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VERTICAL DIFFUSION OF POLLUTANTS IN THE ATMOSPHERIC BOUNDARY LAYER

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Abstract

An atmospheric diffusion model is used to study the main features of vertical distri-bution of pollutants released from a continuous point source at different heights in a thermally stratified atmospheric boundary layer. The model is based on the two-dimen-sional advection-diffusion-deposition equation, with vertical profiles of wind and eddy diffusivity for the atmospheric boundary layer. The formulation permits studying the influence of surface roughness, atmospheric stability, mixing depth and source height on concentration distributions. Numerical simulations of atmospheric diffusion experiments are done. From the two-dimensional concentration fields, different para-meters are calculated in terms of the second, third and fourth order statistical moments which characterize vertical distributions of material. The computed values are satisfactorily compared with those evaluated from observational data. The vertical dispersion coefficient (second order moment root mean square) is also compared with those proposed by other authors.

Resumen

Se utiliza un modelo de difusión atmosférica para analizar las características princi-pales de la distribución vertical de contaminantes emitidos en forma continua, desde diferentes alturas, en una capa límite atmosférica térmicamente estratificada. El modelo se basa en la ecuación bidimensional de advección-difusión-depósito, con perfiles verticales del viento y la difusividad turbulenta válidos para la capa límite atmosfé-rica. La formulación permite analizar la influencia de la rugosidad del terreno, la estabilidad atmosférica y las alturas de la capa de mezcla y de la emisión de contaminan-tes sobre las distribuciones de la concentración. Se realiza la simulación numérica de experimentos de difusión atmosférica y a partir de los campos bidimensionales de con-centración obtenidos, se calculan diferentes parámetros en función de momentos estadís-ticos de segundo, tercero y cuarto orden, para caracterizar las distribuciones vertica-les del material. Los valores hallados se comparan satisfactoriamente con los resultan-tes de los datos observacionales. El coeficiente de dispersión vertical (raíz cuadrada del momento de segundo orden), se compara además con los propuestos por otros autores.

1. Introduction

One of the most serious problems for humanity is the continuous and increasing environment deterioration. Ambient pollution effects may be short- or long-term. The impact on different components of air-ground-water system may range from the local scale to the global one (in the case of possible climatic changes, due to radiative balance alterations), through the regional scale, as acid deposition.

Pollutants released from different sources are submitted in the atmosphere to atmospheric diffusion. Most of them undergo chemical reactions, reaching comparatively more noxious forms. Finally, they are removed from the atmosphere by dry and wet deposition processes.

Atmospheric diffusion models are used to predict air pollution concentrations. Those based on mass conservation equation may potentially incorporate meteorological parameter changes, surface feature effects, different emission conditions and pollutant remotion processes.

Pollutants diffusion is basically described by the dimensions and shape of the distributions in the air downwind of the source.

The operational diffusion model most widely used is the gaussian model. In its application, it is necessary to know, among other parameters, vertical dispersion coefficient values. Generally, this coefficient is estimated through empirical curves and varies with downwind distance and atmospheric stability. However, these empirical coefficients have a validity range restricted by the experimental conditions on which they are based. Application in different conditions may lead to erroneous values in computed concentration levels. Moreover, empirical coefficients are generally obtained through concentration measurements at ground level, assuming a gaussian vertical distribution.

Atmospheric dispersion coefficients, besides distance downwind and stability, depend on terrain features and emission conditions. In all cases, vertical dispersion must be less than the height of the mixing layer, upper boundary to vertical diffusion.

To obtain vertical dispersion coefficient values, Pasquill-Gifford curves (Gifford, 1961) are the most widely used. They are proposed for ground level emissions and rural zones. Combining experimental results and theoretical expressions, Briggs (1973) obtained dispersion curves valid for rural and urban zones separately. Hosker (1973), using previous Smith's (1972) nomograms, found expressions for the vertical dispersion coefficients as a function of surface roughness, for emissions from sources near ground level. There are few studies on emission height effects on vertical dispersion.

