

Impurity distribution in foggy and low cloud cover conditions

*Nurgali Zaurbekov*¹, *Allayarbek Aidosov*², *Gulzat Zaurbekova*^{2*}, and *Nurbike Zaurbekova*³

¹Abai Kazakh National Pedagogical University Almaty, Kazakhstan

²Al-Farabi Kazakh National University Almaty, Kazakhstan

³Kazakh National Women's Teacher Training University, Almaty, Kazakhstan

Abstract. The paper discusses the possibilities of modelling the distribution of harmful impurities emitted into the atmosphere by industrial enterprises in conditions of fog and low clouds. With several sources located along a certain line or dispersed over an area, under conditions of elevated temperature inversion, which is often observed in fogs, additional reasons for increasing pollution of the surface air layer may appear. Under such conditions, calculations show that the distance at which the maximum concentration is observed increases substantially, and the concentration decreases very slowly with the distance after its maximum. As a result, the effect of the mutual overlapping of concentration fields from individual sources increases and more uniform pollution than in convective conditions is created in the industrial area. At the same time, even if the concentration from a separate source is relatively small, the total pollution can be very significant.

1 Introduction

In some previous papers [1-4], we described the possibilities of mathematical modelling of the distribution in the atmospheric air of harmful impurities emitted by oil and gas producing fields. However, in foggy and low cloud conditions, there are some features that need to be considered separately. At present, the conditions of air pollution during fogs and the nature of the direct effect of fogs on the turbulent diffusion of pollutants have been insufficiently studied. The available theoretical and experimental materials are few, insufficiently systematized and analyzed. The influence of fogs on the content of harmful substances in the surface boundary layer of the atmosphere is very complex. In the presence of fogs, specific distribution conditions of meteorological elements are often observed, contributing to an increase in the concentration of harmful substances in the surface layer and air.

In the presence of fogs, the qualitative composition of impurities and the toxic nature of their effects on humans change. They are partially absorbed by water droplets, which sometimes lead to the formation of new substances; in the atmosphere, the impurity concentration decreases. The nature of the interaction of gases and some aerosols, especially hygroscopic, is different.

* Corresponding author: agu_nurgali@mail.ru

On hygroscopic particles, moisture condensation and fog formation may begin at a relative humidity of less than 100%; the microphysical characteristics of the fog are affected by the number and properties of condensation nuclei. Air vapour deposition on aerosols increases their size and sedimentation rate to the earth's surfaces.

Given the complexity of the processes, it is of particular importance to develop theoretical issues of air pollution in fogs with the involvement of physically rich models for their study taking into account the microphysical structure of this issue. At the same time, it is necessary to take into account such characteristics as fog height, water content, temperature stratification as fully and strictly as possible. Since field data on these characteristics are currently insufficient, it is advisable to use theoretical studies for some types of fogs.

2 Mathematical modelling of impurity distribution in foggy and low cloud conditions

The study of turbulent diffusion of pollutants in the presence of non-freezing water bodies or river fogs is of special interest. Often these water surfaces are located in the vicinity of settlements, resort areas or megacities. For clarity, we will explain the essence of the issues discussed by the example of a simpler model of transport of substances in the atmosphere. It is based on the equation, described in [1].

In this paper, it is proposed to determine the distribution of the amount of moisture in the soil η from the equation:

$$\rho_w \frac{\partial \eta}{\partial t} - \frac{\partial Q_s}{\partial z} = 0, \quad (1)$$

where

$$Q_s = K_\eta \rho_w \frac{\partial(\bar{\psi} + z)}{\partial z}, \quad (2)$$

Or

$$Q_s = D_\eta \rho_w \frac{\partial \eta}{\partial z} + K_\eta \rho_w, \quad D_\eta = K_\eta \frac{\partial \psi}{\partial \eta}; \quad (3)$$

here: Q_s – soil moisture flow; K_η – hydraulic conductivity; ρ_w – water density; z – vertical coordinate, defined as in the atmosphere; D_η – diffusion coefficient.

