Advection-diffusion model for the simulation of air pollution distribution from a point source emission

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Abstract. Advection-diffusion model is one of the mathematical models, which can be used to understand the distribution of air pollutant in the atmosphere. It uses the 2D advection-diffusion model with time-dependent to simulate air pollution distribution in order to find out whether the pollutants are more concentrated at ground level or near the source of emission under particular atmospheric conditions such as stable, unstable, and neutral conditions. Wind profile, eddy diffusivity, and temperature are considered in the model as parameters. The model is solved by using explicit finite difference method, which is then visualized by a computer program developed using Lazarus programming software. The results show that the atmospheric conditions alone influencing the level of concentration of pollutants is not conclusive as the parameters in the model have their own effect on each atmospheric condition.

1. Introduction

Air quality recently has been a global problem, because the air has polluted by human activities and natural processes. [1] reported that in 2012, air pollution exposure was responsible for 7 million premature deaths and this occurs annually. Pollution is able to affect an area far from the source of emission that is caused by a bulk motion of wind. The focus of this article is a source of air pollution from a point source emission such as a chimney. The distribution of air pollution can be studied through mathematical simulation which is meteorological parameters can be observed as they have an effect on the distribution of air pollution. [2] stated that simulation of air pollution provides information about the spread of pollutants in an area, the scale, and level of pollution and estimation.

Modelling and simulation of air pollution distribution are to observe the effects meteorological factors such as the wind, temperature, humidity, pressure, etc. have on the dispersal of pollutants. Reference [3] did a study on air pollution dispersion using a steady state two-dimensional mathematical model. They considered mesoscale wind, which is generated by urban heat island. Heat from the Earth’s surface results in the vertical movement and it mixes with the mean wind causing turbulence. Eddy diffusivity plays an important role in turbulence. The strength of turbulence determines the atmospheric stability, whether it is stable, neutral or unstable. The higher the turbulence; the more the atmosphere becomes unstable.

2. Model Development

The advection diffusion model is used to simulate air pollution distribution, which describes the physical processes involved in the dispersal of pollutants. Reference [4] has shown that the dispersion of pollutant
can be expressed by the following equation with time dependent:

\[
\frac{\partial C}{\partial t} + U \frac{\partial C}{\partial x} + V \frac{\partial C}{\partial y} + W \frac{\partial C}{\partial z} = \frac{\partial}{\partial x} \left( K_x \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial y} \left( K_y \frac{\partial C}{\partial y} \right) + \frac{\partial}{\partial z} \left( K_z \frac{\partial C}{\partial z} \right) + S
\]  

(1)

where,

\( C = C(x, y, z, t) \) is the pollutants concentration at any location \((x, y, z)\) at time \(t\);

\( U, V \) and \( W \) are the wind profile in downwind \((x)\), crosswind \((y)\) and vertical \((z)\) direction respectively;

\( K_x, K_y \) and \( K_z \) are eddy diffusivities in the downwind \((x)\), crosswind \((y)\) and vertical \((z)\) direction respectively;

\( S \) is the source or sink term of the air pollutants.

In the physical processes, advection carries the pollutants in the \(x\) - direction and transports them as far as the wind blows. The mixture of the mean wind and the heat from the Earth’s surface together with friction from buildings create a local wind called mesoscale wind. It is considered that the mean wind speed changes with increasing height and the mesoscale wind which is a function that depends on temperature. Diffusion takes place alongside advection and this reduces the concentration of the pollutants by turbulence. It is assumed that the mean concentration of pollutants is constant along the \(y\) - direction, thus reducing it to a two-dimensional problem. It is again considered that the wind profile and eddy diffusivity are constant during pollutants emission. Therefore, Eq. (1) becomes:

\[
\frac{\partial C}{\partial t} + U \frac{\partial C}{\partial x} + W \frac{\partial C}{\partial z} = \frac{\partial}{\partial x} \left( K_x \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial z} \left( K_z \frac{\partial C}{\partial z} \right) + S
\]

(2)

Taking into account the removal mechanism such as precipitation, Eq. (2) can be written as

\[
\frac{\partial C}{\partial t} + U \frac{\partial C}{\partial x} + W \frac{\partial C}{\partial z} = \frac{\partial}{\partial x} \left( K_x \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial z} \left( K_z \frac{\partial C}{\partial z} \right) - \lambda C
\]

(3)

where \( \lambda \) is the removal mechanism rate.

