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Gilice tér 39, H-1181 Budapest, Hungary
E-mail: gmajor@met.hu or *antal@met.hu*
Fax: (36-1) 290-7387

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Temporal variation of the atmospheric sulfur budget over Hungary during 1980–1996

László Bozó

Hungarian Meteorological Service,
H-1675 Budapest P.O. Box 39, Hungary
E-mail: bozo@met.hu

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Abstract—Annual atmospheric sulfur budgets over Hungary for the period of 1980–1996 are investigated in the paper using simple mass balance equations. It was found that the sulfate budget is almost balanced over the country, however, regarding sulfur-dioxid, Hungary is still a net atmospheric exporter in spite of the fact that Hungarian SO_2 emission decreased significantly during the past years. Data for the atmospheric sulfur concentration and deposition gained from the Hungarian regional background air pollution monitoring network are also presented for the period investigated.

Key-words: SO_2-emission, sulfur deposition, atmospheric budget, long-range transport.

1. Introduction

Sulfur components play significant role in acid rain formation. In addition, fine size range sulfate particles can reduce the effect of potential global warming — at least on regional scale — through the direct reflection of incoming solar radiation back to the space. Emission of SO_2 has changed significantly in Europe during the last 15 years which should also be resulted in the temporal variation of the atmospheric sulfur budget of Hungary. On the basis of the method used, the horizontal transport of sulfur can be estimated so conclusions can be drawn on the role and its temporal variation of Hungary in the European atmospheric sulfur budget.

The calculations were made for a box over Hungary up to the tropopause without any exchange with the stratosphere. It is assumed that there is no accumulation of sulfur in the box over a year's period which means that the gains and losses of the box are equal on an annual average.

2. Equations used for the calculations

The mass balance equations for SO_2 and sulfate can be given as follows:

$$E + A_{im} = D_{dry} + D_{wet} + T + A_{ex}, \tag{1}$$

$$T + A^*_{im} = D^*_{dry} + D^*_{wet} + A^*_{ex}, \tag{2}$$

where

E: SO_2 emission from the surface,
A_{im} and A^*_{im} : import terms for SO_2 and sulfate,
A_{ex} and A^*_{ex} : export terms for SO_2 and sulfate,
D_{dry} and D^*_{dry}: dry deposition of SO_2 and sulfate,
D_{wet} and D^*_{wet}: wet deposition of SO_2 and sulfate,
T: mass of SO_2 transformed into sulfate over the country.

Let

$$AD = A_{im} - A_{ex}, \tag{3}$$

$$AD^* = A^*_{im} - A^*_{ex}. \tag{4}$$

These terms are calculated using the mass balance equations (Eqs. (1) and (2)):

$$AD = D_{dry} + D_{wet} + T - E, \tag{5}$$

$$AD^* = D^*_{dry} + D^*_{wet} - T. \tag{6}$$

3. Emission of sulfur compounds in Hungary

The natural source strength of sulfur species has been estimated on the basis of data compilation by *Várhelyi* and *Gravenhorst* (1981). These authors estimated the average continental emission to be 0.01–0.03 g S m^{-2} a^{-1}. Thus, the corresponding value for Hungary is in the order of 10^{-3} Tg S a^{-1} (1 Tg = 10^{12} g). Anthropogenic sulfur emission data — which is represented in the form of SO_2 release — are based on the official national reports of the Ministry of Environment, Hungary. The annual emission values are shown in *Fig. 1*. It can be seen that the release of sulfur dioxid has been decreasing since the early 80's. Its value was 0.82 Tg S a^{-1} in 1980 and 0.38 Tg S a^{-1} in 1996. The decrease can be explained by the reduced industrial activity of the country and — from the 90's —

by the replacement of out-of-date technologies by less energy consumer and more environment friendly ones. Coal, which was used for home heating widely during the previous decades is now also being replaced by natural gas which causes only very low sulfur emission from the heaters.

