

COMMISSION OF THE EUROPEAN COMMUNITIES

# **environment and quality of life**

## **Dispersion simulation models for air pollutants**



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**Published by the**

**COMMISSION OF THE EUROPEAN COMMUNITIES**

**Directorate-General**

**'Scientific and Technical Information  
and Information Management'**

**Bâtiment Jean Monnet - Kirchberg**

**LUXEMBOURG**

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*Printed in Belgium*

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Abstract

In the vicinity of chimneys the micro-meteorology is governed by the emission process itself. There, the eddies carrying the discharged pollutants start their way through the laminar/turbulent moving atmosphere. In the course of time, the eddies decay and coagulate. The transport mechanism for a diluted eddy gas is governed by the Boltzmann equation which can be solved by Monte Carlo techniques. The concentrations of the different chemicals relevant to air pollution change in time according to the local concentration ratios which, in their turn, change in space and time due to eddy diffusion. The adequate mathematical tool is the "first collision concept".



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## 1. Dispersion Mechanism

From turbulence research one knows that an air stream usually contains temporarily stable air parcels, eddies, which lead an independent life during finite time periods and afterwards decay. These eddies vary in size. Obviously as well as the drift component the eddy motion also has a stochastic component. One observes smooth pieces of path and abrupt changes in direction. In macroscopic stationary flows, there are regions with a practically unvarying spectrum of eddy sizes. This, however, does not mean that the eddies have an infinite life- and residence-time, but that there are a lot of individual histories (birth, stability phase, decay) phase-shifted against another.

In such a continuum interspersed by eddies a gas-cloud enriched with pollutants will be released. At this time point, it can be considered as a single parcel. One observes that it undergoes the same fate as the other eddies: it has a certain stability phase, it decays afterwards into smaller "quasi-stable" units which, in their turn, decay again after a certain life-time and which, finally, lose their macroscopic particle structure (RICHARDSON (1922)). Thus, the entrained pollutant will be distributed more or less homogeneously over a certain volume containing, as a rule, yet other eddies of different "ages". If the individual constituents enter a region where eddy-birth takes place, they will be partially joined with constituents of other decaying eddies to form new bigger quasi-stable units (coagulation), and the transport- and decay process continues. In this way, the pollutant is dispersed much faster and more extensively than by molecular diffusion. The eddies thus act as vehicles and the atmosphere as the carrier.

The cascade of pollutant eddies emerging from a decaying waste-gas cloud is, outside the "effective source domain" which has still to be defined, numerically small, in comparison with the number of turbulence-elements there. An interaction between pollutant eddies will therefore be rarer by far than the interaction between pollutant eddies and carrier. Consequently, the ensemble of pollutant eddies, outside the effective source domain, will hardly modify the turbulence field of the atmosphere. This

fact is of fundamental significance for the mathematical description of the pollutant dispersion process ( $\rightarrow$ linearization). After a certain relaxation time, both the polluted eddies and the turbulence elements will have the same size-spectrum, the same velocity-distribution and the same decay-behaviour. There is equilibrium.

Inside the effective source domain, however, a process of accommodation takes place: the elements of the polluted cloud will be "thermalized". One will, therefore, distinguish between a phase of thermalization and a phase of equilibrium.

The observable abrupt changes in direction could be assumed to be "collisions" between eddies. The corresponding interaction mechanism, however, is up to now unknown. Only an intensive study of the eddy structure can give some insight. This however concerns turbulence research and is for us of peripheral interest only. We are not at all interested in the fate of the single eddy, since we need only the space-time dependent density distribution of the atmospheric pollution.

There exist, in the literature, many attempts to use a mechanism, taken from another domain of physics, to describe the dispersion process, such as e.g. that used in the theory of Brownian motion (CHADRASEKHAR (1943)). One should try, however, to combine such a mechanism with the concept of "mean life-time".

The statistical distributions of life-times determining the cascade-decay have to be taken from experimental data; they are probability- or frequency-distributions, like all experimental results in this field, since the observations register only realizations of statistical quantities or sample means. Life-times of polluted eddies depend on

density  
size-spectrum  
velocity-distribution } of the turbulence elements

and on the

size  
velocity } of the polluted eddy.

It seems reasonable to interpret each collision as an absorption followed by an emission. If, at the collision point, the eddy is absorbed, it automatically enriches the carrier with pollutant. If there is, an instant later, an emission, the new eddy may have a gain or loss of pollutant content in comparison with the incoming eddy. We then speak of a "up- or down-scattering".

There are two different models to deal with the decay and coagulation: either the eddy is assumed to be a cluster of uniform "atoms", or an air parcel, homogeneous inside but of irregular shape, to which arbitrary masses can be added or taken away. Another assumption fundamental for the modelling is, that the interactions do not possess any memory effects. Consequently, the velocity direction of an emitted eddy is equally distributed, if the eddy distribution of the carrier is on average isotropic. Without turbulence elements there are, obviously, no collisions.