If pollutants' vertical distribution in the atmosphere has a gaussian shape, specifying plume mean height and vertical dispersion coefficient, the distribution is defined. However, there are experimental and theoretical results suggesting a departure from gaussian distribution (Pasquill and Smith, 1983).

In this paper, a diffusion model is applied to obtain the vertical distributions of substances released from a continuous point source, at different levels, in a thermally stratified atmospheric boundary layer. The model consists of the two-dimensional semiempirical advection-diffusion-deposition equation with vertical profiles of the wind and turbulent diffusivity which permit including stability conditions and roughness. Simulations of the Prairie Grass and the Hanford-67 dispersion experiments are carried out. The spatial distribution of concentrations is obtained from numerical methods, and characteristic parameters of the vertical distributions are calculated, as functions of different statistical moments, comparing the numerical predicted ones with those arising from observational data. Furthermore, the observed vertical dispersion coefficient (second order root mean square), used in operational models, is compared with those predicted by other authors.

2. The Advection-Diffusion-Deposition Model

The two-dimensional semiempirical advection-diffusion-deposition equation, for a continuous point source is (see Pasquill and Smith, 1983):

$$\bar{u}(z)\frac{\partial C}{\partial x}=\frac{\partial}{\partial z}\left[K(z)\frac{\partial C}{\partial z}\right]-v_d\frac{\partial C}{\partial z} \quad (1)$$

where x is alongwind, z is the vertical coordinate, C(x,z) is the crosswind-integrated concentration, $\bar{u}(z)$ is the mean horizontal wind speed, K(z) is the eddy diffusion coefficient, and v_d is the dry deposition velocity.

The vertical profiles of eddy diffusivity and wind speed are obtained from an extension of the Monin-Obukhov hypothesis to the whole atmospheric boundary layer, combined with the gradient-transfer theory. This extension relies on the assumption of local validity of the turbulent energy equation in a steady, horizontally homogeneous atmospheric boundary layer (Businger, 1982).

It is supposed that the friction velocity (u_*) depends on height in the atmospheric boundary layer, following the relation (Yokoyama et al., 1979):

$$u_* = u_{*0}\left[1-\frac{z}{h}\right] \tag{2}$$

where u_{*0} u_{*0} is the surface friction velocity and h the atmospheric boundary layer height.

It is assumed that the eddy diffusivity coefficient is equal to the momentum eddy diffusivity $K_m(z)$, whose vertical profiles are (Ulke, 1992):

- neutral and stable conditions ($h/L \geq 0$):

$$K_m(z) = ku_{*0}h\left[\frac{z}{h}\right]\left[1-\frac{z}{h}\right]\left[1+6.9\frac{h}{L}\frac{z}{h}\right]^{-1} \tag{3.a}$$

- unstable condition ($h/L<0$):

$$K_m(z) = ku_{*0}h\left[\frac{z}{h}\right]\left[1-\frac{z}{h}\right]\left[1-22\frac{h}{L}\frac{z}{h}\right]^{1/4} \tag{3.b}$$

where k is von Kármán's constant (k=0.41), L is the Monin-Obukhov length and h/L is the stability parameter used in the atmospheric boundary layer.

The K_m profile compares well with other theoretical and semiempirical forms (Yokoyama et al., 1979, Wieringa, 1980).

