

## INFLUENCE OF METEOROLOGICAL CONDITIONS ON VERTICAL PROFILES AND FLUXES OF ATMOSPHERIC TRACE SPECIES

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*The physical and quantitative analysis of the influence of meteorological conditions on vertical distribution of trace gases has been performed on the basis of the relationships predicted by the similarity theory. The tables have been composed for the gas-exchange coefficients within a wide range of meteorological conditions and the characteristics of the Earth's surface. The gas-exchange coefficient and the turbulent flux of atmospheric trace species can vary by several tens times under the effect of the above factors.*

Within the scope of recent studies of air pollution and monitoring of the environment the analysis of some peculiarities in the trace species distribution under different meteorological conditions is of urgent importance. Some physical and quantitative aspects of this analysis are discussed in this paper.

It is known that the main portion of anthropogenic species is concentrated in the atmospheric boundary layer at heights up to several hundreds of meters. Therefore we analyze here only the relationships obtained in Ref. 1 as a result of applying the similarity theory to the problem of atmospheric pollution.

From the equation of motion, heat inflow, and contaminant transfer in the boundary layer and by generalizing the theory for the case of nonequilibrium stratification, we derive the following expressions for a trace species concentration ( $q$ ), air temperature ( $T$ ), and wind velocity ( $c$ ):

$$q(z) = q_1 + q_* \ln(\eta/\eta_1); \quad (1)$$

$$T(z) = T_2 + T_* \ln(\eta/\eta_2) - \gamma_a(z - z_2); \quad (2)$$

$$c(z) = (u_*/\kappa) \ln(\eta/\eta_0). \quad (3)$$

Here  $q_*$  and  $T_*$  are the scales (characteristic values) of  $q$  and  $T$  variation in the boundary layer, connected with turbulent fluxes of species,  $Q_q(0)$ , and heat fluxes,  $Q_T(0)$ , by the relationships:

$$q_* = -Q_q(0)/(\kappa \rho_1 u_*); \quad T_* = -Q_T(0)/(\kappa c_p \rho_1 u_*), \quad (4)$$

where  $\eta$  is new variable related to the height  $z$  by the relationship:

$$\eta = \exp(z/L_*) - 1; \quad (5)$$

$q_1$  and  $T_2$  are the trace species concentration and air temperature at the levels  $z_1$  and  $z_2$ , respectively;  $u_*$  is

the dynamic velocity related to the wind velocity  $c_1$  at the level  $z_1$  by the expression:

$$u_* = \kappa c_1 / \ln(\eta_1/\eta_0); \quad (6)$$

$\eta_1$ ,  $\eta_2$ , and  $\eta_0$  are the values of  $\eta$  at  $z_1$ ,  $z_2$ , and  $z_0$ , respectively ( $z_0$  is the parameter of roughness);  $L_*$  is the scale of the atmospheric boundary layer (Monin-Obukhov):

$$L_* = \frac{u_*^2 T_2}{\kappa^2 g T_*} = - \frac{c_p \rho_1 u_*^3 T_2}{\kappa g Q_T(0)}; \quad (7)$$

$\kappa = 0.38$  is the constant;  $\rho_1$  is the air density at  $z_1$ ;  $g$  is the acceleration of gravity.

If formula (2) is written for a certain altitude  $z_3$ , at which the temperature  $T_3$  is measured, and thus determined scale  $T_*$  is introduced into the relationship (7), then, taking into account Eq. (6), we derive the following expression for the scale  $L_*$ :

$$\frac{z_1}{L_*} = \frac{\ln^2(\eta_1/\eta_0)}{\ln(\eta_3/\eta_2)} B, \quad (8)$$

where the dimensionless parameter  $B$  is introduced to represent the finite-difference analog of the Richardson number:

$$B = \frac{g z_1 (T_3 - T_2) - \gamma_a (z_3 - z_2)}{T_2 c_1^2}. \quad (9)$$

If the heights  $z_2$ ,  $z_1$ , and  $z_3$  are chosen so that  $z_2 = z_1/n$  and  $z_3 = n z_1$  (where  $n$  is an arbitrary number), then the right-hand side of Eq (8) proves to be dependent, in addition to  $B$ , only on  $z_1/L_*$  and  $z_0/z_1$ ; the ratios  $z_3/L_*$ ,  $z_2/L_*$ , and  $z_0/L_*$  involved in  $\eta_3$ ,  $\eta_2$  and  $\eta_0$  should be written as:

$$z_3/L_* = n z_1/L_*; \quad z_2/L_* = z_1/(n L_*);$$

$$z_0/L_* = (z_0/z_1)(z_1/L_*).$$

The results of  $z_1/L_*$  calculations at different  $z_0/z_1$  and  $n = 2$  (i.e.,  $z_3 = 2z_1$  and  $z_2 = z_1/2$ ) are given in the figure.

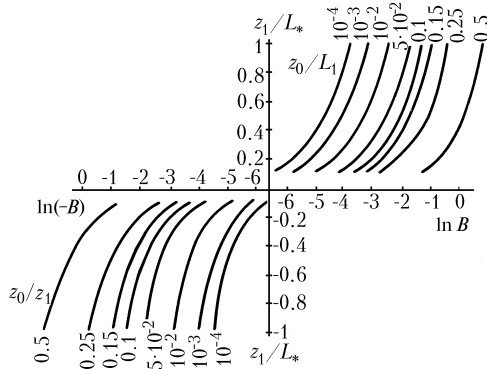


FIG. The dependence of  $z_1/L_*$  on  $B$  and  $z_0/z_1$ .

The exponential-logarithmic formulas (1)–(3) show the basic peculiarities of altitude distribution of the meteorological parameters (wind velocity, air temperature, mass ration of water vapor) and the species concentration.

We use the series expansion of the exponent

$$\eta = \exp(z/L_*) - 1 = z/L_* + z^2/(2L_*^2) + z^3/(6L_*^3) + \dots \quad (10)$$

At low heights and at any values of the parameters  $B$  and  $L_*$  the distribution of  $q$  and  $c$ , as well as of  $T$  is described by the logarithmic formulas (laws):

$$q(z) = q_1 + q_* \ln(z/z_1), \quad (11)$$

$$c(z) = (u_*/\kappa) \ln z/z_0, \quad (12)$$

to which Eqs. (1) and (3) are reduced if in the right-hand side of Eq. (4) only the first (linear) component remains.

If three terms remain in the expansion (10), the formulas (1) and (3) take the form:<sup>1)</sup>

$$q(z) = q_1 + q_* \left( \ln(z/z_1) + \frac{z-z_1}{2L_*} + \frac{z^2-z_1^2}{6L_*^2} \right); \quad (13)$$

<sup>1)</sup> For this purpose  $\eta$  should be written as  $\eta = x(1+x/2+x^2/6)$ , where  $x = z/L_*$ . Having used the representation  $\ln(1+y) \approx y$  at small value of  $y = x/2+x^2/6$ , we obtain  $\ln \eta = \ln x + x/2 + x^2/6$ .

$$c(z) = \frac{u_*}{\kappa} \left( \ln(z/z_0) + \frac{z-z_0}{2L_*} + \frac{z^2-z_0^2}{6L_*^2} \right). \quad (14)$$

Analysis of the observational data has shown that for describing the wind velocity profile at stratification, essentially different from the equilibrium one (when  $|L_*| \sim 5\text{--}50$  m), in the right-hand sides of Eqs. (13) and (14) we should take into account not only the first and second terms (in this case the formula is called linear-logarithmic), but the third (square-law) term as well.

The profiles  $q(z)$ ,  $c(z)$ , and  $T(z)$  well (within the error less than 5%) agree with the logarithmic ones up to the height  $z_* = 0.1 |L_*|$ . With the increase of the height these profiles deviate more and more from the logarithmic ones. The wind velocity grows with the increase of  $z$  more rapidly than  $\ln z$ , at a stable ( $B > 0$ ,  $L_* > 0$ ) stratification of the boundary layer and less rapidly at unstable ( $B < 0$ ,  $L_* < 0$ ) one.

During the last decades numerous data of the gradient measurements of  $c$  and  $T$  in different regions of the world have been analyzed. This analysis has shown that using Eq. (14) and more general formulas (2) and (3) the experimental data are approximated with high precision. As a result, we can conclude that formula (1) describes the altitude distribution  $q$  to a high accuracy.