Following [6] we define the variables K_η , $\bar{\psi}$, D_η using the formulas:

$$\bar{\psi} = \bar{\psi}_H (\bar{\eta}_s / \bar{\eta})^b; \quad (4)$$

$$K_\eta = K_{\eta H} (\bar{\eta} / \bar{\eta}_H)^{2b+3}; \quad (5)$$

$$D_\eta = K_{\eta H} (\bar{\eta} / \bar{\eta}_s)^{b+3}, \quad (6)$$

where the index “ H ” indicates the values of the corresponding function under saturation conditions. The last term of equation (3) describing the impurity runoff in the fog can be expressed by the formula [1, 2]:

$$B_c S = P(r)n(r)dr,$$

where $P(r)$ is the amount of impurity absorbed by a fog drop of radius r per unit time, and $n(r)$ is a function of the droplet size distribution. The value of $P(r)$ is usually determined by studying the interaction of pollutants with fog droplets [1].

In close proximity to the surface, a boundary layer is created in which the concentration of the impurity (denoted by q) is less than s since on the surface of the water droplet due to the absorption of the impurity $q=0$. The concentration distribution q can be approximately determined by solving the equation of molecular diffusion and spherical coordinates:

$$\frac{d}{dp} p^2 \frac{dq}{dp} = 0 \quad (7)$$

where p is the radius vector; using as boundary conditions of equality $q=0$ on the surface of the droplet at $p=r \cdot q \rightarrow S$ at a sufficient distance from the droplet (at $p \rightarrow \infty$). This solution has the form $q \rightarrow S(1-rp^{-1})$, hence the impurity flux to the droplet surface is directly found.

$$P(r) = -V \left. \frac{dq}{dp} \right|_{p=r} \quad (8)$$

where V - the coefficient of molecular diffusion of impurities in the air.

According to [1-4], the size distribution function of fog droplets can be approximated simplistically by the following relation:

$$n(r) = \alpha r^2 \exp(-\beta r), \quad \alpha = \frac{(2.5)^5 q_{l1}}{\pi p_w r_m}; \quad \beta = 5r_m^{-1} \quad (9)$$

here q_{l1} denotes the liquid water content of the fog; r_m is the modal radius of drops corresponding to the maximum of the distribution function and p_w the water density. Substituting expression (9) into (8) by performing integrations, we get:

$$B_c = \frac{0.69 q_{l1}}{p_w r_m^2} = 0$$

From the obtained formula it follows that the absorption of the fog impurity, all other things being equal, rises with an increasing water content of the fog Δ or with decreasing droplet size r_m

3 Experiment and discussion of the results

Example. Let us consider the air mass flow of a thermally inhomogeneous underlying surface - a polygon measuring $20 \times 20 \text{ km}^2$, in the western part of the region there is a non-freezing artificial reservoir with a size of $5 \times 5 \text{ km}^2$. The model input parameters take the following values. Soil temperatures at the initial moment at the level $z=0$ i.e. $T_s(0, t) = 278\text{K}$; relative humidity over land $f = 70\%$, and above water $f = 100\%$; horizontal velocity $U_f = 5 \text{ m/s}$; $\Delta x = \Delta y = 250 \text{ m}$; vertical step varied from 5 m in the lower 50 m layer to 100 m in the layer 500 to 1800 m . The intermediate steps were $6, 12, 50 \text{ m}$. In total, 48 levels in the vertical direction were considered. Source height $H_s = 80\text{m}$; $\Delta t = 300\text{s}$; $z_0 = 0,01\text{m}$ for land and $z_0 = 0.001$ for the water surface. Atmospheric temperature $T_a = 268\text{K}$. The remaining parameters correspond to the data stated in [5].

The advective transformation of a colder air mass on a warm water surface ($T_w = 283\text{K}$), is accompanied by a sharp restructuring of the fields of temperature, humidity, the occurrence of water content, which in turn entails a change in the regime of turbulence and wind and the radiation field.

The non-stationarity of our model allows us to trace additionally the spread of pollutants under the condition of evaporation fog. After 45 minutes of integration of the fog nucleation, its upper boundary is at a height of near 100 m at $x = 5 \text{ km}$ and near 155 m at $x = 10 \text{ km}$. In this case, the upper boundary of the fog is higher than the source. Therefore, the impurity remains below the locking layer, although individual particles still penetrate through the upper boundary of the fog. It should be noted that experiments with the same parameters of the model were carried out without fog formation. In this case, it can be seen from the figure, the impurity spreads freely, its height reaches 300 m .