Between the collisions the eddies follow curved paths, since the buoyancy- and gravity-forces act on them continuously. In view of the need for a mathematical simulation one may ask whether such a trajectory could be approximated sufficiently well by a chain of straight paths, joined together at the collision points. This seems to be permissible, if the corresponding force-field does not noticeably change along the "mean collision-free path". In this case, one can apply the concept of "free paths" fruitful elsewhere; i.e. velocity changes are assumed to occur at discrete space points only, in the form of collisions.

Finally, one should point out that single eddies are always representatives of a turbulence spectrum and that eddies of different sizes have different mobility. Quantitative relations must be taken from turbulence theory.

During the phase of equilibrium the mathematical effort of simulation is considerably smaller than during that of the thermalization. In the case of equilibrium one could assume the kinetic energy of the turbulence elements to be e.g. Maxwellian-distributed. The parameters characterizing such a distribution have to be fixed by fitting experimental data.

The question whether the eddies, emerging a puff later, are already preformed during the puff emission (cluster model) and are travelling together for a while or whether the mode of decay will be decided only at the moment where the eddy hits an obstacle, seems to be irrelevant, but largely determines the mathematical structure of the simulation model.

In the case of a cluster structure of the puffs representing the emission of a point source, the number of the uniform cluster components is proportional to the intensity of the source; the rate of release can vary with time.

At the space-time point where the eddy emission takes place both the eddy and the carrier have an identical concentration spectrum of chemical substances. During the free flight, the eddy represents a closed system without any contact with the carrier material permeated. Because of this, the chemical reactions occurring inside the eddy are independent of the situation outside. At the point of absorption, the local concentration spectrum of the absorbing material becomes modified by the chemicals carried by the eddy. Other more or less simultaneously arriving eddies contribute in a similar way. Obviously each eddy born and starting there carries with itself, the actual concentration spectrum of the birth place, and so forth.

In the vicinity of chimneys the micro-meteorology is governed by the emission process itself. Released hot gases create a buoyancy zone into which surrounding cold air is drawn. That mixing region is the "effective source region" of an emitter. There, the eddies carrying the discharged pollutants start their way through the laminar or turbulent moving atmosphere. In the course of time, the eddies decay and coagulate. Precipitations and chemical reactions change the pollutant concentration in the air, absorption and reflection along solid and fluid surfaces. Because of eddy diffusion and convection caused by the local aerodynamics the polluted air will be distributed through an increasing volume.

## 2. Simulation Techniques

A stochastic simulation model is required which makes use of a submodel for each of the relevant phenomena (local turbulence, buoyancy, wash-out etc.) governing the transport-, dispersion- and dilution processes, which can be replaced by a more sophisticated one, if measurements require it.

Since a regional air monitoring network has only a finite number of stations at its disposal, one will interpret the atmosphere of a region as a 3-dimensional system of compartments, each with its own homogeneous micro-meteorology. All meteorological information belonging to the corresponding compartments is called the meteorological inventory of the region. Defective inventories have to be completed either by interpolation or by additional measurements. A dispersion simulation is only applicable to a region for which complete inventories of meteorology and emission are available.

Only a model simulation of the pollutant dispersion can check the relevance of the single parameters characterizing the different submodels and distinguish between parameters of the local and universal significance. Evidently, both the construction and the validation of such a simulation model need a rather large number of experiments both on the laboratory and the city scale. These experiments serve two different purposes: to determine the dispersion characteristics of a city and to test the model structure. Finally, sensitivity analysis must be used to find the range of values for the different model parameters and to minimize the model input. The decisive minimization, however, can be done only using measured data obtained from dispersion experiments both on the laboratory and on the city scale.

Last but not least, it should be guaranteed that the model has a sound theoretical basis, that a particularly lucid simulation results, and that this approach can conveniently be performed on a computer with economical use of computer time, if one follows the pollutant dispersion from emission to the end of a given observation interval.

Given a set of air quality standards, the complete emission inventory of the region to be controlled and the corresponding meteorological inven-

tory containing a sufficiently detailed description of the local meteorology. Determine a computerized pollutant level forecast indicating the pollutant concentration distribution in adequate detail in space and time, paying regard, in the first instance, to the dangerous situations usually accompanied by irregular or weak winds.

An inspection of smoke diffusing near the ground gives the impression that the dispersion is brought about primarily by the relative motion with the wind but otherwise seems to move at random. An examination of the record of wind speed shows that the oscillations are caused by the passage of whirls and vortices, which, like molecules, appear to have distinct identities. The idea that molecular formalism can describe in an empirical fashion the transport of pollutants within a turbulent atmosphere is taken for granted by many researchers even though there is no proof that this is generally justifiable. Whether this approximation is appropriate can be judged only on the basis of the particular application.