The results of calculations of the dimensionless ratio  $\beta$  by formula (1)

$$\beta = \frac{q(z) - q_1}{q_*} = -\kappa \rho_1 u_* \frac{q(z) - q_1}{Q_q(0)}$$

are given for several values of  $L_*/z_1$  in Table I. Under conditions of usually observed positive fluxes ( $Q_q(0) > 0$ ), the trace species concentration decreases with the height:  $q(z) < q_1$  at  $z/z_1 > 1$ . However, the rate of the  $q$  decrease with height differs essentially at stable ( $L_* > 0$ ) and unstable ( $L_* < 0$ ) stratifications of the atmosphere.

TABLE I. The ratio  $10^2\beta$ .

$L_*/z_1$	$z/z_1$								
	0.25	0.5	2	5	10	20	40	100	200
10	-142	-72	74	182	279	411	623	1225	2225
25	-141	-70	71	169	249	340	457	718	1120
$\infty$	-139	-69	69	161	230	300	369	460	530
-25	-137	-68	67	153	213	264	301	322	324
-10	-135	-67	64	142	189	221	233	235	235

At  $L_* > 0$  the concentration decreases with the height much rapidly than at  $L_* < 0$ , i.e., at  $z/z_1 = 100$  the ratio  $\beta$  amounts to 12.25 at  $L_*/z_1 = 10$  and equals only 2.39 at  $L_*/z_1 = -10$ .

By means of the parameters  $L_*$  and  $u_*$  the difference  $q_1 - q(z)$  rather complicatedly depends on the wind velocity, the temperature lapse rate ( $\gamma = -\partial T/\partial z$ ), and the roughness parameter ( $z_0$ ).

Let us determine the difference  $q_1 - q(z)$  for the two values of  $c_1$ , assuming that all other parameters ( $Q_T, Q_q, z_0$ ) are the same. Assume that under a light breeze ( $c'_1$ ) the scale  $L'_*/z_1$  is close to 10 (at  $B > 0$ ). If the wind velocity increases up to  $c''_1$ , being more than  $c'_1$  by a factor of 5, then  $L_*$ , according to Eqs. (6) and (7) increases by about 125 times. At the value of  $L_*/z_1 \approx 1250$  the height distribution of  $q$  is described by the logarithmic formula (11).

The values of the ratio  $b = (q_1 - q)' / (q_1 - q)''$  of the differences  $q_1 - q$  at  $c'_1$  and  $c''_1 = 5c'_1$  are

$z/z_1$	0.25	0.5	2	5	10	25	100	200
$b$	5.17	5.23	5.43	5.72	6.14	7.34	13.5	21.2

With the increase of wind velocity the difference  $q_1 - q$  grows more rapidly than the ratio  $\beta$ .

According to Eq. (6), an increase in the roughness parameter  $z_0$  affects the values  $\beta$  and  $b$  in the same manner as the growth of  $c_1$ . Let us give the values of the ratio  $a = (q_1 - q)' / (q_1 - q)''$  of the difference  $q_1 - q$ , at  $z_0/z_1 = 10^{-2}$  and  $z''_0/z_1 = 0.25$ :

$z/z_1$	0.25	0.5	2	5	10	25	100	200
$a$	3.43	3.51	3.60	3.80	4.08	4.87	8.95	14.1

The dependence of the difference  $q_1 - q$  on  $\gamma$ , having a pronounced effect on the heat flux  $Q_T(0)$ , and owing to it, on  $L_*/z_1$ , follows immediately from the data of Table I. Thus, if at a certain value of  $\gamma' < \gamma_a$  (stable stratification:  $Q_T(0) < 0$ ) the scale  $L_*/z_1 = 10$  while at the other value of  $\gamma'' < \gamma_a$  ( $Q_T(0) > 0$ ) the scale  $L_*/z_1 = -10$ , the ratio  $(q_1 - q)' / (q_1 - q)''$  varies from 1.08 at  $z/z_1 = 0.25$  to 9.31 at  $z/z_1 = 200$ .