Temperature would be considered as one of the parameters that influences the distribution of pollutants as it affects turbulence. Therefore, the change in temperature \(T\) with time would be as follows:

\[
\frac{\partial T}{\partial t} = Q_x \frac{\partial^2 T}{\partial x^2} + Q_z \frac{\partial^2 T}{\partial z^2}
\]

(4)

where \( Q_x, Q_z \) are the heat diffusion rate in the \(x\) and \(z\) directions respectively.

3. Method of Solution

3.1 The Domain

The objective is to analyze the pollutants concentration in a given two-dimensional domain \((x, z)\) at time \(t\) by developing a program using Lazarus software. Eq. (3) and Eq. (4) would be solved numerically by explicit finite difference scheme to obtain the solution. To apply the finite difference scheme, the domain \((x, z)\) is subdivided into a set of similar rectangles of sides \(\Delta x\) and \(\Delta z\), by equally spaced grid lines, parallel to the \(z\) - axis, determined by \(x = i\Delta x, i = 0, 1, 2, ..., M\) and parallel to \(x\) - axis, determined by \(z = j\Delta z, j = 0, 1, 2, ..., N\). Hence, the domain of interest belongs \(M \times N\) grids and the intersection of grid lines (grid points) determine the concentration of pollutants \(C\) and also the temperature \(T\). The entire grid would be computed with time difference \(\Delta t\). The concentration \(C\) at a grid point \((i, j)\) with time \(n\) is denoted by \(C_{i,j}^n\) and the temperature \(T\) at a grid point \((i, j)\) with time \(n\) is denoted by \(T_{i,j}^n\).
3.2 Initial and Boundary Condition
Before the system is run, the initial value is needed to set the system up. At the initial time \( t = 0 \), temperature \( T_0 \) is set in the domain. The boundary for the domain is taken and it is assumed that at the beginning when \( t = 0 \) there is pollution emission \( C_0 \) at some point in the domain. Thus, \( C(x, z, 0) = C_0 \) at \( t = 0 \) for \( 0 < x < M \) and \( 0 < z < N \) \((M - \text{horizontal and } N - \text{vertical grid points})\) and where \( C_0 \) is the pollutants concentration at position \((x, z)\) at \( t = 0 \). As a boundary value problem, the boundary condition is needed to control the system, with \( C = C_0 \) where the value of the solution at the boundary is known.

3.3 Meteorological Parameters
Meteorological parameters have an influence on air pollutants distribution. The wind can transport the pollutants far away from the source of emission and the diffusivity determines the rate at which pollutants diffuse. These parameters are dependent on the intensity of turbulence, which is influenced by atmospheric stability. In reality, to solve the Eq. (3) the variable wind velocity and the eddy diffusivity are considered to be functions of vertical distance, as suggested by [5]:

\[
u = U_r \left( \frac{z}{z_r} \right)^\alpha
\]

\[
K_z = K_r \left( \frac{z}{z_r} \right)^\beta
\]

where \( U_r \) and \( K_r \) are the measured wind speed \((m/s)\) and vertical diffusivity at a reference height \( z_r (m) \), \( \alpha \) and \( \beta \) are the constants depending on the atmospheric stability and surface roughness.

The mathematical forms of mesoscale wind in the horizontal and vertical directions as suggested by [6] are:

\[
U_x = -ax \left( \frac{z}{z_r} \right)^a
\]

\[
W_z = \frac{az^2}{\alpha + 1} \left( \frac{z}{z_r} \right)^\alpha + b \left( \frac{T - \min T}{\max T - \min T} \right)
\]

where \( a \) is a proportionality constant \((s^')\).

In the realistic form, temperature affects turbulence, which then affects the mesoscale wind. Therefore, the mathematical form of the mesoscale wind in the vertical direction becomes

\[
W = \frac{az^2}{\alpha + 1} \left( \frac{z}{z_r} \right)^\alpha + \frac{b}{\max T - \min T} \left( T - \min T \right)
\]

where \( b \) is the air flow constant which is caused by temperature.