Fundamentally, the pollutant dispersion can be described from both the Eulerian and the Lagrangian points of view.

#### a. Eulerian Models

The transport mechanism for a diluted eddy gas is governed by the linear Boltzmann equation<sup>1)</sup>:

$$(1) \quad \frac{df}{dt} = -\alpha f + \mathcal{S}_{m', \vec{v}'} \beta(m', \vec{v}'; m, \vec{v}) f(m', \vec{v}') + \mathcal{J}$$

This is a balance equation. The left-hand side is the substantial derivative of  $f$ , which is the number of eddies of type  $m$  per phase-space volume element, whereas the right-hand side represents the sum of gains and losses there. Using the "free path concept", i.e. polygonal trajectories consisting of straight paths interrupted by instantaneous deviations, we get

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1)  $\mathcal{S}$  denotes a superposition, a discrete summation  $\sum$  or a continuous integration  $\int$  or both.

$$(2) \quad \frac{df}{dt} = \frac{\partial f}{\partial t} + \vec{v} \cdot \text{grad } f$$

$\vec{v}$  denotes the velocity vector of the individual,  $\alpha$  the "total interaction coefficient" and  $L = 1/\alpha$  the "mean life-time",  $\beta$  the "scattering coefficient". To solve the transport equation (1), a Monte Carlo approach has been developed, in the past, which constructs a series of random walks for a stochastically selected but representative sample of members of an expanding particle cloud. For a more detailed explanation see, for instance DAVISON (1957).

A simultaneous treatment of chemical reactions, however, requires a more complex kernel  $\beta$ , that is a kernel which depends on the density  $f$  itself. The classical Monte Carlo techniques here meet difficulties. One, therefore, tries to change eq. (1) into a more convenient form. To do so, one takes the so-called "first collision concept" successfully applied to reactor physics (DAVISON (1957)).

All collision can be treated as first collision by assuming that an eddy suffering any collision is absorbed. In this picture the collision density

$$(3) \quad \psi(\vec{r}, t) = v\alpha \sum_{m, \vec{v}} f(m, \vec{v}) = v\alpha \chi(\vec{r}, t)$$

satisfies the equation

$$(4) \quad \left\{ \begin{array}{l} \psi(\vec{r}, L) = \sum_{\vec{\rho}} K(|\vec{r} - \vec{\rho}|) [S(\vec{\rho}, \tau) + h(\vec{\rho}, \tau) \psi(\vec{\rho}, \tau)] \\ \tau = t - |\vec{r} - \vec{\rho}|/v \end{array} \right.$$

$K(|\vec{r} - \vec{\rho}|)$  is the probability for an eddy starting from  $\vec{\rho}$  to suffer its first collision at  $\vec{r}$ , e.g.

$$K(|\vec{r} - \vec{\rho}|) \sim \frac{v\alpha}{4\pi|\vec{r} - \vec{\rho}|^2} \exp(-v\alpha|\vec{r} - \vec{\rho}|)$$

$h(\vec{r},t)$  new eddies appear at the absorption point, provided that the emission is isotropic.

Nonlinearities characteristic for chemical reactions enter here via the "multiplication factor",  $h(\vec{r},t)$ , which is now dependent on the densities of all the substances involved in the chemical transformation and which has to be derived from the kinetic equations taken from the literature (e.g. LEIGHTON (1961)). Eq. (2) can be solved by an iteration starting from plausible density distributions. Eqs. (1) and (4) represent the basis of our considerations.

We shall now discuss the dispersion of inert substances and come back to the chemical transformations at the end of this paper.

The space-time dependent number of eddies  $\chi(\vec{r},t)$  satisfies the differential equation

$$(5) \quad \text{div} (\mathbb{D} \text{grad}) \chi = a \chi + \frac{\partial \chi}{\partial t} + T \frac{\partial^2 \chi}{\partial t^2} + \sum_{k=3}^{\infty} \epsilon_k \frac{\partial^k \chi}{\partial t^k} + Q$$

where the  $\epsilon_k$ 's, the absorption coefficient  $a$ , the relaxation time  $T$  and the eddy diffusion tensor  $\mathbb{D}$  depend on  $\alpha, \beta, \vec{v}$  and space-time  $(\vec{r},t)$ . (SOODAK (1962); MÜLLER (1975)).

To keep the mathematics within reasonable bounds, we shall, in a first step, confine ourselves to discrete eddy-classes and to the so-called "hyperbolic approximation" characterized by

$$(6) \quad \epsilon_k \equiv 0 \quad \text{for} \quad k \geq 3$$

Only when it turns out that this approximation is not sufficient for the interpretation of an experiment, shall we take into consideration higher time derivatives also.

