

BASIC MATHEMATICAL MODELING OF THE ENVIRONMENTAL PROCESSES IN THE LOWER DANUBE REGION [I]

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ABSTRACT

In most environmental pollution problems, the pollutant is released to the environment by the, almost always, turbulent flow of a carrier fluid. The pollutant mixes with the surrounding fluid (air or water) and undergoes chemical transformations. A proper account of "where the pollutant went" and "what happened to it" necessitates a theory of turbulent reacting flows, i.e. the simultaneous treatment of mixing and chemical reactions. The important field of Air Quality Modelling is an interdisciplinary field borrowing elements from Fluid Mechanics, Atmospheric Chemistry, Meteorology and others.

KEYWORDS: Air Quality Modelling, Fluid Mechanics, Meteorology, turbulent flow

1. Introduction

1.1 Atmospheric dispersion models [1]

Atmospheric dispersion models are computer programs that use mathematical algorithms to simulate how pollutants in the ambient atmosphere disperse and, in some cases, how they react in the atmosphere. The dispersion models are used to estimate or to predict the downwind concentration of air pollutants emitted from sources such as industrial plants and vehicular traffic.

2. Air Quality Modelling and plume dispersion

2.1 Box models [2]

Consider the following Fig. 2.1: where we have enclosed a whole city in a control volume. Assume that the air in the box is **fully uniform** in concentration and that there is uniform wind of velocity U flowing along the x-direction. Assume that the box extends up to the mixing height H . Assume that there is background pollution b (a convenient and common unit is in kg of pollutant per m^3 of air) that is being advected towards the city by the wind and

that the city itself generates q kg/m^2 /s of the pollutant.

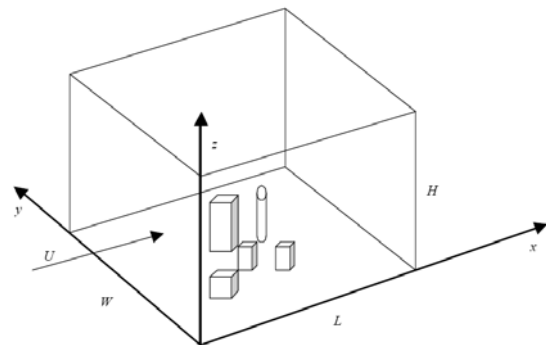


Fig. 2.1

Then, the conservation of mass for this pollutant gives for its concentration c in the box (in the same units as b):

$$WLH \frac{dc}{dt} = UWHb - UWHc + qWL + WHL\dot{w}_c \quad 2.1$$

$$\Leftrightarrow \frac{dc}{dt} = (b - c) \frac{U}{L} + \frac{q}{H} + \dot{w}_c \quad 2.2$$

The l.h.s. of Eq. (2.1) is the unsteady accumulation of the pollutant. The first term on the r.h.s. is the amount of pollutant advected *into* the box by the wind; the second term is the amount advected *out* of the box (note that what is being advected out has concentration c , the concentration of the wellmixed box); the third term shows how much c per unit time is released in the city (e.g. by factories or cars); the last denotes how much c is being generated by chemical reactions (e.g. by transformations from other pollutants). The reaction rate w_c has units kg of pollutant per unit volume. Very often we are interested only in the steady state, i.e. $dc/dt = 0$. Let us also neglect reactions, which is a good approximation if the particular pollutant reacts very slowly compared to the residence time L/U . Then, Eq. (2.2) gives that the pollutant will be in a concentration c_{bm} above the city, given by:

$$c_{bm} = b + \frac{qL}{UH} \quad 2.3$$

This is the "standard" box model result used in Air Quality Modelling practice and hence the subscript *bm* in Eq. (2.3). Equation (4.3) involves many assumptions, the most important of which are that the pollutant is uniformly distributed in the box, that the wind is uniform (despite the boundary layer!), and that the emission is uniformly distributed across the area of the city. Clearly, none of these assumptions is really justified. Nevertheless, Eq. (2.3) shows the correct scaling with H and U : low mixing heights and low winds imply a higher concentration of pollutant. Note also that the local city meteorology affects the pollution concentration through the wind U and the mixing height H . Hence we expect a larger concentration of pollutant at night (small H , small U) than at day, although this may be counterbalanced by the higher emissions during daytime (e.g. from traffic).