3.4 Numerical Solution
The solution of Eq. (3) and Eq. (4) would be found numerically by explicit finite difference scheme. The explicit finite difference scheme implies that:

\[
\frac{\partial C}{\partial t} = \frac{C_{i,j}^{n+1} - C_{i,j}^n}{\Delta t}
\]
The central difference scheme is used for the advection and diffusion terms; the first derivative is as follows:

$$\frac{\partial C}{\partial x} = \frac{C_{i+1,j}^n - C_{i-1,j}^n}{2\Delta x}$$  \hspace{1cm} (6)

And the second derivative would be

$$\frac{\partial^2 C}{\partial x^2} = \frac{C_{i+1,j}^n - 2C_{i,j}^n + C_{i-1,j}^n}{\Delta x^2}$$  \hspace{1cm} (7)

The approximations (5), (6) and (7) are applied to Eq. (3) and (4). Substituting the approximations into Eq. (3) results in the equation below,

$$\frac{C_{i,j}^{n+1} - C_{i,j}^n}{\Delta t} + U \frac{C_{i+1,j}^n - C_{i,j}^n}{2\Delta x} + W \frac{C_{i,j+1}^n - C_{i,j}^n}{2\Delta z} = K \frac{C_{i+1,j}^n - 2C_{i,j}^n + C_{i-1,j}^n}{\Delta x^2} + K \frac{C_{i,j+1}^n - 2C_{i,j}^n + C_{i,j-1}^n}{\Delta z^2} - \lambda (c_{i,j}^{n+1})$$  \hspace{1cm} (8)

Rearranging Eq. (8) in the finite difference form, $C_{i,j}^{n+1}$ is obtained as:

$$C_{i,j}^{n+1} = A_i C_{i+1,j}^n + A_j C_{i,j+1}^n + A_{ij} C_{i,j}^n + A_{ij+1} C_{i,j+1}^n + A_{ij-1} C_{i,j-1}^n,$$  \hspace{1cm} (9)

for $i = 1, 2, 3, ..., M$ and $j = 1, 2, 3, ..., N$, where,

$$A_i = \frac{U \Delta t}{2\Delta x} + \frac{K \Delta t}{\Delta x^2},$$

$$A_j = \frac{W \Delta t}{2\Delta z} + \frac{K \Delta t}{\Delta z^2},$$

$$A_{ij} = \frac{2K \Delta t}{\Delta x^2} - 2 \frac{K \Delta t}{\Delta z^2} - \lambda \Delta t + 1.$$

The explicit scheme will converge and be stable when $0 \leq \frac{\Delta t}{\Delta x^2} \leq \frac{1}{4}$.

The second derivative is applied to the spatial dimensions and then approximations (5) and (7) would be substituted into Eq. (4), it would be:

$$\frac{x_{i,j}^n - x_{i,j}^{n-1}}{\Delta t} = \frac{Q_x}{\Delta x^2} \left( T_{i+1,j}^{n-1} - 2T_{i,j}^{n-1} + T_{i-1,j}^{n-1} \right) + \frac{Q_z}{\Delta z^2} \left( T_{i,j+1}^{n-1} - 2T_{i,j}^{n-1} + T_{i,j-1}^{n-1} \right)$$  \hspace{1cm} (10)

Rearranging Eq. (10) gives:

$$x_{i,j}^n = \frac{Q_x}{\Delta x^2} T_{i+1,j}^{n-1} + \frac{Q_z}{\Delta z^2} T_{i,j+1}^{n-1} + \frac{Q_x}{\Delta x^2} T_{i-1,j}^{n-1} + \frac{Q_z}{\Delta z^2} T_{i,j-1}^{n-1} + \left( -2 \frac{Q_x}{\Delta x^2} + 2 \frac{Q_z}{\Delta z^2} + 1 \right) T_{i,j}^{n-1}$$  \hspace{1cm} (11)