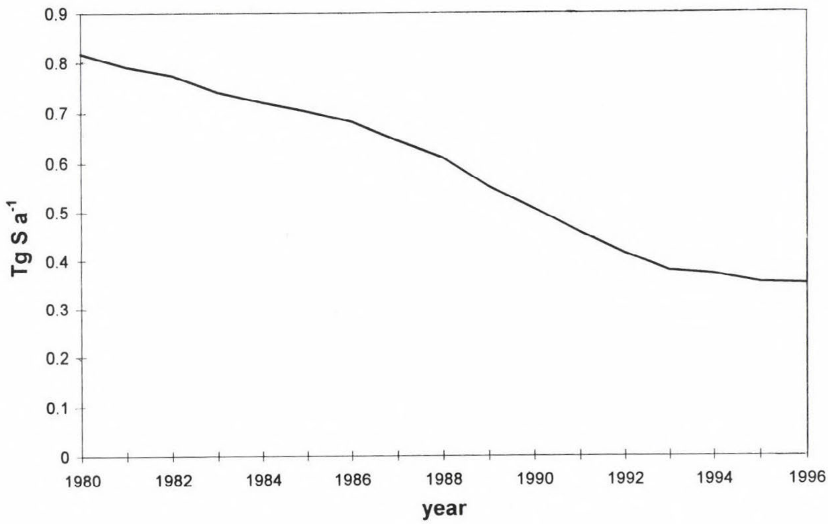

Fig. 1. Temporal variation of SO_2 emission from Hungary.

4. Concentration of SO_2 and sulfate over Hungary

Monitoring of SO_2 and sulfate under regional background conditions was started in the 70's in Hungary. The longest Hungarian measurement record is available from K-puszta. In the early 80's Farkasfa station also joined the SO_2 monitoring activity. Since 1996 there have been two additional regional background stations in operation in Hungary measuring also SO_2 and sulfate. Locations of stations are mapped in *Fig. 2.*

Temporal variation of SO_2 and sulfate concentrations under regional background conditions at K-puszta station is shown in *Fig. 3.* Due to the fact that during the period of 1992–1994 a lot of SO_2 data were missing for technical reasons, annual averages used in the atmospheric budget calculations were taken from the EMEP calculations (*Barrett et al., 1995*). One can see from the figure that SO_2 concentrations were higher in the first half of the period investigated (1980–1988) than in the second one. It is in accordance with the reduced sulfur emission from Hungary and other European countries.

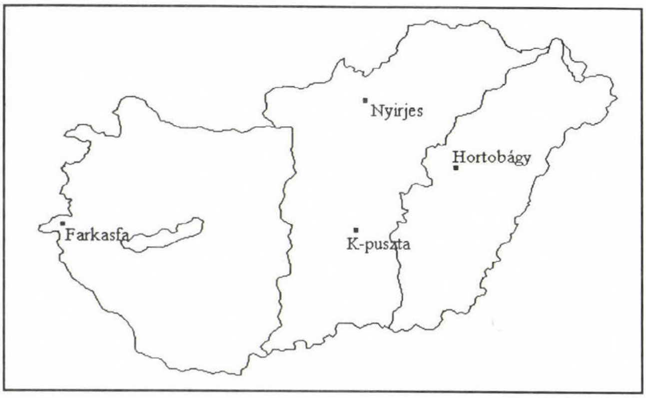

Fig. 2. Regional background monitoring stations in Hungary.