It is readily seen that all the above regularities are explained by the influence of turbulent exchange on the transfer and altitude distribution of pollutants. In the model discussed the dependence of the turbulence coefficient  $k_z$  on the altitude is described by the following formula:

$$k_z = \kappa u_* L_* (1 - \exp(-z/L_*)). \tag{15}$$

At a stable stratification ( $B > 0, \gamma < \gamma_a$ ) and at a certain altitude,  $k_z$  is the larger, the greater are  $L_*$  and  $u_*$ .

On the one hand, the pollutant flux  $Q_q = -\rho k_z (\partial q / \partial z)$  is constant, and, on the other hand, this value is determined by the amount of pollutants emitted from different sources near the ground surface. Hence, the increase of the turbulent exchange intensity (the increase of  $c_1, z_0$ , and  $\gamma$  is useful in this case;  $c_1$  and  $z_0$ , affect  $k_z$  in two ways through  $u_*$  and  $L_*$ ) is accompanied by a decrease of  $-\partial q / \partial z$ ; the slower the pollutant concentration decreases with height, the larger are  $c_1, z_0$ , and  $\gamma$ .

At the unstable stratification ( $a < 0, \gamma > \gamma_a$ ), when  $L_* < 0$ , at a given height  $k_z$  also grows with increasing  $L_*$  (i.e., with the decrease of  $L_*$  absolute value). Therefore in this case the pollutant concentration height decrease is the slower, the greater is  $\gamma$ . However, the dependence of the distribution of  $q$  on  $c_1$  and  $z_0$  at  $\gamma > \gamma_a$  is not single-valued, as at  $\gamma < \gamma_a$ . At  $\gamma > \gamma_a$  ( $L_* < 0$ ) the growth of  $c_1$  and  $z_0$  results via  $u_*$  in the increase of  $k_z$ , and through  $L_*$  it results in the decrease of  $k_z$ . Thus, if at  $L_*/z_1 = -10$  and  $c'_1 = 1$  m/s the wind velocity increases up to  $c''_1 = 5$  m/s, then  $k_z$  at 10 m height grows almost 5 times under the effect of  $u_*$  and decreases by a factor of 1.72 under the effect from  $L_*$ , i.e., as a result,  $k_z$  increases by a factor of 2.9. The above-mentioned ratio  $L = (q_1 - q)' / (q_1 - q)''$  takes the following values at ( $c''_1 = 5 c'_1, L_*/z_1 = -10$ ):

$z/z_1$	0.2	0.5	2	5	10	25	100	200
$b$	4.86	4.82	4.65	4.40	4.11	3.52	2.55	2.22

Assume that the turbulent flux of a trace species  $Q_q(0)$  has the form:

$$Q_q(0) = -\rho_1 \alpha_q c_1 (q_3 - q_2), \tag{16}$$

where  $q_3$  and  $q_2$  are the species concentrations at the levels  $z_3$  and  $z_2$ ;  $c_1$  is the wind velocity at  $z_1$ ;  $\alpha_q$  is the gas exchange coefficient.

At vertical distribution of  $q(z)$  and  $c(z)$  described by the formulas (1) and (3), for  $\alpha_q$  we derive the ratio:

$$\alpha_q = \frac{\kappa^2}{\ln(\eta_3/\eta_2) \ln(\eta_1/\eta_0)}, \tag{17}$$

where  $\eta_3, \eta_2, \eta_1$ , and  $\eta_0$  are the values of the variable  $\eta$  at  $z_3, z_2, z_1$ , and  $z_0$ , and  $z_3 = nz_1; z_2 = z_1/n$ .

Results of  $\alpha_q$  calculation at  $n = 2$  are given in Table II. At a constant ratio  $z_0/z_1$  the gas exchange coefficient increases by several times, when passing from a very stable stratification, when  $L_* \sim (5.0 \div 1.0)z_1$ , to a very unstable one, when  $L_* \sim -(5.0 \div 1.0)z_1$ . The value of  $\alpha_q$  essentially depends on the roughness parameter, namely, at a constant  $L_*$  the value of the gas exchange coefficient increases by one order of magnitude with the growth of  $z_0/z_1$  from between  $10^{-4}$  and  $10^{-3}$  to 0.25-0.50.