Emphasis on chemistry

Equation (2.2) is not restricted to a single pollutant. Various researchers use it for examining also the chemistry and emphasize the reaction rate term, rather than the wind transport. In such box models, the governing equation for each species i becomes:

$$\frac{dc_i}{dt} = \dot{w}_i \quad 2.4$$

Equation (2.4) is solved by numerical methods subject to a particular set of initial conditions and the solutions can help identify how the various pollutants are transformed during the day. Comparisons with experimental data can then assist in developing chemical mechanisms, such as "tuning" the rates of the various reactions like the smog-forming reactions. Developing detailed chemical mechanisms for atmospheric pollution is a very

active research area at present, not least because more and more chemicals come under regulation.

Emphasis on inhomogeneity

It is not a bad assumption to take the pollutant concentration as uniform in the z -direction, especially during unstable stratification. However, the assumption of homogeneous concentration in the wind direction is usually much worse because often q is a function of x . This can be partly dealt with by re-deriving Eq. (2.2) for a thin strip of thickness Δx and hence obtaining a differential equation for dc/dx .

Emphasis on yearly averages

Very common in Air Quality Modelling, Eq. (2.3) is used for a range of wind directions and speeds and a range of mixing heights, so that various meteorological conditions can be examined to find the corresponding pollutant concentrations. These are then weighted by the probability of occurrence of these particular conditions and hence a yearly average pollutant concentration can be calculated. Such calculations are important, e.g. for planning anti-pollution measures, for calculating the extra environmental burden of new industrial plants, etc.

2.2 Gaussian dispersion models

Model problem

The paradigm problem concerning pollution relatively close (e.g. a few km) to a source is the „chimney plume”. This is shown on the following Fig. 2.2:

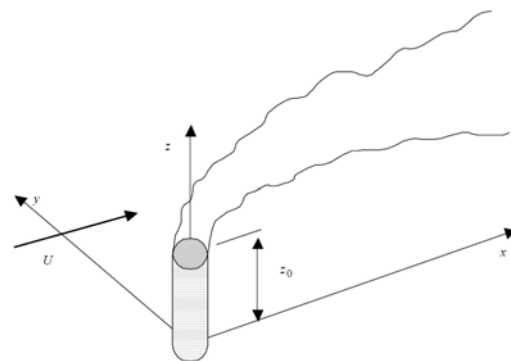


Fig. 2.2.

We are interested in: (a) the width of the plume downwind; (b) the concentration of the pollutant across the plume and particularly on the ground; (c) the difference between a steady emission (a „plume”) and an unsteady emission (a „puff”). To calculate these items is very important from a practical point of view and forms the topic of this chapter [4].

Governing equation for a reacting scalar

Conservation of mass

Consider an infinitesimal control volume ΔV . Inside the CV we have a uniform mixture of species undergoing chemical reactions.



Mass may cross the surfaces of the CV. For simplicity of presentation we assume a one-dimensional geometry.

Then, the principle of mass conservation of each species i reads:

[Rate of accumulation] = [Rate at which species comes in] – [Rate at which species leaves] + [Rate of generation due to reaction]

In mathematical terms:

$$\frac{\partial(m_V Y_i)}{\partial t} = m_i'' \Delta y \Delta z - (m_i'' + \Delta m_i'') \Delta y \Delta z + \dot{w}_i \quad 2.5$$

where: m_V (Kg): total mass of mixture inside the CV,

$$m_V = \rho \Delta x \Delta y \Delta z$$

Y_i : mass fraction of i ; ρ (Kg/m³): mixture density; m_i'' (Kg/m²/s): mass flow of species i per unit time per unit surface, the **mass flux**; \dot{w}_i (Kg/m³/s): mass of species generated per unit volume per unit time due to chemical reactions