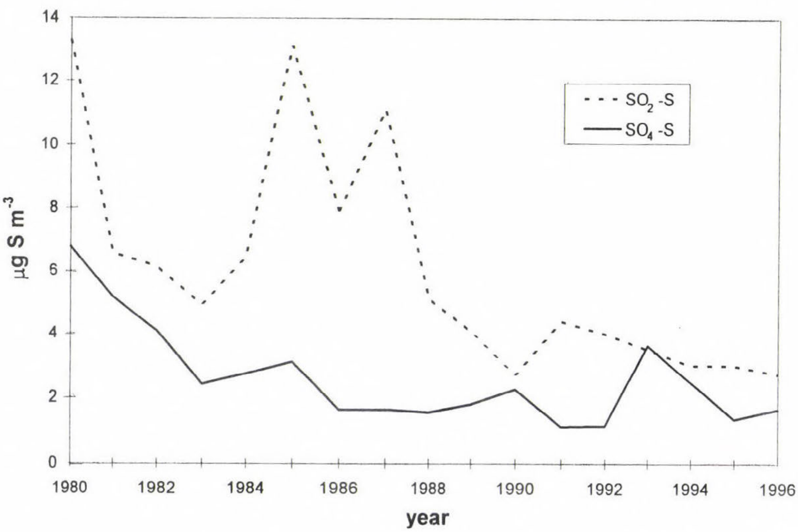

Fig. 3. Temporal variation of SO_2 and sulfate concentration.

5. Atmospheric SO_2-sulfate transformation and sulfur deposition over Hungary

The oxidation process of SO_2 in the troposphere is initiated by its reaction with free OH^- radicals producing SO_3 in two steps. Then SO_3 reacts with water vapour producing sulfuric acid. Transformation ratio can be estimated by both laboratory experiments and direct field measurements. The results show relatively big variation (*Warneck*, 1988). Laboratory experiments indicate lower values for the intensity of transformation which can be explained by the fact that liquid phase atmospheric reactions might speed up the transformation of sulfur dioxide into sulfate. In present paper the transformation ratio of 3.6%/hour was used for the calculations measured by *Horváth* and *Bónis* (1980).

Dry deposition of atmospheric sulfur compounds can be determined by multiplying the averaged concentration near the surface and deposition velocity. This latter value was taken from the field measurements of *Horváth et al.* (1996) who carried out vertical profile measurements for SO_2 and meteorological parameters in Hungary so that to estimate the dry flux of SO_2 from the atmosphere onto the surface. Based on their measurements and calculation an annual average of 0.6 cm s^{-1} was used for atmospheric budget estimations. Dry deposition velocity for sulfate particles was taken from *Möller* (1983). Its value is 0.1 cm s^{-1}.

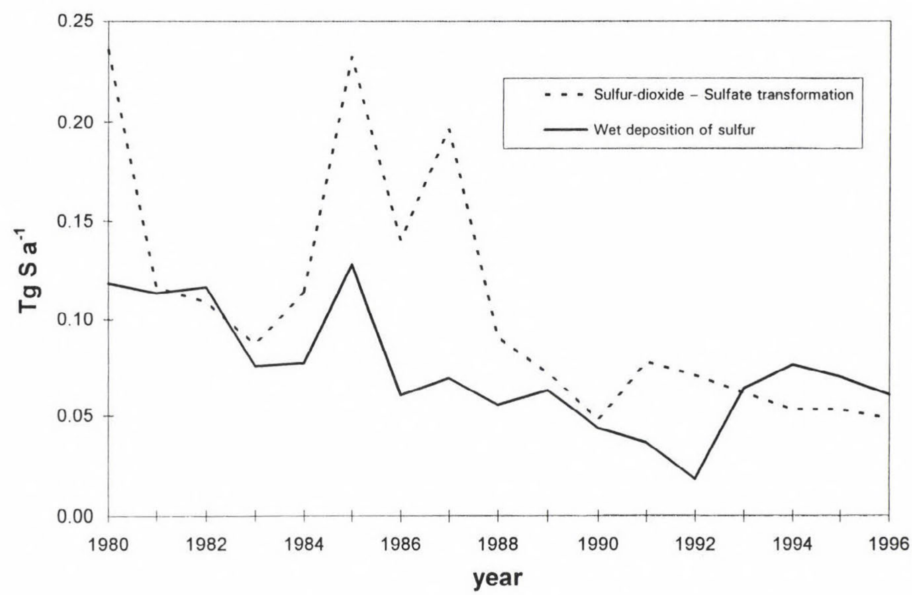

Fig. 4. Temporal variation of SO_2-sulfate transformation and wet deposition of sulfur over Hungary.