The wind profile is obtained from the K-theory using eqs. (2) and (3):

- neutral and stable conditions ($h/L \geq 0$):

$$\bar{u}(z) = \frac{u_{*0}}{k}\left\{\ln\frac{z}{z_0} - \left[1-6.9\frac{h}{L}\right]\left[\frac{z-z_0}{h}\right]\frac{6.9}{2}\frac{h}{L}\left[\frac{z^2}{h^2}-\frac{z_0^2}{h^2}\right]\right\} \tag{4.a}$$

- unstable condition ($h/L<0$):

$$\bar{u}(z)=\frac{u_{*0}}{k}\left\{\ln\frac{z}{z_0}+\ln\left[\frac{(1+\mu_0^2)(1+\mu_0)^2}{(1+\mu^2)(1+\mu)^2}\right]+2\left[\tan^{-1}\mu-\tan^{-1}\mu_0\right]+\frac{2L}{33h}\left[\mu^3-\mu_0^3\right]\right\} \quad (4.b)$$

with

$$\mu=\left[1-22\frac{h}{L}\frac{z}{h}\right]^{1/4} \qquad \mu_0=\left[1-22\frac{h}{L}\frac{z_0}{h}\right]^{1/4}$$

where z_0 is surface roughness length.

Expressions (3) and (4) tend, with small z/h, to the forms generally used in surface layer (see Panofsky and Dutton, 1984).

Eq. (1) with (3) and (4) is solved numerically (van Buijtenen et al., 1973). Spatial increments on both directions are variable in order to get a better description of diffusion near the source. Boundary conditions include dry deposition.

3. Vertical Distribution of Pollutants

The main features of the vertical distribution of pollutants can be specified using the statistical moments of that distribution.

The second order moment is defined as (see Pasquill and Smith, 1983):

$$\sigma_z^2(x)=\left[\int_0^\infty z^2C(x,z)dz\,/\int_0^\infty C(x,z)dz\right]-\left[\int_0^\infty zC(x,z)dz\,/\int_0^\infty c(x,z)dz\right]^2 \quad (5)$$

The vertical dispersion coefficient (σ_z), which represents the vertical extent of the plume, is obtained from the second order moment. In eq. (5), the root mean square of the second term is the mean vertical displacement of material.

Skewness (A_z) and kurtosis (K_z) coefficients, which characterize distribution shape, are defined respectively in relation with third and fourth order moments (see Wallington, 1968):

$$A_c(x)=\left\{\left[\int_0^\infty z^3C(x,z)dz\,/\int_0^\infty C(x,z)dz\right]-3\bar{z}\sigma_z^2-\bar{z}^3\right\}\sigma_z^{-3} \quad (6)$$

$$K_c(x)=\left\{\left[\int_0^\infty z^4 C(x,z)dz \Big/ \int_0^\infty C(x,z)dz\right]-4\bar{z}A_c\sigma_z^3-6\bar{z}^2\sigma_z^2-\bar{z}^4\right\}\sigma_z^{-4} \tag{7}$$

From the two-dimensional concentration field obtained with the model, it is possible to calculate σ_z, A_z and K_z at different distances of the source, numerically solving the integrals in (5), (6) and (7), with trapezoidal method.

4. Comparison With Expiremental Data

Numerical simulations are made of atmospheric diffusion field experiments carried out in the Projects Prairie Grass (Barad (ed), 1958) and Hanford-67 (Nickola, 1977).

In the Prairie Grass Project, SO_2 was released from a continuous point source at ground level. Concentration measurements were made at several vertical levels in towers located 100m from the source.

During Hanford-67 Project different particle tracers (Z_nS, F, Rh B) and a radioactive inert gas (Kr-85) were released. The emission heights were 1m, 2m, 26m, 56m and 111m. Concentration measurements were taken at different levels in towers located at distances 200m, 800m, 1600m and 3200m from the source.

Meteorological data for numerical simulations are: roughness length (z_0), surface friction velocity (u_{*0}), Monin-Obukhov length (L) and mixing height (h).

From the wind speed and temperature data at different levels we computed, in the following order: Richardson number (Ri), L (from Ri), (Businger et al., 1971), z_0 and u_{*0}. Mixing height, h, is found from the radiosonde and airplane temperature and humidity data in the case of Project Prairie Grass, and from theoretical expressions for Hanford-67 Project (see Ulke, 1992).