Letting Δx go to zero, we obtain the **species conservation equation**:

$$\frac{\partial(\rho Y_i)}{\partial t} = -\frac{\partial m_i''}{\partial x} + \dot{w}_i \quad 2.6$$

The mass flux m_i'' for each species that appears in the species conservation equation is composed of two parts: an **advective** and a **diffusive** part.

$$m_i'' = m_{i,ADV}'' + m_{i,DIFF}'' \quad 2.7$$

The advective mass flux is due to the bulk fluid motion and is given by:

$$m_{i,ADV}'' = Y_i m'' = Y_i \rho u \quad 2.8$$

The diffusive mass flux is given by **Fick's Law**:

$$m_{i,DIFF}'' = -\rho D \frac{\partial Y_i}{\partial x} \quad 2.9$$

Fick's Law states that the mass flux is proportional to the gradient of the mass fraction of the species. This is a diffusion process because it tends to make concentration gradients more uniform, i.e. it mixes the various species together. The coefficient D (m²/s) is the **diffusion coefficient** and, in general, depends on the nature of the diffusing species. For gases, it is a common approximation that the diffusion of heat and mass follow the same rate, i.e. D is related to the conductivity λ :

$$\rho D = \frac{\lambda}{c_p} \quad 2.10$$

Final instantaneous species conservation equation

With these expressions, the species conservation equation takes the final form:

$$\frac{\partial(\rho Y_i)}{\partial t} + \frac{\partial(\rho u Y_i)}{\partial x} = \frac{\partial}{\partial x} \left(\rho D \frac{\partial Y_i}{\partial x} \right) + \dot{w}_i \quad 2.11$$

It is important to know the physical mechanisms contributing to this equation: the first term in the l.h.s. corresponds to accumulation of species i , the second to advection by the bulk fluid motion, the first term in the r.h.s. corresponds to molecular diffusion and the last to the generation by the chemical reactions.

In more dimensions and for a generic scalar ϕ that is proportional to the mass fraction (e.g. our usual concentration in atmospheric pollution expressed in kg/m³), the governing transport conservation equation becomes:

$$\frac{\partial \phi}{\partial t} + u_j \frac{\partial \phi}{\partial x_j} = D \frac{\partial^2 \phi}{\partial x_j^2} + \dot{w} \quad 2.12$$

in Cartesian tensor notation, where we have assumed an incompressible flow and a constant diffusivity, typically excellent assumptions in environmental fluid mechanics. If the scalar is inert, then simply $\dot{w} = 0$. Equation (2.12) is the starting point for examining turbulent mixing.

Averaged species conservation equation

In a turbulent flow, we can write that the instantaneous mass fraction of a scalar is $\phi = \bar{\phi} + \phi'$ and that the velocity is $u = \bar{u} + u'$. It is easy to see that, by performing Reynolds decomposition and performing the averaging procedure on the Eq. 2.12 we get:

$$\frac{\partial \bar{\phi}}{\partial t} + \frac{\partial(\bar{u}_j \bar{\phi})}{\partial x_j} + \frac{\partial(\overline{u'_j \phi'})}{\partial x_j} = D \frac{\partial^2 \bar{\phi}}{\partial x_j^2} + \bar{w} \quad 2.13$$

The first term in the l.h.s. is the unsteady accumulation of ϕ , the second is due to mean advection, and the third is due to turbulent transport (or turbulent diffusion). The first term in the r.h.s. is due to molecular diffusion and the second is the mean reaction rate.

Modelling the scalar flux – the eddy diffusivity

It is usual engineering practice to model the turbulent transport term using the **eddy diffusivity** concept, also known as the **gradient approximation**. This model is motivated from the Kinetic Theory of Gases, where the mass flux is found to be proportional to the gradient of the mass fraction (Eq. 2.9) and the molecular diffusivity D is found to be proportional to the mean molecular speed and the mean free path between molecular collisions.