The calculations revealed that for conditions of fog evaporation is characterized by the absorption of a significant part of the impurity by fog drops above the water body and close to the coast, where the highest values of fog water content are observed; almost no impurities remain in the gaseous state at all heights in the fog layer; on the windward coast, this effect will weaken with a distance from the reservoir and a decrease in water content. It is noted that in the case when the source of pollution is located above the fog layer, a decrease in concentration is observed not only inside, but also above it. This can be explained by the fact that due to the intense diffusion flow of impurities directed from top to bottom.

Tables 1, 2 show the results of calculating the concentration of gaseous contaminants from a source 100 m high, located on the west coast 1.0 km from the reservoir, and the turbulent exchange coefficient over the mirror of the water surface for fog conditions.

The value of the radius of the droplets is r_m , corresponding to the maximum of the distribution function $P(r)$, which was taken equal to 2μ . Table 3 shows the s values at heights of $2, 25, 50$ and 75 m above the reservoir at a distance of up to 16 km from the source.

Table 1. Fog water content (g/kg) (numerator) and coefficient of turbulent exchange Kz (m^2s) (denominator)

X, km	Height Z,m						
	2	25	50	75	100	125	150
2	0.05/2.7	0.09/5.4	0.11/7.9	0.08/9.3	0.01/4.4	-2.7	-1.3
4	0.12/3.2	0.14/6.2	0.18/8.9	0.11/9.4	0.02/4.5	-2.6	-1.4
6	0.19/4.7	0.24/7.1	0.25/9.3	0.17/13.5	0.08/15.5	0.02/10.1	-2.4
8	0.30/5.1	0.32/9.2	0.27/11.9	0.17/17.1	0.08/18.2	0.02/10.5	-3.2
10	0.32/6.2	0.36/10.3	0.25/12.2	0.16/16.4	0.04/15.1	-9.2	-2.9
12	0.09/7.1	0.12/10.4	0.14/12.8	0.14/12.9	0.10/11.7	0.02/10.1	-3.4
14	0.05/7.2	0.9/7.4	0.10/7.9	0.05/7.8	0.01/9.1	-5.4	-3.2
16	0.03/7.0	0.08/7.2	0.06/7.7	0.05/7.7	0.01/7.6	-4.1	-2.7

Table 1 shows Fog water content (g/kg) (numerator) and coefficient of turbulent exchange K_z (m²s) (denominator) for $\Delta T_{w-a}=15^\circ\text{C}$, $U_\varphi= 5\text{m/s}$, $f_0=70\%$. Here the X coordinate is the distance from the source of pollution (located in the western shore, 1.0 km from the reservoir), km; Z – height above the water surface, m; i.e. indicators of fog water content (g/kg) (numerator) and turbulent exchange coefficient K_z (m²s) (denominator) at 2, 4, 6, 8, 10, 12, 14 and 16 km from the source of pollution and at 2, 25, 50, 75, 100, 125 and 150 m above the surface of the reservoir. (for example, at 2 km in length and 2 m in height from the source of pollution, the water content of the fog is 0.05 g/kg, and the turbulent exchange coefficient is $K_z=2,7\text{m}^2\text{s}$)

From this table, it follows that in a fog the concentration of pollutants in the atmosphere decreases sharply due to its dissolution in drops. Although the levels of 75 and 100 m are located above the fog, and at these levels, the concentration decreases several times. Thus, the presence of fog leads to the fact that not only the polluting impurity that would be in the vicinity of the underlying surface in the absence of fog is concentrated in its droplets, but also a significant part of the impurity from the higher-lying and, in this case, the most polluted levels. Thus, the fog droplets accumulate an impurity from the extended layer, which significantly increases the total pollution of the atmosphere in the vicinity of the underlying surface. This is significantly affected by an increase in the temperature of the fog, which is formed by the release of latent heat of condensation. The maximum temperature in the fog above the surrounding atmosphere reaches about 3-4.5 °C.

From [6-34] it follows that we use the concept of total concentration $s_n=S=S_f$, where S_f is the concentration of contaminants a in the fog droplets, calculated per unit volume of an air mass. From the numerical calculation, it follows that in the area of moisture condensation, the total concentration of contaminants is higher than that which would be here in the absence of fog. Above the fog, on the contrary, the total concentration equal to the concentration of the impurity in the atmosphere is less than the concentration that would be observed here in the absence of fog.

The structural features of the atmospheric boundary layer with developed advective radiation fogs are a blocking temperature inversion (inversion "cap") at their upper boundary, and the atmosphere is neutrally stratified. In the presence of temperature inversions, there is a “smoke” of the air from the operating source.