The hyperbolic approximation leads to classical equations of mathematical physics. In the past, numerous integration approaches have been developed which we can use. (See e.g. FRANK - v. MISES (1961)).

Equation (1) degenerates, for n different types of eddies, to the set of wave equations

$$(7) \quad T_i \frac{\partial^2 \chi_i}{\partial t^2} + \frac{\partial \chi_i}{\partial t} + \sum_{j=1}^n b_{ij} \chi_j = \nabla \cdot (D_i \nabla \chi_i) + Q_i$$

with  $i = 1, 2, \dots, n$ .

Since the solution  $\chi_i^{as}$  of the parabolic equations, i.e. the diffusion equations

$$(8) \quad \frac{\partial \chi_i^{as}}{\partial t} + \sum_{j=1}^n b_{ij} \chi_j = \nabla \cdot (D_i \nabla \chi_i) + Q_i$$

converges, for increasing t, to  $\chi_i$ , i.e.

$$(9) \quad \chi_i^{as} \approx \chi_i \quad \text{for} \quad t > T$$

one shall also test the solutions of (8) by experiments, to find their practical usefulness.

Several attempts have been made to justify the application of both the wave equations (7) and the diffusion equations (8) to pollutant dispersion (e.g. MONIN (1959), LAMB-SEINFELD (1973), PASQUILL (1974)). The a priori free parameters of the derived equations can be fixed either by fitting their solutions to experimentally measured concentrations  $\chi_i$  or by theoretical reasoning.

For solving codes for the equations (7) and (8) see e.g. OECD-NEA (1976).

It should be said here that the solutions of all the dispersion equations mentioned above can - via syllogisms from other domains of physics - be interpreted as a sum of stochastic micro-processes and mathematically be solved by Monte Carlo techniques (e.g. MÜLLER (1975)).

b. Lagrangian Models

One has here two different possibilities of interpreting the pollutant dispersion, that is, by allowing the eddies

- to be "interwoven" in the turbulence field, to move in harmony with it, or
- to lead an autonomous existence there (within the meaning of the Brownian motion).

The choice will be influenced mainly by the measured data available, such as frequency distributions of wind-speed and direction, of eddy energies and sizes, and two- or more-point correlations and turbulence scales of type (e.g. ROTTA (1972))

$$(10) \quad \left\{ \begin{array}{l} L_{ijk}(\vec{r}, t) = \gamma_{ij} \int_{-\infty}^{+\infty} R_{ij}(\vec{r}, t; \vec{p}_k, 0) d\vec{p}_k \\ T_{ij}(\vec{r}, t) = \gamma_{ij} \int_{-\infty}^{+\infty} R_{ij}(\vec{r}, t; 0, \tau) d\tau \\ \gamma_{ij} = [2 \overline{u'_i(\vec{r}, t) u'_j(\vec{r}, t)}]^{-1} \\ R_{ij}(\vec{r}, t; \vec{p}, \tau) = \frac{\overline{u'_i(\vec{r}, t) u'_j(\vec{r} + \vec{p}, t + \tau)}}{[\overline{u'^2(\vec{r}, t)} \overline{u'^2(\vec{r} + \vec{p}, t + \tau)}]^{1/2}} \end{array} \right.$$

Continuum Theory

The Eulerian velocity field  $\vec{u}$  composed into  $\vec{u} = \vec{u}_m + \vec{u}'$  possesses, apart from the mean velocity  $\vec{u}_m$ , a so-called "stochastic component"

$$(11) \quad \vec{u}'(\vec{r}, t) = \sum_{\vec{k}, \omega} \vec{U}(\vec{k}, \omega) \exp(i\omega t + i\vec{k} \cdot \vec{r})$$

which is a superposition of all the air motions not describable up to now.

in a deterministic sense. Usually the amplitude  $\vec{U}$ , the wave number  $\vec{k}$  and the frequency  $\omega$  will be taken from long-time observations. As one knows, the moving atmosphere carries air masses behaving like lumps of a certain consistency and moving as units during finite time intervals, i.e. clusters of submasses having nearly identical histories of motion and highly correlated velocity vectors. Two- and more-point correlations allow the identification of such "air parcels" experimentally. Within the framework of a mathematical modelling of pollutant dispersion the possibility of calculating one or more of the quantities  $\vec{U}, \vec{k}$  and  $\omega$  by the aid of random generators and to simulate, finally, the eddy trajectories as random walks suggests itself.