By making an analogy between the random turbulent motions of "fluid particles" and the random molecular motion in a fluid, the turbulent transport term is written as:

$$\overline{u'_j \phi'} = -D_T \frac{\partial \bar{\phi}}{\partial x_j} \quad 2.14$$

with the eddy diffusivity D_T given by

$$D_T = C u' L_{turb} \quad 2.15$$

By a trial-and-error procedure and comparison with experimental data, the constant C is found to be around 0.1, but this depends on how L_{turb} is defined. There is a lot of criticism behind the use of the gradient approximation for modelling turbulent transport and indeed sometimes Eqs. (2.14) and/or (2.15) fail to predict the correct magnitude of mean $(u'_j \phi')$. Nevertheless, the eddy diffusivity concept remains a very useful approximation for providing a tractable closure to Eq. (2.13), which then becomes:

$$\frac{\partial \bar{\phi}}{\partial t} + \bar{u}_j \frac{\partial \bar{\phi}}{\partial x_j} = \frac{\partial}{\partial x_j} \left[(D + D_T) \frac{\partial \bar{\phi}}{\partial x_j} \right] + \bar{w} \quad 2.16$$

Note that D_T may be a function of space and hence should be kept inside the derivative in the r.h.s. of Eq. (2.16). The eddy diffusivity concept is usually much better for an *inert* scalar than for a *reacting* scalar, but we use it anyway.

For high Reynolds numbers, $D \ll D_T$, which suggests that the molecular diffusion may be neglected. To illustrate this, consider a wind flow of 5 m/s with a typical turbulence intensity of 10%, so that $u' = 0.5$ m/s. In the atmospheric boundary layer, the lengthscale is proportional to the height above the ground. Let us take that $L_{turb} = 500$ m. Then $D_T = 25$ m²/s. At standard temperature and pressure, the molecular diffusivity of air is 2.2×10^{-5} m²/s (see Eq. 2.10). Therefore the diffusion caused by molecular motions is negligible compared to the diffusion due to turbulence, which is a typical feature of turbulent flows at large Reynolds numbers. Molecular action is *always* present at the smallest scales, but these contribute very little to the overall diffusion of the scalar (the small eddies just don't "move far enough"). In other words, "where the smoke goes" is a function of the large scales only and the turbulent diffusivity suffices.

Turbulent diffusion of an unsteady puff

In many cases, we are interested in the way a pollutant spreads under the action of turbulent diffusion, when the emission of the pollutant is not continuous, but occurs only for a short time. Our full governing equation (Eq. 2.16) is our starting point, but to demonstrate how the solution comes about, let us assume zero mean velocities and finite and spatially-uniform turbulent diffusivity.

Then, Eq. (2.16) becomes:

$$\frac{\partial \bar{\phi}}{\partial t} = K_j \frac{\partial^2 \bar{\phi}}{\partial x_j^2} + \bar{w} \quad 2.17$$

The overbar denotes the mean concentration of the pollutant and the eddy diffusivity is now allowed to be a function of the direction. This is usually the case in atmospheric turbulence: the velocity fluctuations are not really equal in the three directions, and hence the eddy viscosity is strictly speaking a (diagonal) tensor. Hence the appearance of the subscript j in K in Eq. (2.17). We have also denoted the turbulent diffusivity by the symbol K (rather than D_T) to conform to the standard notation in atmospheric pollution.