The characteristic coefficients of vertical distribution at each sampling distance are estimated from the two-dimensional concentration fields numerically obtained. The respective coefficient values for observational data are found from concentration vertical measurements. In both cases, they are calculated from the parameters definition, using expressions (5) to (7), with trapezoidal integration.

The observational vertical dispersion coefficients are also compared with the values obtained from Briggs' and Smith-Hosker's curves.

The departures of vertical dispersion, skewness and kurtosis coefficients obtained from numerical simulations from those obtained from observational data, are quantitatively studied. For this purpose, the global, difference and correlation measurements for air quality models validation are calculated (Willmott, 1982). This is also done for comparison of semiempirical vertical dispersion coefficients with observational data.

5. Results and Discussion

Figure 1 shows qualitatively the comparison between observed and model estimated vertical dispersion coefficients. A satisfactory global agreement is observed.

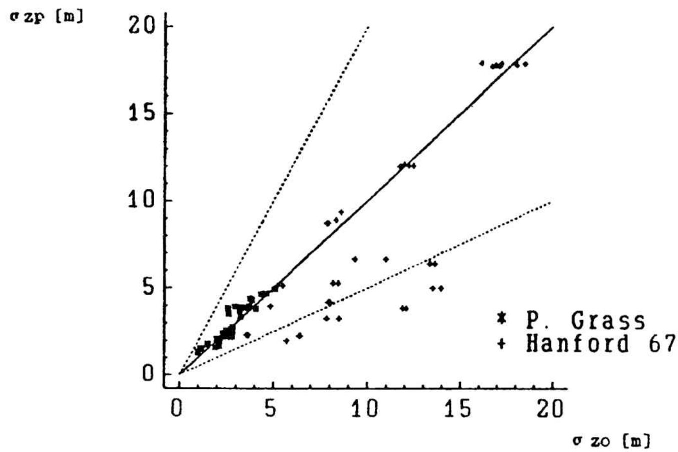

Fig. 1: Comparison of model predicting vertical dispersion coefficients (with those obtained from data)

The vertical dispersion coefficient is slightly underestimated, in the mean, with the proposed model. The observed coefficients show a slightly greater variability than the predicted ones. 15% of underestimated values is found in a factor less than or equal to 0.5.

In Table I it can be seen that root mean square error ($RMSE_u$) is due mainly to unsystematic errors (RMSE). The mean fractional error (MFE) shows mean underestimation factors of 0.85 with a 38% standard deviation (MFESD). There is an important observed-proposed values agreement (d=index of agreement). The linear regression parameters (a=intercept, b=slope, r^2 =coefficient of determination) show a good correlation.

	σ_z			A_z	K_z
	MODEL	BRIGGS	SMITH-HOSKER	MODEL	MODEL
MBE	1.07	-14	-10	-0.089	-0.164
S^2_d	6.30	1477	663	0.065	0.975
RMSE	2.72	40.71	27.52	0.269	0.995
$RMSE_u$	2.34	33.70	20.86	0.211	0.744
$RMSE_s$	1.38	22.84	17.95	0.166	0.660
MFE	0.16	-0.65	-0.45	0.095	-0.073
MFESD	0.38	0.49	0.45	2.92	0.20
d	0.917	0.215	0.337	0.926	0.855
a	0.101	-10.08	-9.88	0.291	1.582
b	0.821	4.69	4.05	0.743	0.592
r^2	0.745	0.314	0.471	0.787	0.608
N	80	80	80	80	80

MBE: Mean bias error S^2 d: Variance of the differences
RMSE: Root mean square error
RMSEu : Unsystematic root mean square error
RMSEs: Systematic root mean square error
MFE: Mean fractional error
MFESD: Mean fractional error standard deviation.
d: Index of agreement
a: Intercept b: Slope r_2: Coefficient of determination

Table 1: Statistical measurements calculated for the comparison of predicted vertical dispersion, skewness and kurtosis coefficients with those obtained from observational data.