Long-term variation of sulfur wet deposition as well as SO_2-sulfate transformation is plotted on *Fig. 4.* Wet only samplers at 10 background sites of Hungary were used for collecting precipitation samples. Wet deposition of sulfur is estimated by averaging the wet deposition rates from the stations. It can be seen from the figure that the lowest deposition rate were detected in 1992 and the highest in 1985. Since the wet deposition of sulfur dioxide and sulfate can only been measured altogether as the sulfate content of precipitation, it was needed to separate the rain-out and wash-out processes for SO_2 and sulfate. Scavenging ratios applied in the EMEP model (*Barrett et al.,* 1995) were used for our budget calculations.

6. The atmospheric sulfur budget over Hungary

The differences between horizontal atmospheric import and export terms (see Eqs. (3) and (4)) are plotted in *Fig. 5.* Sulfate budget is more balanced as compared to that of sulfur-dioxide. Its largest negative value was calculated for 1987. During the last four years it was going to be close to zero. Regarding AD, its value was strongly negative in the early 80's. During the period of 1993–1996 it is stabilized at around -0.20 Tg S a^{-1}. It means that even in 1996, more sulfur dioxide was exported horizontally in the atmosphere from Hungary than the country imported from other countries, so Hungary is still playing a net exporter role in the European sulfur budget. In *Fig. 6* it is plotted how much percent of the Hungarian sulfur emission is compensated by total sulfur deposition and transformation. Its lowest value was detected in 1990 while the highest one — over 50% — in 1985.

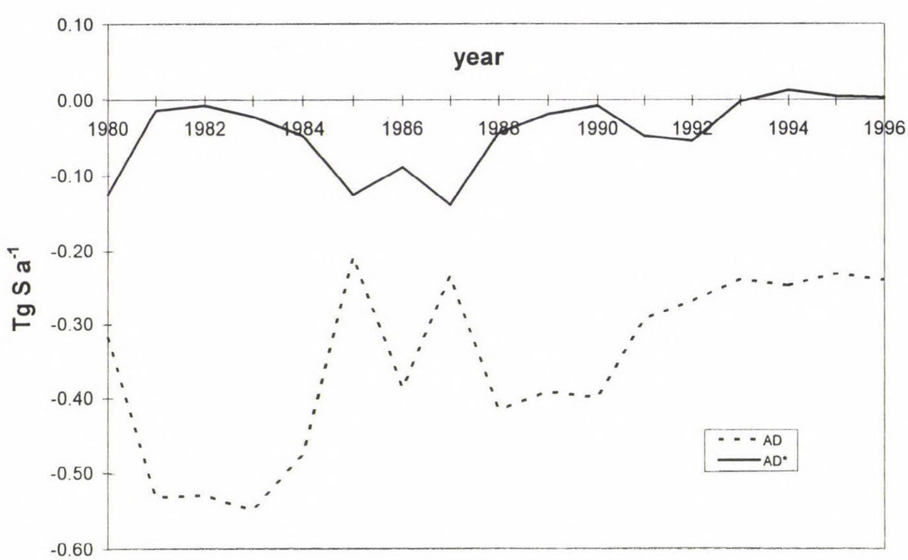

Fig. 5. Temporal variation of horizontal transport terms of sulfur over Hungary.