KRAICHNAN (1970) developed a model for eddy diffusion in which the velocity field was represented by a Fourier series

$$(12) \quad \vec{u}(\vec{r}, t) = \sum_{-\infty}^{+\infty} \vec{V}_n \times \vec{k}_n \exp(i\omega_n t + i\vec{k}_n \cdot \vec{r})$$

where the  $\vec{V}_n$  and  $\omega_n$  were taken from Gaussian distributions and the  $\vec{k}_n$  from classical energy spectra. ORZAG (1969), McCOY (1975) and others used similar approaches. PATTERSON/CORRSIN (1966) established a random field of velocities on a predetermined space-time lattice. As a particle moves, it obeys the velocity instructions that it finds as it arrives at lattice points. TOMPSON (1971) created particle trajectories directly in  $\vec{r}$ -space by generating  $\vec{u}$  according to measured or theoretically calculated statistical means like  $\sigma_i^2 = \langle u_i'^2 \rangle$ .

The diversity of measured frequency distributions and disposable turbulence scales, like (10), offers a multiplicity of similar structured models for eddy diffusion and thus automatically for pollutant dispersion. (e.g. JOYNT-BLACKMAN (1976)).

RICHARDSON (1922) developed the hypothesis that fully developed turbulence consists of a continuous hierarchy of eddies of various orders of size and energy content. The eddies have a characteristic life-time, after which larger eddies give up their energy to smaller ones. The

lowest order eddies dissipate their energy heating up the atmosphere. This decay, sometimes locally accompanied by eddy coagulations, makes a realistic modelling rather complicated. The literature contains few appropriate references.

### Particle Theory

One interprets the turbulence as a cloud of particles interspersed in a continuous air flow and leading an individual existence. Thus, one concedes to the atmospheric motion a degree of freedom higher than usual, with the aim of equipping the model with an ample set of free parameters and granting a greater flexibility which, in its turn, permits us to take into account dispersion relevant information inherent in the flood of meteorological data continuously registered over many years and characterizing the local situation. The simplest type of such a description is, to concede an individual existence only to the polluted eddies and to interpret the "clean" turbulence field as a continuum in the above mentioned sense. The situation resembles a set of pearls (eddies) dancing about a deforming "flying carpet" (turbulence field). It is obviously a sort of Brownian motion.

Wind- and temperature-field fluctuations expressed by frequency distributions, correlation coefficients etc. supply both statistical measures like  $\vec{u}_m$ ;  $\sigma_u^2 = \langle \vec{u}'^2 \rangle$  and  $\theta_m$ ;  $\sigma_\theta^2 = \langle \theta'^2 \rangle$  characterizing, in their turn, statistical distributions like the Gaussian and, in addition, particle properties like sizes, free paths and free flight times, in short, basic information to construct particle trajectories in the form of random paths. The variety of these gives us the opportunity to calculate a "plume axis", a "plume rise" and "dispersion parameters ( $\sigma_1$ ,  $\sigma_2$ ,  $\sigma_3$ )" as a set of ensemble means. These parameters determined by small-scale experiments can, sometimes, be used as input to city-scale pollutant dispersion models like these of TURNER (1964), FORTAK (1970) etc.

A more sophisticated simulation assumes that the "clean" turbulence field is composed, at least partially, of an eddy cloud also, migrating in an ocean of air, such as ring- and spherical-shaped vortices or

clusters of such units (MILNE-THOMSON (1960)). The gas of polluted eddies permeates the turbulence gas. Whether the eddies under consideration are really identical with these classical vortices or not has still to be clarified, but this is a task for turbulence research. Pollutant dispersion modellers will, with thanks, accept such results and apply them in their business.

Now some remarks concerning the mathematization of the dispersion processes.

One usually starts here from the Newton equation

$$(13) \quad \frac{d\vec{v}}{dt} = -\mathbb{A}\vec{v} + \vec{a}$$

where  $\mathbb{A}$  is the friction tensor and  $\vec{a}$  any acceleration acting on the particle as a whole.  $\vec{v}$  and  $\vec{a}$  are mean values representative for the eddy motion. A formal integration leads to

$$(14) \quad \vec{v}(t) = e^{-\mathbb{A}t} \vec{v}(t_0) + \int_{t_0}^t e^{-\mathbb{A}(t-\tau)} \vec{a}(\tau) d\tau$$

This result shows that the velocity  $\vec{v}$  consists of two additive components, one dependent on the initial velocity  $\vec{v}(t_0)$  and decreasing in time. The remaining component is the end-result of all the influences affecting the particle during the time-interval  $(t - t_0)$ .

The classical theory of Brownian motion interprets  $\vec{a}$  as a series of randomly spaced Dirac-impulses caused by collisions between the Brownian particle and the molecules of the carrier. As a result of the particle bombardments, the velocity of the diffusing Brownian particle at any instant is random in magnitude and direction, its velocity-history being thus a stochastic process. The time-integral of the velocity-history is the displacement-history which is necessarily again a stochastic process. The corresponding random-walk approach divides the total displacement of a Brownian particle into a number of independent steps

$$(15) \quad \Delta \vec{x} = \int_{\Delta t} \vec{v}(\tau) d\tau$$

The probability distribution of a particle executing such random steps is a standard problem of probability theory with an extensive literature (e.g. CRAMER (1946), BARTLETT (1956), FELLER (1957)).