Table 2. Impurity concentration in fog conditions (denominator) and without it (numerator)

X, km	Height, m									
	0.5	2	4	10	30	50	100	150	200	250
2	0.08/0.17	0.05/0.12	0.02/0.09	-/0.09	-/0.02	-/-	-/-	-/-	-/-	-/-
4	0.15/0.29	0.07/0.22	0.03/0.016	0.01/0.12	-/0.08	-/0.02	-/-	-/-	-/-	-/-
6	0.53/0.48	0.48/0.36	0.41/0.32	0.39/0.27	0.25/0.10	0.17/0.03	0.11/-	0.09/-	-0.32/-	0.08/-
8	0.55/0.49	0.49/0.37	0.47/0.36	0.43/0.31	0.38/0.20	0.27/0.12	0.19/0.04	0.14/-	0.09/-	0.08/-
10	0.56/0.49	0.50/0.39	0.49/0.37	0.44/0.32	0.38/0.21	0.27/0.12	0.20/0.05	0.14/-	0.10/-	0.06/-
12	0.41/0.39	0.30/0.32	0.25/0.30	0.21/0.28	0.12/0.15	0.07/0.09	0.06/0.01	0.04/-	0.02/-	0.01/-
14	0.30/0.34	0.21/0.28	0.12/0.27	0.08/0.21	0.03/0.10	0.02/0.07	0.05/-	0.04/-	0.01/-	-/-

Table 2 shows the impurity concentration in fog conditions and in absence of fog. Here the X is the distance from the source of pollution located in the western bank of 1.0 km from the reservoir, km; Z is the height above the water surface, m; i.e. impurity concentration in fog conditions (denominator) and without it (when there is no fog) (numerator) at 2, 4, 6, 8, 10, 12 and 14 km from the source of pollution and at 0,5; 2; 4; 10; 30; 50; 100; 150; 200 and 250 m above the water surface (for example, at 2 km in length and 0.5 m in height from the source of pollution, the concentration of impurity in fog conditions is 0.008 mg/m², and in the absence of fog 0.17 mg/m²).

Let us present the results of another experiment. We consider the important features of impurity scattering in the presence of a raised inversion located directly above the source, and first, we neglect the absorption of the impurity by water droplets of fog. These calculations were used for purposes. First, for this type of fog, the raised inversions are a consequence of their existence, and it was of interest to evaluate the quantitatively related changes in concentration. Secondly, and this is the main thing, as in the case of fogs, it is possible to judge the direct effect of the absorption of impurities by fog droplets on the basis of comparison of calculations with neglect and taking into account this effect in the elevated inversion of temperature over the source to concentrations in its absence. It was assumed that $a = 0$; the height of the source located at the lower boundary of the inversion is 100 m, the wind speed is $V_i = 2$ m/s.

It is known that in the absence of inversion near the source, the maximum concentrations are observed at $z = H$, and the concentrations decrease approximately symmetrically; with distance from the source, they decrease approximately symmetrically; with distance from the source, the vertical concentration gradients decrease, and the line of maximum concentrations gradually approaches the underlying surface.

The presence of a raised inversion above the source leads to asymmetry of the vertical distribution of concentration at close distances due to a sharp increase in air pollution in the sub-inversion layer, where the main part of the impurity is concentrated. At a sufficiently large horizontal distance from the source, the impurity gradually penetrates the lower part of the inversion, and it also becomes contaminated.

Table 3. The ratio of the concentration of impurities in the presence of inversion and without it

Height, m	Distance from source, m				
	2	25	50	75	100
2	0.05/2.7	0.09/5.4	0.11/7.9	0.08/9.3	0.01/4.4
4	0.12/3.2	0.14/6.2	0.18/8.9	0.11/9.4	0.02/4.5
6	0.19/4.7	0.24/7.1	0.25/9.3	0.17/13.5	0.08/15.5
8	0.30/5.1	0.32/9.2	0.27/11.9	0.17/17.1	0.08/18.2
10	0.32/6.2	0.36/10.3	0.25/12.2	0.16/16.4	0.04/15.1
12	0.09/7.1	0.12/10.4	0.14/12.8	0.14/12.9	0.10/11.7
14	0.05/7.2	0.9/7.4	0.10/7.9	0.05/7.8	0.01/9.1
16	0.03/7.0	0.08/7.2	0.06/7.7	0.05/7.7	0.01/7.6