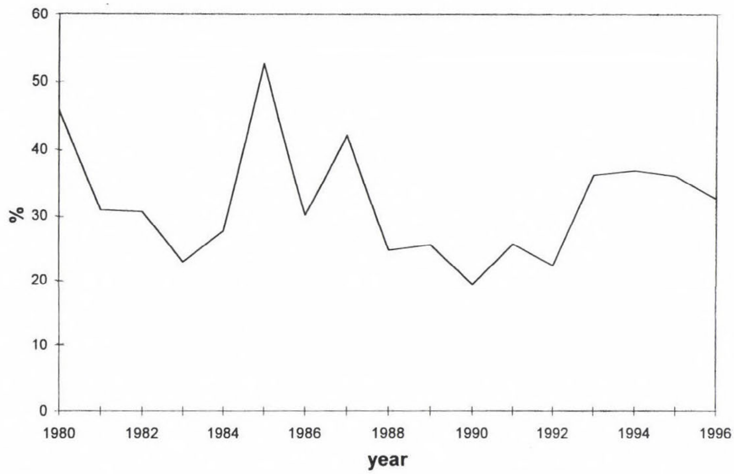

Fig. 6. Hungarian sulfur emission compensated by total deposition and transformation.

7. Conclusions

Although the sulfur dioxid emission of Hungary decreased rapidly during the last decade, the country still plays a net atmospheric exporter role for sulfur dioxide in Europe. Atmospheric sulfate budget is balanced, thus the amount of sulfate imported by the country is roughly equal to that of exported by Hungary. It is expected that sulfur budget will also vary in the future due to the continuous completion of the second Sulfur Protocol: Hungary should reduce its sulfur-dioxide emission by 50% until year of 2005 and by 60% until 2010 as related to the emission level reported in 1980. On the basis of these numbers, Hungarian SO_2 emission in 2010 must not exceed the limit of 0.327 Tg S a^{-1}.

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Non-parametric estimation of climate trends

István Matyasovszky

Department of Meteorology, Eötvös Loránd University,
H-1083 Budapest, Ludovika tér 2; E-mail: matya@ludens.elte.hu

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Abstract—One of the most important statistical characteristics of climate variables is their expected value. Under a changing climate the expected value can also change. The expected value at a given time can be simply estimated by averaging observations being in an interval around this time. A basic question is how to define the length of this interval. It may be supposed that a weighted average results in a more accurate estimate than ordinary averaging. How to choose weights in that case? As the intensity of change of climate can also change it is reasonable to use variable averaging time length. Which techniques are available for this purpose? In the paper, a summary of mathematical details is presented and three examples are shown to illustrate the importance of above questions.

Key-words: smoothing, weighted local regression, bandwidth, kernel function.

1. Introduction

Consider the observations $y_0, y_1, \ldots, y_n$ of a climate variable at $t_0 < t_1 < \ldots < t_n$ in the following form

$$y_j = f(t_j) + e_j, \quad j = 0,\ldots,n$$

where $f(t)$ is the so-called trend function representing temporal behavior of the expected value. We may generally assume that the superposing fluctuations come from a white noise process, i.e.,

$$E[e_j] = 0, \quad E[e_j^2] = d^2, \quad E[e_i e_j] = 0, \quad i \neq j$$

where E denotes the expected value operation.

There are two basic approaches to estimate trend functions. One of them includes the *parametric method* when $f(t)$ is considered as a linear combination of known functions $g_0(t),\ldots, g_{k-1}(t)$ $(k > 1)$. The task is then to estimate optimum parameters of the linear combination generally by the method of least squares. Besides the simplicity of this procedure, an important advantage is its ability to assess the statistical significance of changes in observed series. However, the functions $g_0(t),\ldots, g_{k-1}(t)$ are generally not known in practice and an appropriate choice of them is a difficult problem.