For an inert pollutant, Eq. (2.17) is identical to the unsteady heat conduction equation for an "instantaneous source" and has a known solution. If Q kg/s of pollutant are released over a (very short) time Δt at point (x_0, y_0, z_0) , the solution of Eq. (2.17) gives for the mean pollutant concentration $\bar{\phi}$ in one, two, and three dimensions (in kg/m, kg/m², kg/m³ respectively):

$$\bar{\phi}(x) = \frac{Q\Delta t}{2(\pi)^{1/2}(K_x)^{1/2}} \exp\left[-\frac{1}{4t} \frac{(x-x_0)^2}{K_x}\right] \quad 2.18$$

$$\bar{\phi}(x, y) = \frac{Q\Delta t}{4(\pi)(K_x K_y)^{1/2}} \exp\left[-\frac{1}{4t} \left(\frac{(x-x_0)^2}{K_x} + \frac{(y-y_0)^2}{K_y}\right)\right] \quad 2.19$$

$$\bar{\phi}(x, y, z) = \frac{Q\Delta t}{8(\pi)^{3/2}(K_x K_y K_z)^{1/2}} \exp\left[-\frac{1}{4t} \left(\frac{(x-x_0)^2}{K_x} + \frac{(y-y_0)^2}{K_y} + \frac{(z-z_0)^2}{K_z}\right)\right] \quad 2.20$$

where t is the time from the release. Note the "symmetry" of the terms in the exponential, but also note that the behaviour of the maximum concentration at the centre of the cloud (i.e. at x_0, y_0, z_0) has a different scaling with time depending on the dimensionality of the problem.

In practical atmospheric dispersion of pollutant clouds, the wind has to be taken into account. The situation is visualized in the following Fig. 2.3.

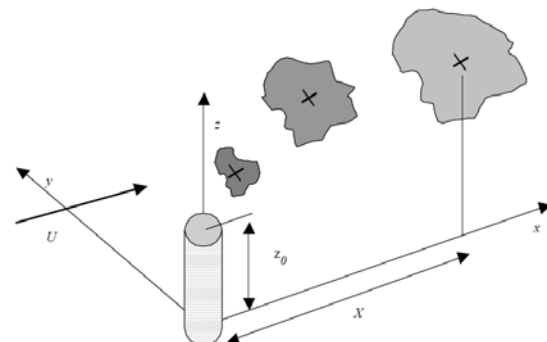


Fig. 2.3.

The governing equation becomes (for an inert pollutant):

$$\frac{\partial \bar{\phi}}{\partial t} + U \frac{\partial \bar{\phi}}{\partial x} = K_x \frac{\partial^2 \bar{\phi}}{\partial x^2} + K_y \frac{\partial^2 \bar{\phi}}{\partial y^2} + K_z \frac{\partial^2 \bar{\phi}}{\partial z^2} \quad 2.21$$



The solution of Eq. (2.21) is again Eq. (2.20), but now we must interpret the time t as the downwind distance X of the **centre of the pollutant cloud**, divided by the wind speed U , and x, y, z as the distances from the centre of the cloud. In Eulerian coordinates (more useful!), x should be replaced by $(x-X)$, with $X=Ut$. Usually, the coordinate system we use has an origin immediately underneath the release point on the ground (i.e. $x_0=y_0=0$) (Fig. 2.3) and so the "standard" form of the solution of Eq. (2.21) reads:

$$\bar{\phi}(x, y, z) = \frac{Q\Delta t}{8(\pi)^{3/2}(K_x K_y K_z)^{1/2}} \exp\left[-\frac{1}{4t}\left(\frac{(x-Ut)^2}{K_x} + \frac{y^2}{K_y} + \frac{(z-z_0)^2}{K_z}\right)\right]$$

Depending on the relative magnitude of 2.22 , the cloud may take a spherical or ellipsoidal or even a disk-like shape.

Final equation

Very often in atmospheric dispersion, we do not use the eddy diffusivities directly, but we work with the **dispersion coefficients** [5], defined by:

$$\sigma_x^2 = 2\frac{X}{U}K_x, \quad \sigma_y^2 = 2\frac{X}{U}K_y, \quad \sigma_z^2 = 2\frac{X}{U}K_z \quad 2.23$$

Using the dispersion coefficients (units: m), Eq. (2.22) becomes:

$$\bar{\phi}(x, y, z, t) = \frac{Q\Delta t}{(2\pi)^{3/2}\sigma_x\sigma_y\sigma_z} \exp\left[-\left(\frac{(x-Ut)^2}{2\sigma_x^2} + \frac{y^2}{2\sigma_y^2} + \frac{(z-z_0)^2}{2\sigma_z^2}\right)\right]$$

2.24

which is a "standard" Gaussian expression. The dispersion coefficients are usually functions of X . It is important to note that Eq. (2.24) is valid even if the eddy diffusivities K are not constant. This would be the case for short times from the release because then the constant eddy diffusivity idea breaks down: for small cloud widths, the pollutant is being dispersed by the action of only a small range of the turbulent eddies.