Figs. 2 and 3 present the resulting comparison of semiempirical vertical dispersion coefficients estimated with the expressions proposed by Briggs (1973) and Hosker (1973) with experimental data. In both cases, a higher departure than that obtained in the previous comparison is noted. In general, a great overestimation of vertical dispersion coefficients is found with the two models, especially for the Briggs' curve system. Furthermore, a higher variability in the values obtained from semiempirical curves is observed.

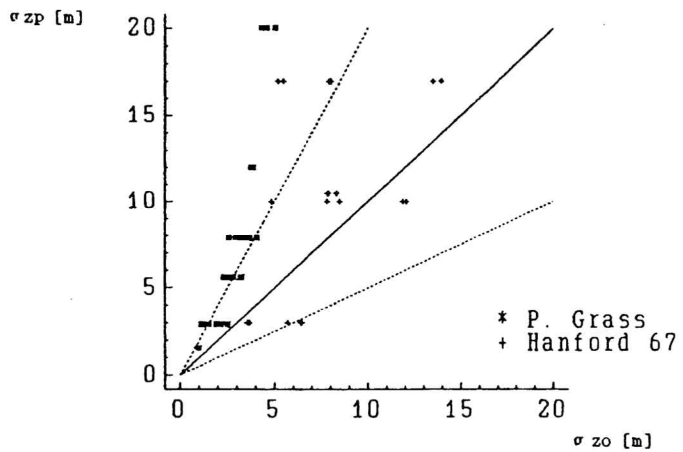

Fig. 2: Comparison of vertical dispersion coefficients predicted by Briggs formulas (Czp) with those obtained from data (Czc).

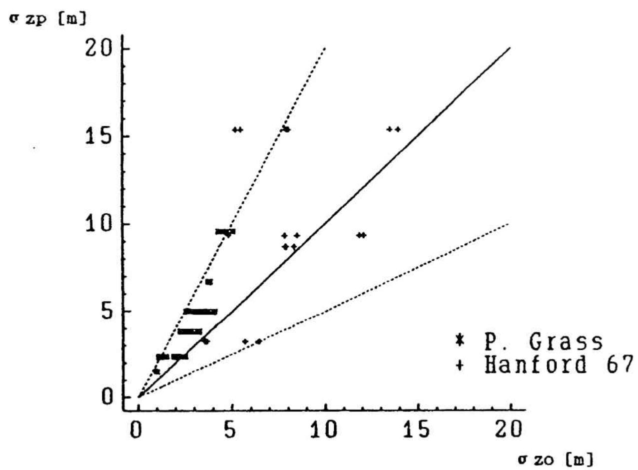

Fig.3: Comparison of vertical dispersion coefficients predicted by Hosker's formulas (Czp) with those obtained from data (Czc).

The global agreement between proposed and observed values is, in general, relatively small.

For Briggs' curves, 61% of overestimated values is found in a factor greater than or equal to 2, and for Smith-Hosker's curves the proportion is 29%. 2.5% of underestimated values is observed in a factor less than or equal to 0.5, for Briggs' curves.

In Table I it is observed that there is a great variance of the differences (S_d^2), between observed and theoretical values. The larger contribution to mean square error is due to unsystematic errors. The agreement between semiempirical values and observational data is low.

Mean fractional errors show overestimation factors from 1.6 to 2, with great standard deviations (nearly 50%).

The linear regression coefficients point out, in coincidence with the other parameters, a departure from the best fitting.

Figure 4 shows the comparison of skewness coefficients estimated from numerical simulations with those obtained from observational data. It is of interest to point out that most skewness coefficients, both observed and proposed, are greater than zero.