The other basic approach includes *non-parametric methods* when no assumption on the shape of $f(t)$ is needed. The trend function is then approximated locally by different polynomials at each t. This results in the following weighted averaging of observations:

$$\hat{f}(t) = \sum_{j=0}^{n} w_j(t) y_j, \tag{1}$$

which is frequently called smoothing of data. Several techniques are available to choose the weights $w_j(t)$ (*Nadaraya*, 1964; *Cleveland*, 1979; *Gasser* and *Muller*, 1984; *Eubank*, 1988), which, however, have the same asymptotic properties. Asymptotics are defined such that the support of $f(t)$ is fixed, and taking $n \to \infty$ (i.e., distance between neighboring observations becomes smaller and smaller) the properties of estimation Eq. (1) are obtained. For simplicity but without any loss of generality the support of $f(t)$ is chosen as the interval [0,1] for mathematical formulation. (Obviously, the actual support of $f(t)$ can result in [0,1] after a scale transformation of time t.)

The following section summarizes asymptotic properties of non-parametric regression techniques. Choice of weights and the averaging time is also discussed here. Section 3 presents the best estimator Eq. (1) of regression functions $f(t)$ for fixed numbers of observations. Section 4 shows three examples to demonstrate some theoretical problems in practice. Finally, a section for discussion is provided.

2. *Asymptotic properties*

A reasonable assumption, when estimating $f(t)$, is that observations at instants close to t have relatively large weights, while observations far from t have small weights in Eq. (1). There are several traditional versions for the concept, such as binomial, exponential, or Gaussian smoothers. However, the choice of both the actual smoother and the averaging time is quite arbitrary and these smoothers reflect only basic characteristics of time series. Therefore, there was a need for a well-developed theory in order to overcome above difficulties. To illustrate this theory we are focusing on weighted local regression (WLR) (*Cleveland*, 1979). Weighted local version of the method of least squares at t results in

$$\hat{f}(t) = \hat{a}_0,$$

where $\hat{a}_0$ is obtained by minimizing

$$\sum_{j=0}^{n} (y_j - [a_0 + a_1(t - t_j) + \ldots + a_{k-1}(t - t_j)^{k-1}])^2 K((t - t_j)/h) \tag{2}$$

with respect to $a_0, a_1, \ldots, a_{k-1}$. The kernel function K determines how to decrease the influence of a squared difference in the sum of squared differences Eq. (2) when moving off t, and the bandwidth h determines the rate of this decrease.

In order to construct an estimate having "good" properties some criteria should be satisfied for the kernel. K is defined on the interval [-1,1] and is called of order k if

$$\int_{-1}^{1} K(z) z^j dz = \begin{cases} 0, & 0 < j < k \\ 1, & j = 0 \end{cases}$$

is satisfied. Then, assuming that kth derivative of f exists, under conditions $n \to \infty,\ h \to 0,\ nh \to \infty$
the bias

$$E[\hat{f}(t)] - f(t) = u_k(t),$$

the variance

$$E[(\hat{f}(t) - E[\hat{f}(t)])^2] = v_k^2(t)$$

and the mean squared error

$$MSE(t) = E[(\hat{f}(t) - f(t))^2] = u_k^2(t) + v_k^2(t) \tag{3}$$

of WLR are obtained asymptotically by

$$u_k(t) = \frac{(-1)^k}{k!} h^k B_k f^{(k)}(t), \quad v_k^2(t) = \frac{d^2}{nh} V_k f^2(t), \tag{4}$$

where

$$B_k = \int_{-1}^{1} K(z) z^k dz, \quad V_k = \int_{-1}^{1} K^2(z) dz,$$

and $f^{(k)}$ denotes kth derivative of f. Eq. (4) shows that the bias is proportional to $f^{(k)}$ and is small when the smoothing is small. The variance is proportional to f^2 and is large when the smoothing is small. A small bias, therefore, requires small bandwidths, while a small variance needs large bandwidths. A trade-off between the two requirements, i.e., an estimate of the optimal bandwidth could be defined by minimizing the asymptotic integrated mean squared error

$$IMSE = \int_0^1 (u_k^2(t) + v_k^2(t))\,dt \tag{5}$$

when the trend function and its kth derivative were known. Due to lack of knowing these functions an unbiased and consistent estimate of Eq. (5) is constructed as

$$CV(h) = \frac{1}{n+1} \sum_{i=0}^{n} (y_i - \hat{f}_i(t_i))^2 \tag{6}$$

and its minimum with respect to h gives the optimal bandwidth. Here $\hat{f}_i(t_i)$ is an estimate Eq. (1) of $f(t_i)$ but leaving out y_i.