According to (15) the random step can be taken in the form

$$(16) \quad \Delta x_i = p_i(t) \Delta t + q_i(t) r_i(t) \Delta t$$

where  $r_i(t) \Delta t$  is any random process with independent increments, e.g. a Gaussian white noise, and where  $p_i$  and  $q_i$  are Eulerian quantities which can be obtained either from measurements of the flow conditions or from theoretical or empirical correlations.

BULLIN-DUKLER (1974) simulated this method on a hybrid computer. Time-varying fluctuating velocities were simulated using white noise generators filtered in a specific manner. The procedure can readily accommodate time-varying boundary conditions and source strengths as well as time and position dependent flow conditions. The computer time required is very short.

CHANDRASEKHAR (1943), GOLDSTEIN (1951), MONIN (1959), KIRMSE (1964), RILEY-CORRSIN (1971) and the already cited authors relate the coefficients of equation (16) to the physics of the problem and adopt the "free path concept" already mentioned above. They follow an ensemble of uniform and unchanging particles over a series of equal time intervals, but they do not consider the life-time, the decay and the coagulation conditioned by the local structure of the atmosphere. The simultaneous existence of eddies of different sizes and pollutant content are also neglected. One wonders whether this deficiency can be compensated for by a more refined consideration of meteorological quantities only.

Most of these authors show that their "Lagrangian model" is equivalent to a Eulerian "diffusion equation" or a "telegrapher's equation", at

least asymptotically. The cascade decay, however, already demonstrates that instead of a single dispersion differential equation (parabolic or hyperbolic) one has to apply a set of those equations and that the pollutant concentration calculated by the superposition of the different particle densities does not necessarily have to satisfy such a second-order differential equation.

### 3. Eddy Diffusion

Flying balloons or tetroons move along polygonial trajectories composed of free paths some meters in length. Quasi-stable parcels of air show similar random walks. Profiting from this similarity we construct an analogous Monte Carlo approach corresponding to the linear Boltzmann equation (1) and thus solve it.

A local turbulence field induced by an obstacle or by thermal effects in a laminar meso-wind field will be interpreted as an eddy cloud. The eddy velocity is composed of a drift-component and a stochastic one; outside the cloud the eddy density is much lower. A polluted air-parcel entering the turbulence field will be thermalized there.

Polluted eddies are assumed to be particles of a diluted gas, released from a known position in space and time. Let  $x_i(t)$  be the coordinate of this moving particle. Such coordinates are random variables. Each such release of a particle can be expected to result in a different time function  $x_i(t)$ . Its location is recorded at  $t = T$ . This is repeated many times for each  $T$  and for a series of values of  $T$  of interest. The local concentration  $\chi$  can be found from a sample mean by dividing the number of particles recorded as being in some small volume by that volume.  $x_i(t)$  is assumed to be a Markovian process. Thus, the statistics of the particle locations at time  $t$  in the future depends only on its position at the present and is independent of the past. This implies in our case, that the location of the eddy is independent of the history of the turbulence.

a. Life-time

These particles interact with the gas of the turbulence elements characterized by a given particle density  $\delta$  and a given velocity distribution  $g(V)dV$ . The interaction is a sort of scattering or moderation. Let the particles of the moderating gas have a velocity-independent cross-section  $\sigma$  and the trajectories of the colliding particles, before the interaction, an angle  $\xi$  ( $\cos \xi = \mu$ ). Since all directions are equally likely for both the particles, the probability of a collision angle between  $\xi$  and  $(\xi + d\xi)$  is  $d\mu/2$ . Thus the number of such collisions per space-time volume element is given by

$$(17) \quad d\chi = v_r \sigma \delta g(V) dV d\mu/2 = v \sigma ds = v d\alpha$$

where  $v_r$  = relative velocity between the colliding particles and  $ds$  = differential cross section for a particle of velocity  $v$ .

In the centre-of-mass system the velocity only changes its direction; the scattering is assumed to be isotropic here. The total scattering cross-section  $\alpha(v)$  can now be obtained by integrating  $d\alpha$  over  $V$  and  $\mu$ . If we assume, in addition, that a particle cloud decays according to a first-order rate law, the "life-time" of an individual particle is exponentially distributed. The probability that the particle will not decay in the time interval  $\Delta t$  is, therefore,  $\exp(-\alpha \Delta t)$ , where  $\alpha$  is the decay-coefficient. The corresponding "mean life-time" is given by  $L = 1/\alpha$ .