The source of pollution is located in the western shore of 1.0 km from the reservoir i.e. impurity concentration in the presence of inversion (denominator) and in the absence of inversion (when there is no inversion) (numerator) at 2, 4, 6, 8, 10, 12 and 14 m in height and 2, 25, 50, 75 and 100 m in length from the source of pollution (for example, at 2 m in length and 2 m in height from the source of pollution, the concentration of impurity in inversion conditions is $0,05 \text{ mg/m}^2$, and in the absence of inversion $2,7 \text{ mg/m}^2$)

The calculations showed that in the case under consideration, a decrease in the concentration of the impurity is observed at all levels, and the layer of elevated inversion is also cleared, in which, as indicated, at a large distance from the source, in the absence of fog, a part of the impurity is concentrated. This confirms the conclusion made earlier in the calculations of the effect of river fogs on the diffusion of impurities from high-altitude sources about the "accumulation" of impurities by fog from the overlying layers, leading to an increase in the total concentration at levels located in the fog and a decrease in the concentration of gaseous impurities above it. The presence of fog under inversion leads to the fact that the impurity does not penetrate into the inversion layer even at large distances from the source and almost all of it passes into water droplets, turning them into a solution of harmful substances. As another example, we describe the calculation results under the same meteorological conditions, but at a

source height of $H = 150$ m. In this case, the source was located inside the inversion layer at a distance of 50 m from its lower boundary. Due to weak turbulence at heights, the impurity scatters slowly and moves to fog over large distances in the form of a concentrated torch. Only a small distance from the source drops to the ground. The presence of a layer of weak turbulence between the source and the fog prevents a noticeable accumulation of impurities from the overlying layers, although, as in the cases discussed above, most of the impurities entering the fog dissolve in its droplets.

Of considerable interest is the study of the effect of fog on the diffusion of sulfur dioxide (SO_2), which is one of the most common harmful pollutants in the atmosphere. The conversion of sulfur dioxide can be carried out in a heterogeneous zone. By heterogeneous transformation, we mean a chemical reaction that occurs not in the gas phase, but in droplets or on the surface of particles in the atmosphere. It should be noted, another interesting way to convert sulfur dioxide. During the combustion of carbonaceous fuel, highly dispersed soot particles are formed on the surface of which sulfur dioxide is adsorbed. This leads to structural changes in the sulfur dioxide molecule, contributing to its transformation into sulfate. This transformation occurs more intensively if the number of soot particles entering the air increases, for example, when the heating is turned on. The dissolution of sulfur dioxide in fog droplets leads to the formation of an aerosol of sulfurous acid. The latter, in comparison with sulfur dioxide, has greater toxicity, in its presence in the atmosphere, corrosion of metal objects is significantly increased.

In addition, sulfur dioxide dissolved in fog droplets is significantly faster oxidized to sulfuric anhydride than sulfur dioxide in the gaseous state. This is due to the fact that usually fog droplets contain some trace elements that have catalytic properties, in their presence, oxidation is more intense. In view of the fact that sulfuric anhydride in interaction with water forms sulfuric acid, it is possible for simplicity to talk about the partial oxidation of sulfur dioxide in the atmosphere to sulfuric acid.

In the next version of the calculation, the influence of the earth's topography on the dispersion of pollutants over a freezing reservoir in fog conditions is considered. The relief is represented by a single, perpendicular to the flow, and the water temperature is 15°C higher relative to the ambient temperature. Let's set the horizontal velocity flow $U_f=5\text{m/s}$, $V_f=0$ m/s, with background stratification $S=0.053^\circ\text{C/m}$ at the input of the region. On the left bank, there is a continuously operating pollution source with a height of $H_s=100\text{m}$.

The evolution of fog over the water surface is accompanied by a temperature inversion which at midnight is located at an altitude of $\sim 300\text{m}$, under it, the atmosphere has an unstable stratification. During this period the fog has a large capacity and is localized over the surface of the reservoir.

From Figure 1 we can conclude that the flow almost does not flow around the characteristic relief of the basin and does not form recirculation flows, moving over the mountain valley and practically without distortion. Thus, the calculations confirm that in mountain circuses or near ridges under conditions of blocking inversions of the atmosphere, the impurity can become locked and remain practically motionless for a long time regardless of the external flow. But with the prolonged action of high background flow velocities, it is set in motion and a recirculation exchange is formed in the valley due to the flow energy and turbulence of diffusion of the underlying levels of the locking inversion. The absorption of gaseous pollutants by fog droplets occurs mainly in the upper layer of fog, and in the vicinity of reservoir X, the concentration is close to zero.