Eq. (9) and (10) can be computed for each $i = 1, 2, 3, ..., M$ and $j = 1, 2, 3, ..., N$ with $n = 1, 2, 3, ...$, which is true for interior grid points. At the boundary grid points, the boundary conditions are needed for discretization. Therefore, initial concentration value $C_{i,j}^0$ is an input data from user, for

$$i = \left( \frac{M}{2} - 1 \right), ..., \left( \frac{M}{2} + 1 \right) \quad \text{and} \quad j = \left( \frac{N}{2} - 1 \right), ..., \left( \frac{N}{2} + 1 \right) \quad \text{whereby} \quad M \quad \text{and} \quad N \quad \text{must be even number}.$$

Boundary conditions for the concentration of pollutants are as follows:

$$C_{i,0}^n = C_{i,N}^n = 0, \quad \text{for} \quad i = 0, 1, 2, ..., M \quad \text{and} \quad j = 0, 1, 2, ..., N,$$

$$C_{0,j}^n = C_{M,j}^n = 0, \quad \text{for} \quad i = 0, 1, 2, ..., M \quad \text{and} \quad j = 0, 1, 2, ..., N.$$


The following is the initial conditions of the temperature:

1) Lower temperature (LowT): \( T_{i,j}^n \) are given by input data from user for \( i = 0,1,2,...,M \) and \( j = 0,1,2,...,N \).

2) Upper temperature (UpperT): \( T_{i,j}^n \) are given by input data from user for \( i = 0,1,2,...,M \) and \( j = N-2,N-1,N \).

3) Temperature between ground level and atmosphere
   \[ \Delta \text{Temperature} = \frac{\text{LowT} - \text{UpperT}}{N-7} \]
   \[ T_{i,j}^n = (\text{LowT} - \Delta \text{Temperature}) \ast (j-3), \text{for } i = 0,1,2,...,M \text{ and } j = 4,...,N-3. \]

While for the boundary conditions are:

1) \( T_{i,j}^n = \text{LowT}, \text{for } i = 0,1,2,...,M \text{ and } j = 0,1,2,3, \)

2) \( T_{i,j}^n = \text{UpperT}, \text{for } i = 0,1,2,...,M \text{ and } j = N-2,N-1,N, \)

3) \( T_{i,j}^n = (\text{LowT} - \Delta \text{Temperature}) \ast (j-3), \text{for } i = 0 \text{ and } j = 4,...,N-3, \)

4) \( T_{i,j}^n = (\text{LowT} - \Delta \text{Temperature}) \ast (j-3), \text{for } i = M \text{ and } j = 4,...,N-3. \)

As stated earlier, the wind component and eddy diffusivity are functions of vertical distance. Therefore, the function of the large-scale wind becomes \( U = u + u_r \), hence

\[
U[i, j] = Ur \left( \frac{j}{z_r} \right)^a + q(i) \left( \frac{j}{z_r} \right)^a \tag{12}
\]

where \( q \) is proportionality constant and the function for the mesoscale wind, which is dependent on the change of temperature, becomes

\[
w[i, j] = \frac{a(j)}{\alpha + 1} \left( \frac{j}{z_r} \right)^a + b \left( \frac{T_{i,j}^n - \min T}{\max T - \min T} \right) \tag{13}
\]

For eddy diffusivity,

\[
K_z[j] = K_z \left( \frac{j}{z_r} \right) \tag{14}
\]

\[
K_x[j] = K_x \left( \frac{i}{z_r} \right) \tag{15}
\]

All these parameters are counted from \( i = 1,2,3,...,M \) and \( j = 1,2,3,...,N \), which is the same in the downwind \((x)\) direction with \( j \) becoming \( i \).

4. Result and Discussion

This study is to analyze the distribution of air pollution distribution under the influence of large-scale wind, mesoscale wind, and eddy diffusivity. Wind profile was defined in Eq. (12) as large-scale wind and Eq. (13) as mesoscale wind. While eddy diffusivity in vertical and horizontal direction were defined in Eq. (14) and Eq. (15) respectively. There are some unknown parameters, which are \( Ur,a,K_z,\alpha,\beta \) required for the computation of air pollution concentration. The range of values for some of the parameters are as follows: \( Ur = -50 \text{ to } 50 \text{ m/s}, K_z = 0 \text{ to } 5 \text{ m/s}^2, \alpha = -1 \text{ to } 1 \text{ s}^{-1}, b = -1 \text{ to } 1 \text{ s}^{-1}, z_r = 10 \text{ m} \).