In the case of a gas-mixture entered by a polluted particle, one will obtain a decay coefficient linearly composed of the decay-coefficients  $\alpha_i$  corresponding to all the gas-components  $i$  representing the mixture. The (partial) densities  $\delta_i$  have to be taken from the eddy-size spectrum offered by turbulence theory.

Example: If the moderating gas consists of uniform particles of mass  $M$  (= multiple of the incoming eddy mass) and if the gas particles have a Maxwellian velocity distribution

$$(18) \quad g(V) dV = 4a^2 V^2 \exp(-a^2 V^2) dV$$

where  $a^2 = M/2k\Theta$  and  $\Theta =$  gas temperature, we obtain for the decay-coefficient

$$(19) \quad \alpha(v) = \sigma \delta \left[ \frac{\exp(-b^2)}{\sqrt{\pi} b} + \left(1 + \frac{1}{2b^2}\right) \operatorname{erf}(b) \right]$$

where  $b = v/a$  (WIGNER-WILKINS (1944)).

Such a description of the turbulence field is called a "temperature concept" of turbulence. Let us now look at the collision process itself.

We base our model upon the following hypotheses:

1) The streaming air will be characterized by the velocity vector  $\vec{u}_m$ . The interspersed turbulence elements possess known size-spectra and particle-densities, and a velocity  $V\vec{\Omega}$  relative to  $\vec{u}_m$ , where  $V$  satisfies the distribution  $g(V)dV$  and  $\vec{\Omega}$  is a uniformly distributed random-variable.

2) All the eddies are clusters.

3) Inside the effective source domain (ESD), the life-time of a polluted eddy is longer than the time of the free flight (TFF) between two collisions, i.e. each collision is a scattering without change of pollutant content.

4) Outside the ESD, the life-time and the TFF are identical, i.e. at each collision point the chain will be passed through:

absorption  $\longrightarrow$  disintegration  $\longrightarrow$  coagulation  $\longrightarrow$  emission.

The pollutant content is usually changed thereby.

#### b. Equilibrium

The eddies will be classified according to their sizes. At the collision point each polluted eddy disintegrates into its constituents, the "atoms"

and consequently augments the local pollutant density. All the atoms simultaneously accumulated in the volume element including the collision point are at this moment disjoint, but coagulate afterwards in a manner specific for the collision point, and finally leave the volume element, so reducing the pollutant density there. The same process occurs at each collision point.

For the mathematical description one assumes that there are eddies of different size-categories, characterized by the number of atoms involved. Each polluted atom of an incoming eddy may be attached to any category during the collision. The corresponding probability has to be chosen from the local eddy size-spectrum. The attachment takes place via a linear indexing of the arriving polluted atoms by a random address. The interaction mechanism of the next collision is characterized again according to both the local situation and the category to which the respective atom belongs. The pollutant concentration at the collision point increases instantaneously but only by the quantum carried by the polluted atom under consideration, since the eddy to which it belongs is only a temporary vehicle. The other polluted atoms traveling in company with it will be separately followed along their way from emission point to detector. The mean local expansion velocity of an eddy cloud depends essentially on both the local eddy size-spectrum and the "mean turbulence temperature",  $\Theta_i$ , there. This model also allows one to pay regard to a loss of pollutant, as caused by a wash-out, chemical transformations etc. The tool best suited here is the so-called space-time dependent "importance factor".

### c. Thermalization

The puff leaving a chimney is part of a hot-gas stream which possesses a considerable vertical momentum being transmitted over time to the surrounding atmosphere. There is a mixing domain with a strong turbulence, a pronounced mean vertical velocity, a higher mean turbulence temperature and a specific eddy size-spectrum: the so-called "effective source domain" of the chimney. From here, the pollutant eddies start their way through the phase of equilibrium. An estimate of the dimen-

sions of this source domain is of importance for practical purposes. One could take here a so-called "plume rise formula" which, in general, will be determined in an experimental way (BRIGGS (1969)). In stationary cases the velocity field near to the source is governed almost exclusively by the hot waste gas stream. Turbulence theory gives corresponding information subsumed under the key words "the turbulent hot free jet". This given velocity field is entered by a pollutant puff, i.e. an eddy cloud. Scattering mechanisms can be constructed there on different hypotheses, e.g. with the picture of inelastically colliding particles, or with that of interacting vortex rings (THOMSON (1883), FOHL-TURNER (1975)).