This makes the diffusivity a function of time, until the cloud becomes large relative to the turbulence lengthscale. In atmospheric problems, this is hardly ever achieved and the dispersion coefficients in the above equations must be provided empirically.

Equation (2.22) (or Eq. 2.24) is called the "Gaussian puff equation" and is very useful in providing numerical estimates on the average amount of pollutant reaching a given point at a given time after a release. Such estimates are crucial for assessing the danger after accidents, such as the Chernobyl nuclear disaster or chemical releases. They are also used to assess the origin of the pollutant, given a set of measurements of the pollutant concentration at a particular point and the meteorological conditions prevailing at that time.

Cloud width

Another use of Eq. (2.24) is to find the width of the cloud. This can be defined in various ways, since the concentration reaches zero asymptotically towards the edge of the cloud. A common way is to use the length:

$$\sqrt{2\sigma_y^2} = 2\sqrt{K_y X/U} \quad 2.25$$

as a characteristic half-width of the Gaussian cloud in the y -direction and similarly for the other directions. This length marks the location from the cloud centre where the concentration falls to $1/e$ of the centre value.

Turbulent diffusion of a steady plume

One way to understand the continuous emission from a chimney (Fig. 2.2) is to view it as a series of "instantaneous puffs". Based on the linearity of Eq. (2.21), the overall concentration $\bar{\phi}$ downwind would be the sum over all such puffs. Assume then that the chimney continuously releases Q kg/s of pollutant and we use $t=x/U$. We are interested in the pollutant concentration at a particular point (x, y, z) at time t . Equation (2.24) can be integrated over time, to give the mean concentration due to a continuous source:

$$\bar{\phi}(x, y, z) = \frac{Q}{(2\pi)^{3/2}} \int_0^\infty \exp\left[-\left(\frac{(x-Ut')^2}{2\sigma_x^2} + \frac{y^2}{2\sigma_y^2} + \frac{(z-z_0)^2}{2\sigma_z^2}\right)\right] \frac{1}{\sigma_x\sigma_y\sigma_z} dt' \quad 2.26$$

It is important to remember that we are now dealing with diffusion at **early times**, i.e. we are interested in the region close (e.g. of the order of 10 to 104 m) to the plume source. There, the width of the plume is clearly not much greater than the turbulence length scale, which is one of the conditions for validity of the constant eddy diffusivity. For short distances from the source, it turns

out that the dispersion coefficients are proportional to time:

$$\sigma_x = u't, \quad \sigma_y = v't, \quad \sigma_z = w't \quad 2.27$$

where u' , v' , w' are the r.m.s. turbulent velocities in the three directions, assumed constant. Equation (2.26) can then be integrated to give:

$$\bar{\phi}(x, y, z) = \frac{Q}{(2\pi)^{3/2}} \frac{u'}{v'w'r^2} \exp\left(-\frac{U^2}{2u'^2}\right) \left[1 + \sqrt{\frac{\pi}{2}} \frac{Ux}{u'r} \exp\left(\frac{U^2 x^2}{2u'^2 r^2}\right) \operatorname{erfc}\left(-\frac{1}{\sqrt{2}} \frac{Ux}{u'r}\right)\right] \quad 2.28$$

with

$$r^2 = x^2 + \frac{u'^2}{v'^2} y^2 + \frac{u'^2}{w'^2} (z-z_0)^2$$

Usually, the plume is *slender*, which implies that $r \approx x$, and usually $u'/U = \theta$ ($0.1 \ll 1$). Both these assumptions are used to simplify Eq. (2.28) into:

$$\bar{\phi}(x, y, z) = \frac{Q}{2\pi U \sigma_y \sigma_z} \exp\left[-\left(\frac{y^2}{2\sigma_y^2} + \frac{(z-z_0)^2}{2\sigma_z^2}\right)\right]$$