The values of other unknown parameters \( \alpha \) and \( \beta \) are taken according to the stability conditions of the atmosphere and surface roughness as described in some previous studies which is shown in table 1 [7].
with these values of $\alpha$ (table 1) corresponding to different stability classes, the value of $\beta$ can be obtained as $\beta = 1 - \alpha$, based on Schmidt’s conjugate power law [8].

**Table 1. Variations of the values of $\alpha$ in different atmospheric condition [7].**

| Stability   | $\alpha$ |
|-------------|----------|
| Case I      | Unstable | 0.17    |
| Case II     | Neutral  | 0.27    |
| Case III    | Stable   | 0.61    |

It is assumed that pollutants are emitted at a constant rate in a uniformly distributed domain. The area extends up to 500 m downwind and 500 m in the vertical direction. The initial concentration and temperature are taken to be $C_0 = 1$ and $T_0 = 27^\circ$C and it is assumed that the range of heat diffusion $Q_x = Q_z$ from 0 to 5 m/s$^2$. The removal of pollutants is assumed to be taking place by either dry deposition or wet deposition processes. The value for the removal parameter $\lambda = 10^{-5}$ s$^{-1}$ is assumed to be constant in all atmospheric conditions.

A numerical method based on explicit scheme requires solutions to be within their range of validity. This analysis is done for $M = 100$ and $N = 100$ independent grids.

To be able to gain insight clearly on air pollution distribution under the effect of large-scale wind, mesoscale wind, and eddy diffusivity for unstable atmospheric condition, the computed concentration values have been analyzed and displayed graphically in figures 2, 3, and 4.

![Figure 1. Air pollution simulation interface.](image)

Figure 1. shows the user interface for the program developed using Lazarus programming software. The various buttons are described to show how the program works and the simulation carried out, are done by varying the atmospheric conditions and the parameter values. The various buttons and check boxes are the part where all the parameters are adjusted in order to have a clear understanding of how the pollutants distribute. Arbitrary points are chosen from the plane on the right by changing the X and Z label. When the program is initialized and run, pollutants are emitted from the stack and as the model parameters are adjusted, they then distributed in the plane and the corresponding graph is plotted which shows the decrease or increase in the level of concentration of pollutants by varying any of the parameters in the model.
The model parameters are varied and used for the unstable atmospheric condition. This is done to see the effect they have on the distribution of pollutants in the atmosphere. In the simulations, one particular parameter is varied whiles the others are kept constant. The simulations carried out in Figure 1 start with a large-scale wind of 7.5 m/s and this value is varied to 12.8 m/s and 22.4 m/s whiles the other parameters are kept constant. Also the level of constancy in the concentration of pollutants at 8, 7.5 and 6.5 unit of time for the small value, medium value and large value respectively, the concentration of pollutants maintains some level of constancy. This is due to the fact that, the emission is continuous and at the point the change in concentration of the pollutants and the atmosphere is the same so there is not any significance change. Also, the pollutants undergo an advection and diffusion process and therefore there would always be a drop or increase in the concentration levels in the presence of the model parameters. Table 2 shows the model parameters and their values.

| Table 2. Varying large-scale wind. |
|------------------------------------|
| Large-scale wind ($U_r$) | 7.5 | 12.8 | 22.4 |
| Heat Diffusion ($Q_r$) | 0.2 | 0.16 | 0.08 |
| Air Flow ($b$) | 0.2 | 0.16 | 0.08 |
| Mesoscale Wind ($a$) | 0.08 | 1.5 | 0.00159 |

From the figures, it can be seen that as the large-scale wind increases the level of concentration decreases. This is because the bulk transfer of pollutants is as a result of the intensity of the wind in a particular area. As the speed of wind increases, pollutants are carried farther away from the source of emission.
As studied by [3], the mesoscale wind prevents the dispersal of pollutants which results in the increase of pollution concentration in the atmosphere. From Figure 3, the level of concentration of pollutants increase as the mesoscale wind is increased from $0.08 \, m/s$ to $0.3 \, m/s$ but decreased when it was increased to $0.82 \, m/s$. This is because of the position of the arbitrary point that was chosen. The concentration of the pollutants was far from the arbitrary point and hence a decrease in the concentration level. The other model parameters are kept constant as the value of the mesoscale wind is varied. The level of constancy in the concentration of pollutants at 5, 3.5 and 3 for the small value, medium value and large value respectively is as a result of the same reason given under Figure 2. Table 3 shows the values used in each of the simulations carried out.