To calculate trace species flux by formula (16), one needs in addition to the wind velocity  $c_1$  at the level  $z_1$ , to have the data on the species concentration at two levels,  $z_3$  and  $z_2$ . In practice the measurements of  $q$  are normally performed only at one level. In this connection, along with the calculations at  $n = 2$  the value of  $\alpha_q$  is calculated at  $n = 50$  (Table III). If in this case  $z_1 = 10$  m, then  $z_3 = 500$  m and  $z_2 = 0.2$  m. The estimates show that at 500 m height the species concentration  $q_3$  is by

about one order of magnitude less than its value  $q_2$  close to the earth's surface. By this it is meant that the calculation of  $Q_q(0)$  can be made with an error less than 10% only using the concentration  $q_2$  measured at level  $z_2$ :

$$Q_q(0) = \rho_1 \alpha_q c_1 q_2, \tag{18}$$

where  $c_1$  is the wind velocity at 10 m level.

TABLE II. The gas exchange coefficient  $10^4 \alpha_q$  ( $n = 2$ :  $z_3 = 2z_1$ ;  $z_2 = 0.5z_1$ ).

$z_1/L_*$	$z_0/z_1$							
	$10^{-4}$	$10^{-3}$	$10^{-2}$	$5 \cdot 10^{-2}$	0.1	0.15	0.25	0.50
1.0	64.7	84.7	123	179	226	267	351	648
0.8	71.9	94.5	137	203	257	310	405	811
0.6	80.1	106	154	232	293	358	468	893
0.4	89.6	119	172	264	340	410	545	1057
0.2	101	134	197	304	389	470	640	1258
0.1	107	142	204	319	409	500	692	1301
0.05	109	146	218	337	436	529	708	1437
0.025	110	149	223	343	446	542	739	1475
0	113	151	226	348	452	549	751	1503
-0.025	115	153	230	354	460	559	766	1536
-0.05	116	155	234	360	469	570	782	1571
-0.1	120	160	241	374	486	593	815	1646
-0.2	127	171	258	402	525	641	885	1804
-0.4	144	193	294	465	619	749	1046	2172
-0.6	163	220	337	540	711	879	1239	2628
-0.8	185	250	384	628	830	1032	1470	3183
-1.0	211	286	444	720	974	1218	1756	3889

The data given in Tables II and III show that the trace gas flux under the influence of meteorological conditions (thermal stratification, wind velocity) and roughness of the earth's surface may vary by several tens or even hundreds times.

TABLE III. The gas exchange coefficient  $10^4 \alpha_q$  ( $n = 50$ :  $z_3 = 50z_1$ ;  $z_2 = 0.02z_1$ ).

$z_1/L_*$	$z_0/z_1$							
	$10^{-4}$	$10^{-3}$	$10^{-2}$	$5 \cdot 10^{-2}$	0.1	0.15	0.25	0.50
1.0	2.75	3.59	5.22	7.59	9.55	11.3	14.9	27.5
0.8	3.40	4.46	6.47	9.59	12.1	14.6	19.1	38.3
0.6	4.40	5.83	8.46	12.8	16.1	19.2	25.7	49.1
0.4	6.17	8.20	11.8	18.2	23.4	28.3	37.6	72.8
0.2	10.0	13.3	19.6	30.2	38.2	46.7	63.6	125
0.1	14.0	18.5	26.6	41.6	53.4	65.2	83.8	170
0.05	16.6	22.3	33.3	51.5	66.6	80.8	108	220
0.025	18.3	24.6	36.8	56.6	73.6	89.4	122	243
0	20.0	26.7	40.0	61.6	80.0	97.2	133	266
-0.025	21.7	28.8	43.3	56.7	86.7	105	144	289
-0.05	22.9	30.6	46.3	71.2	92.7	113	155	311
-0.1	25.4	33.8	50.9	79.1	103	125	172	348
-0.2	28.6	38.5	58.1	90.5	118	144	199	406
-0.4	33.1	44.4	67.6	92.5	142	172	241	500
-0.6	36.5	49.3	75.5	121	159	197	277	588
-0.8	39.5	53.4	81.9	134	177	220	314	679
-1.0	42.2	57.4	89.1	144	196	244	352	781

REFERENCES

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