 1.** The characteristic  $F(0)$  as the function of  $T/\tau$ . Curves 1–4 correspond to the values  $I = 0.25; 0.5; 1$ ; and  $2$ .

Thus, a longer sampling time not only decreases the variance, but also reduces the probability of zero concentrations. As is seen, this probability can be rather significant.

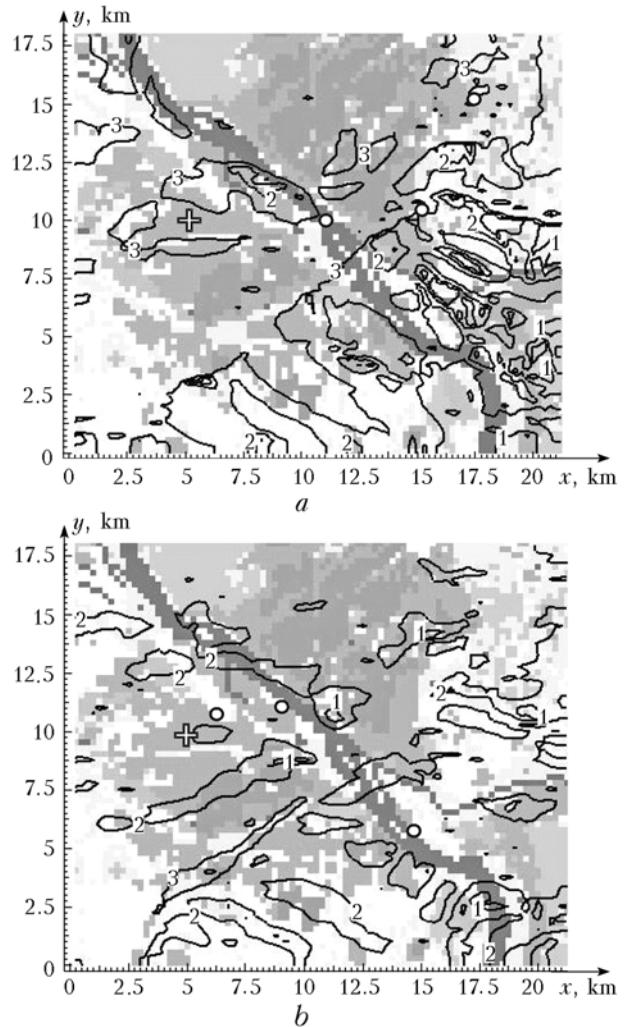
The empirical relation for the Eulerian time scale of the wind velocity fluctuation  $\tau_u$  based on the analysis of the time series of the parameter, recorded with the acoustic anemometer in the surface atmospheric layer is given in Ref. 4:

$$\tau_u \approx (45 \pm 8) \frac{z}{\bar{U}}, \quad (13)$$

where  $z$  is the height above the underlying surface, m;  $\bar{U}$  is the mean modulus of the wind velocity, m/s. A similar expression was used in Ref. 5 to determine the Lagrangian time scale of the concentration fluctuations. Assume that Eq. (13) is applicable to estimates of the Eulerian time scale  $\tau$  as well.

Now, as an example, consider the calculation of the Eulerian time scale by Eq. (13) for Novosibirsk city. The wind velocity fields for this region were found through the numerical–analytical model.<sup>6</sup> In the first variant of calculations we set meteorological conditions typical for the region at 3 a.m., i.e., western wind of 1 m/s at the vane height at the left boundary of the considered area. The second variant assumed wind velocity of 5 m/s at 3 p.m.

Figure 2 shows isolines of the  $\tau$  calculated values for both variants at a height of 10 m above the surface.



**Fig. 2.** Isolines for  $\tau$  calculated for 3 a.m. (a) and 3 p.m. (b) of the local time. Isolines 1–3 correspond to  $\tau = 20, 40$ , and  $60$  s (a);  $300, 600$ , and  $1200$  s (b).

As is seen, the characteristic time scales of fluctuations for the night and daytime conditions

differ substantially. The isoline fields have a complex structure due to different types of the underlying surface within the considered domain. Since daytime values of  $\tau$  are higher than the night ones, the period of the correct daytime sampling must be much longer than the night one.

For the above meteorological conditions we estimated the probability of the pollutant zero concentration. To do this, in a point with coordinates  $x_0 = 5$  km,  $y_0 = 10$  km,  $z_0 = 50$  m we positioned a stationary point source of pollution with the power  $Q_0 = 10^{10}$  relative units and a particle size of 10  $\mu\text{m}$  in diameter (the cross in Fig. 2). The values for  $\bar{C}$  and  $\sigma^2$  were found by solving a semi-empirical equation for turbulent diffusion and the equation for the concentration variance,<sup>7,8</sup> respectively. The turbulent diffusion coefficients were set according to the algebraic model like in Ref. 9.

Table 2 gives the examples of  $F(0)$  calculation for several pollutant concentration values at several points in the plane  $z = 10$  m. As is seen, the greater the concentration fluctuation intensity, the higher the probability of zero concentrations. Therefore, at points with strong fluctuations, concentration measurements must cover a much longer period than at the points with weak fluctuations.

**Table 2. Zero concentration probability for some points of the area**

*Point coordinates $x$ ; $y$ , km; in the plane $z = 10$ m	$I = \sigma/\bar{C}$	$F(0) = 1 - \text{erf}\left(\frac{\bar{C}}{\beta}\right)$
<i>Variant 1:</i>		
11.25; 10.25	0.1	0
15.00; 10.75	0.495	0.045
17.50; 15.50	2	0.746
<i>Variant 2:</i>		
8.25; 10.75	0.108	0
5.75; 11.00	0.492	0.043
12.50; 8.25	2.01	0.747

\* Indicated as circles in Fig. 2.

A similar analysis can be performed for the case of nonstationary concentration fields of the transported pollutant, when  $\bar{C} = \bar{C}(t)$ . In this case,

a quasi-stationary approximation for the correlation function of the concentration variance<sup>4</sup> can be used:

$$B(t, \xi) = \sigma^2(t) \exp\left(-\frac{|\xi|}{\tau}\right), \quad (14)$$

which is consistent with the semi-empirical approach to the description of the atmospheric pollution propagation process.<sup>4</sup> In this case, the expression for the concentration variance (3) takes the form

$$\sigma_T^2 = 2\tau \int_0^T \sigma^2(t) \left[1 - \exp\left(-\frac{t}{\tau}\right)\right] dt. \quad (15)$$

Thus, when monitoring the atmospheric pollution from some known source under typical meteorological parameters, it is possible to estimate the sampling errors connected with statistics of the process of atmospheric pollution transport. Generally, when the pollution source cannot be correctly allocated, the estimate of the error in pollutant concentration measurements becomes a more complicated problem.

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