**Table 3. Varying mesoscale wind.**

| Parameter                  | Value 1 | Value 2 | Value 3 |
|----------------------------|---------|---------|---------|
| Large-scale wind ($U_r$)   | 7.5     | 0.2     | 0.16    |
| Heat Diffusion ($Q_r$)     | 0.3     | 0.82    |         |
| Air Flow ($b$)             | 0.08    | 1.5     |         |
| Delta t                    | 0.00159 |         |         |

**Figure 4. Varying diffusion rate.**

Diffusion is the movement of molecules from a higher concentration to a lower concentration. In this study, the higher concentration point is the source of emission of pollutants and the lower concentration is the arbitrary point that was chosen. As the diffusivity is increased from $0.6 \, m^2/s$ to $1.5 \, m^2/s$ and the finally to $3.7 \, m^2/s$, the level of concentration of pollutants increase as shown in Figure 35. In this case, the level of constancy in the concentration of pollutants at 9.5, 6.5 and 4.5 unit of time for the small value, medium value and large value respectively can be explained in the same way as that of Figure 2. This due to the fact that the pollutants are diffusing at a faster rate to the arbitrary point which tend to increase the concentration at that point. Table 4 shows the model values that were used to carry out the simulations.

**Table 4. Varying diffusivity.**

| Parameter                  | Value 1 | Value 2 | Value 3 |
|----------------------------|---------|---------|---------|
| Large-scale wind ($U_r$)   | 7.5     | 0.2     | 0.16    |
| Heat Diffusion ($Q_r$)     | 0.3     | 0.82    |         |
| Air Flow ($b$)             | 0.08    | 1.5     |         |
| Delta t                    | 0.00159 |         |         |
5. Conclusion
Air pollution is one of the major atmospheric problems the world faces nowadays. Mathematical models are used to estimate the impact of the concentration of air pollution, which have on the living organisms as well the entire ecosystem. By using the mathematical model, it becomes easier to learn the basic concept of the physical phenomena that take place during the distribution of pollutants as they undergo advection and diffusion processes under the influence of meteorological parameters. In this study, these parameters represent large-scale wind, mesoscale wind and eddy diffusivity. From the results, various simulations were carried out in order to access how the distribution of air pollution is affected when any of the meteorological parameters are varied.

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References
[1] WHO 2017 7 Million Premature Deaths Annually Linked to Air Pollution. http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/ 11 August 2017 (20:10)
[2] Arystanbekova N K 2004 Application of Gaussian plume models for air pollution simulation at instantaneous emissions Mathematics and Computers in Simulation 67 pp. 451-8
[3] Agarwal M and Tandon A 2009 Modeling of the urban heat island in the form of mesoscale wind and of its effect on air pollution dispersal Applied Mathematical Modeling 34 pp. 2520-30
[4] Lakshminarayananchari K, Sudheer P K L, Siddalinga P M and Pandurangappa C 2013 Advection – diffusion numerical model of air pollutants emitted from an urban area source with removal mechanisms by considering point source on the boundary International J. App. or Innov. in Eng. and Management 2 pp 251-268
[5] Lin J S and Hildemann L M 1996 Analytical solution of the atmospheric diffusion equation with multiple sources and height dependent wind speed and eddy diffusivities Atmos. Environ 30 pp. 239-254
[6] Dilley J F and Yen K T 1971 Effect of a mesoscale type wind on the pollutant distribution from a line source Atmos. Environ 6 pp. 843-851
[7] Erwin J S 1979 Technical note: A theoretic variation of the wind profile power law exponent as a function of surface roughness and stability Atmos. Environ 13 pp.191-4
[8] Sharan M and Kumar P 2009 An analytical model for crosswind integrated concentration released from a continuous source in a finite atmospheric boundary layer Atmos. Environ 43 pp. 2268-77