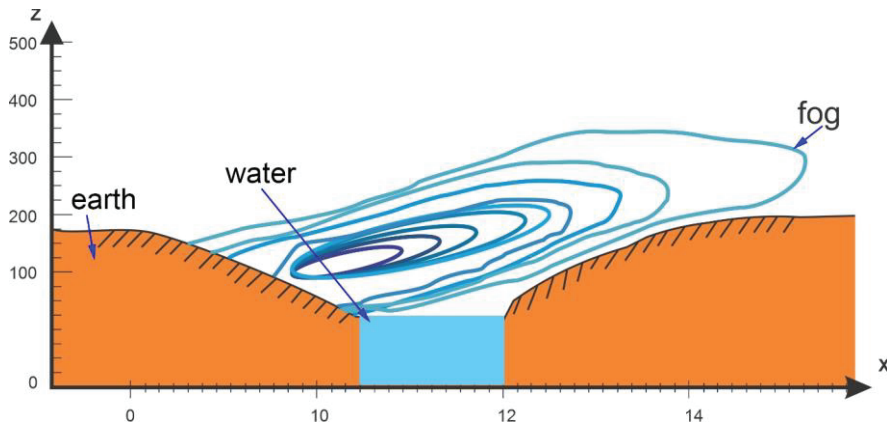


Fig. 1. Impurity distribution in fog conditions in the presence of a complex relief, the X coordinate is the distance from the source of pollution, Z is the height above the water surface, both axes in meters.

For a number of harmful substances, the toxicity of which increases in solutions so quickly dissolving them in droplets leads to a sharp increase in polluted air under conditions of strong fog. If the source of the impurity is on the shore of the reservoir, it does not let the impurity into a steadily stratified layer. The impurity propagates, scattering relatively weakly, until it crosses the boundary of the inner boundary layer at some distance from the pipe. After this, a rapid vertical dispersion of the impurity occurs, and it reaches the underlying surface. This process is called smoke, and is the greatest danger of the environmental plan.

The distribution of the contaminants in areas with complex orography under fog conditions is very different from the scattering on the plain, which can lead to an erroneous assessment of pollution in the area of industrial facilities.

The differences are due to the action of a number of physical phenomena: the appearance of an internal mixing layer, stagnant zones with increased turbulence and the formation of fog with high power. If for such areas to use the existing methods of calculation for flat terrain, the calculated values of the concentration may differ greatly from the measured several times.

In some urban smog, intensification of the oxidation of sulfur dioxide to sulfuric acid occurs in the daytime during photochemical reactions that occur under the influence of solar radiation. According to the data, the strongest oxidation is observed here with a weak wind, temperature inversion at low altitudes and a large number of oxidizers; in this case, 20% of the sulfur contained in the sulfur dioxide emitted into the atmosphere will be oxidized to sulfuric acid. The relative amount of oxidized sulfur dioxide decreases with increasing concentration.

Attention should be paid to the increase in the weight concentration of harmful impurities that occurs during the interaction of sulfur and sulfur anhydrides with water and the formation of sulfur and sulfuric acids, respectively. So, for example, during the oxidation of 1 g of sulfurous anhydride (molecular weight 64), 1.5 g of sulfuric acid is formed (molecular weight 98).

4 Conclusions

A number of factors that can significantly increase atmospheric pollution from fog and their toxicity in the case of a single source have been noted above. When several sources are located along a certain line or dispersed over an area, under conditions of elevated temperature inversion, which is often observed during fogs, additional reasons of increasing pollution of the surface air layer may appear. Under such conditions, as shown by theoretical calculations [3], the distance at which the maximum concentration is observed increases substantially, and the concentration decreases very slowly with the distance after its maximum. As a result, the

influence of the mutual overlapping of concentration fields from individual sources increases and a more uniform pollution than in convective conditions is created in the industrial area. At the same time, even if the concentrations from a single source are relatively small, the total pollution can be very significant.

A certain role in the pollution of surface air layers can be played by the settling of large droplets of fog, in which a dissolved impurity from overlying and often very contaminated layers is transferred to the underlying surface falling drizzle, the droplets of which contain appreciable concentrations of sulfuric acid. This phenomenon has been called acid rain.

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