#### d. Buoyancy

Buoyancy effects modify the momentum of the eddies according to the thermal stratification of the atmosphere. One refers to it as "stable" (unstable) if the stratification damps (intensifies) the vertical motions of air parcels and "neutral" if it does not influence the motions. Along the relatively short flight between two consecutive collisions the buoyancy  $K$  itself usually does not change significantly; as modification of the velocity vector  $\vec{v}$  we can take  $d\vec{v} = (0,0,Kdt)$ . The literature gives the expression

$$(20) \quad K = \frac{g}{\Theta} \left( \frac{\partial \Theta}{\partial z} + \Gamma \right) = R_i \left( \frac{\partial \bar{u}}{\partial z} \right)^2$$

coupling the force  $K$  with the Richardson number  $R_i$ , i.e. with the gradient of the  $\ln \Theta$  - vertical profile (ELIASSEN-KLEINSCHMIDT (1957)).

#### e. Boundary Conditions

At solid and liquid interfaces the particles are partly reflected and partly absorbed. Because of surface roughness the reflection is mostly diffuse. The use of an isotropic angular distribution seems to be adequate. If the characteristic length of surface roughness is smaller than the eddy size the surface is assumed to be smooth.

The space-time domain under consideration is, in general, finite. Its boundary shape and the particle behaviour there can be chosen arbitrarily. It may happen, that a particle moving around crosses the space boundaries of the region several times (in  $\longleftrightarrow$  out), before it (1) finally leaves the region or (2) is absorbed or (3) the observation time is over. All these possibilities represent - for the observer - a suitable termination of the single-particle history. To avoid unnecessary use of computer time, we shall construct a cut-off mechanism to treat multiple crossings. The so-called "albedo", i.e. a certain reflection factor,  $q < 1$ , seems to be best suited here.

Every particle emitted bears the "weight"  $w = 1$ . After  $n$  boundary crossings (in  $\longleftrightarrow$  out), however,  $w = q^n$ . By the introduction of a lower-limit  $q_{\min}$ , we shall judge the history finished when  $q^n \leq q_{\min}$  (MÜLLER (1975)).

At thermal inversion layers one can also observe a partial reflection. Here too the albedo concept is a good way to parametrize the rejection phenomenon. Corresponding measurements are inevitable.

Some attention should be paid to time-dependent boundary conditions, which are usually associated with inversion layers. Since the inversion dynamics is of lower frequency than the reflection process, the albedo concept mentioned above can be applied without any change. One needs only be aware of the actual position and spacing of the layer.

#### 4. Chemical Reactions

The concentrations of the different chemicals relevant to air pollution change in time and according to the local concentration ratios which, in their turn, change in space and time due to eddy diffusion. The adequate mathematical tool is, as already said, the "first collision concept". Another fact to be taken into account is, that the chemical concentrations inside an eddy "grow older" in the same way as the concen-

trations remaining at the birth place of the eddy do. The corresponding formalism is given by the iteration system

$$(21) \quad \left\{ \begin{array}{l} \chi_i^j(\vec{r}, 0) = \lim_{t \rightarrow 0} \chi_i^{j-1}(\vec{r}, t) \\ \frac{\partial \chi_i^j(\vec{r}, t)}{\partial t} = F_i(\chi_1^j, \chi_2^j, \dots) \\ \chi_i^{j+1}(\vec{r}, t) = \chi_i^j(\vec{r}, t) + \sum_{\vec{\rho}, \tau} \varphi(\vec{r}-\vec{\rho}, t-\tau) p(\vec{\rho}, \tau) \chi_i^j(\vec{\rho}, t) \end{array} \right.$$

where the index  $j$  counts the collisions and  $\chi_i^0(\vec{r}, 0) = \chi_o(\vec{r})$  gives the known background distribution of substance  $i$ , and  $\varphi(\vec{r}-\vec{\rho}, t-\tau)$  denotes the transfer function between birth place  $(\vec{\rho}, \tau)$  and first collision point  $(\vec{r}, t)$ . The  $\varphi$  has to be calculated,  $\varphi(\vec{r}, t) = \mathbf{S}g(\mathbf{m}, \vec{v})$ , from the solution  $g$  of the version,  $\beta \equiv 0$ , of equation (1), for a Dirac source,  $S \sim \delta(\mathbf{r}) \delta(t)$ ;  $p \cdot \chi_i$  is the amount of substance  $i$  carried away from  $(\vec{\rho}, \tau)$  by eddies. The quantity  $p$  depends on the eddy density. Stationary turbulence means  $\frac{\partial p}{\partial t} \equiv 0$ . The concentrations  $\chi_i$  after  $n$  collisions result from  $n$  repetitions of system (21). This procedure represents for an increasing  $n$  a successive approximation to the measurable Eulerian concentrations, i.e.

$$(22) \quad \chi_i(\vec{r}, t) = \lim_{n \rightarrow \infty} \chi_i^n(\vec{r}, t)$$

A first order approximation can be obtained by taking the mean life-time  $L$  instead of the real life-time, i.e. replacing the statistical distribution by its mean value.

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