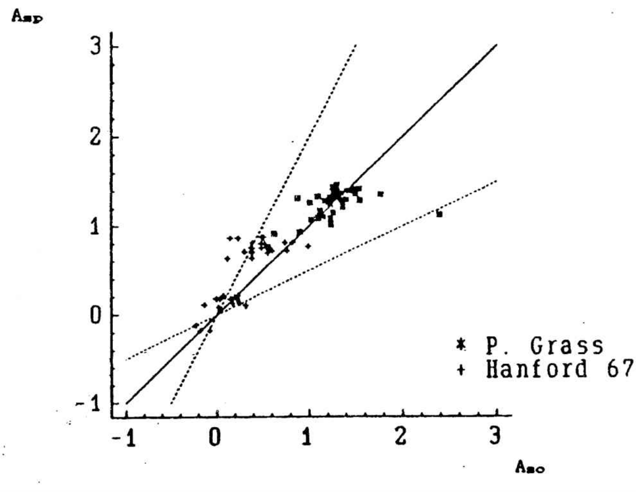

Fig.4: Comparison of model predicted skewnwss coefficients (Azp) with those obtained from data (Azc)

In general a slight overestimation of skewness coefficients from the proposed model is noted. The variability of the observed values is greater than that of the predicted ones. There is 16% of overestimated values in a factor greater than or equal to 2, and 5% underestimated in a factor less than or equal to 0.5.

In Table I it can be seen that the mean square error is due in a larger proportion to unsystematic errors.

The mean fractional error, in opposition to the other parameters. shows a very slight underestimation of skewness coefficients, in a factor 0.91, with an important standard deviation.

The index of agreement shows a satisfactory relation between proposed and experimental coefficients. A similar conclusion is obtained from the linear regression parameter values.

Figure 5 presents the comparison results for kurtosis coefficients.

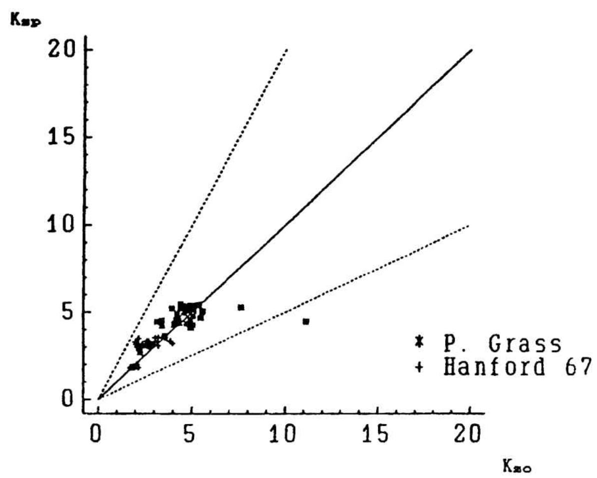

Fig.5: Comarison of model predicted kurtosis coefficients (Kzp) with those obtained from data (Kzo)

For the Prairie Grass data most of the kurtosis coefficients are greater than 3. In the case of Hanford-67 Project, the major proportion is different from 3.

A very small mean difference is found between predicted and observed kurtosis coefficients, and, on the other hand, the latter show greater variability.

The global agreement between proposed and observed values is acceptable.

There is only one observed kurtosis coefficient underestimated by the model in a factor less than or equal to 0.5.

In Table I it can be seen that the mean fractional error shows a slight overestimation of observed kurtosis coefficients, with an associated factor of 1.07 and 20% standard deviation. Unsystematic errors compound, in a larger proportion, the mean square error. The index of agreement shows a satisfactory predicted-observed relation.

Linear regression parameters suggest a departure from the best fitting.

5. Conclusions

In general, vertical concentration distributions show a departure from a gaussian shape. This is related with vertical variations of wind speed and eddy diffusivity and with the influences of surface and top mixing height.

Vertical dispersion coefficients proposed by other methodologies and those obtained from observational data show less agreement than that found in the comparison of the model predicted coefficients with data.

The vertical dispersion, skewness and kurtosis coefficients computed by the model are in reasonable agreement with the experimental data.

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