2.29

This is the final result for the plume diffusion problem. It is often called the "Gaussian plume equation". The x dependence comes indirectly through the dependence of the dispersion coefficients on x , while the dependence on y and z comes directly from the exponential.

2.3 Effects of Lower Danube region meteorology [6]

Effect of inversions

We mentioned previously that the case of one-dimensional diffusion in the horizontal direction approximately corresponds to the case where the plume is perfectly mixed in the vertical direction. If the mixing height is higher than the source height, i.e. if $H > (h_s + h_{pr})$, then there is every possibility that the plume will hit the inversion lid from below, from where it will be reflected downward etc. This situation is shown in the following figure (Fig. 2.5), which also demonstrates how the image source idea can be used to account for the mixing height. It is clear that the vertical direction will eventually become quite homogeneous.

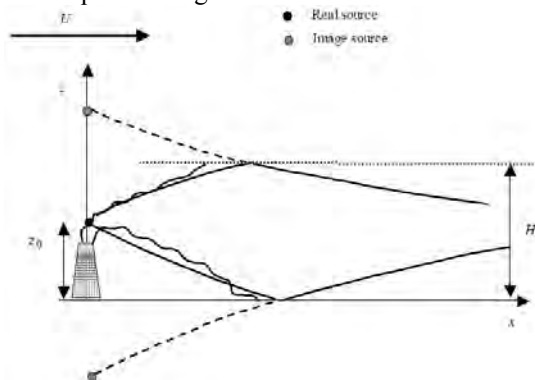


Fig 2.5. Effect of inversion lids on diffusion (source below the inversion)



Fig. 2.6. Pollution above Galati city

Low mixing heights will result in higher ground concentrations.

Inversions assist in "trapping" pollution above, for example Galati city, which can be thought as composed of a very large number of point sources. In addition to more intense photochemistry, these

conditions are favorable for the creation of inversion lids and hence such city encounter high levels of pollution often, see the following Figure 2.6.

2.4 Practical Air Quality Modelling

In previous chapter, we presented the "Box Model", which is a very useful tool for estimating the pollution above a city. Such box models are used in scales of a few km. If one is interested in finer scales (e.g. a few hundred meters), and this may be imperative for accurate calculations of dosage, e.g. from a new industrial plant or a planned motorway, then we need to consider the dispersion very close to the source. For uniform wind conditions and well-defined sources, the Gaussian plume theory is approximately correct for up to a few tens of km, but eventually our assumption of constant wind will become invalid. For *synoptic scales*, i.e. longer than 200 km, we need to couple our pollution dispersion models with proper meteorological (i.e. weather prediction) codes like Eulerian codes and Lagrangian codes. An atmospheric dispersion model [3] based on atmospheric boundary layer turbulence structure and scaling concepts including treatment of multiple ground-level and elevated point, area and volume sources is AERMOD. It handles flat or complex, rural or urban terrain and includes algorithms for building effects and plume penetration of inversions aloft seen in Figure 2.7 [6]



Fig. 2.7. SO₂ concentrations in Galati area modeled with AERMOD

3. Conclusions

The dispersion models require the input of data which includes:

- Meteorological conditions such as wind speed and direction, the amount of atmospheric turbulence (as characterized by the stability class), the ambient air temperature and the height to the bottom of any inversion aloft that may be present;



- Emissions parameters such as source location and height, source vent stack diameter and exit velocity, exit temperature and mass flow rate;
- Terrain elevations at the source location and at the receptor location;
- The location, height and width of any obstructions (such as buildings or other structures) in the path of the emitted gaseous plume.

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