Eqs. (3) and (4) show that the mean squared error of WLR depends on properties of f. This fact motivates the choice of locally varying bandwidths. A smaller bandwidth where f is rough ($f^{(k)}$ is, in absolute value, large) reduces bias and a larger bandwidth in the smooth regions of f ($f^{(k)}$ is, in absolute value, small) reduces variance. In general, construction of a local bandwidth estimator entails a two-step procedure. The first step produces a pilot estimator using a fixed bandwidth, and the second stage yields the local bandwidth estimator. For instance, having a global bandwidth obtained by minimizing the quantity Eq. (6) an estimation of $f(t)$ is performed, then using a finite difference scheme its kth derivative is also estimated. We mention that derivatives of trend functions can be directly estimated from observations using some generalized versions of non-parametric regressions (*Fan* and *Gijbels*, 1995). $f(t)$ and $f^{(k)}(t)$ in Eqs. (3) and (4) are then substituted by their corresponding estimates, and Eq. (3) is minimized at each t.

Note that the above technique is based on asymptotics. One can hope that usage of exact properties can yield better estimates. Exact bias and variance can be written as

$$u(t) = \int_{-1}^{1} K(z) f(t - h(t)z)\,dz - f(t),$$

$$v^2(t) = \frac{1}{nh(t)} \int_{-1}^{1} K^2(z) f(t - h(t)z)\, dz.$$

After substituting *f(t)* by its pilot estimate, the exact mean squared error

$$MSE(t) = u^2(t) + v^2(t)$$

is minimized at each t to have local bandwidth (*Staniswalis,* 1989). A further advantage of the method is that it does not require derivatives of f. It is crucial in areas where $f^{(k)}$ equals to zero and, therefore, the estimated local bandwidth can be extremely large when using asymptotic formulae. An important question is how the local bandwidth depends on pilot global bandwidths. Theoretical considerations and simulation experiments show that for a large range of pilot bandwidths, the estimated trend functions differ slightly (*Staniswalis*, 1989).

Another important question is the choice of kernel. Evidently, it is useful to choose that kernel which delivers the smallest asymptotic variance for a fixed k. These kernels for different k can be found in *Gasser et al.* (1985). *Muller* (1984) produced kernels minimizing variance of *m*th ($m \geq 0$) derivative of $\hat{f}$. The only remaining question now is the order of kernels, i.e., the choice of k. Theoretical reasons and simulation work show that a small value of k is reasonable. Generally, $k = 2$ (locally linear approximation of f) is satisfactory since the choice of bandwidth is a much more serious problem than the choice of the order of the kernel (*Simonoff,* 1996).

3. *Optimal smoothing*

Knowing asymptotic properties of non-parametric techniques is highly useful for several reasons. However, the final question is that which method in practice is preferred, when the number of observations is given, i.e., n is fixed. After *Fan* (1993), the best local linear estimation ($k = 2$)

$$\hat{f}(t) = \sum_{j=0}^{n} \alpha_j(t) y_j \tag{7}$$

of f is the weighted local regression, and the weights can be calculated as

$$\alpha_j(t) = w_j(t) / \sum_{i=0}^{n} w_i(t),$$

where

$$w_j(t) = K((t - t_j)/h)\,(s_2 - (t - t_j)s_1), \quad j = 0,\ldots,n$$

and

$$s_l = \sum_{j=0}^{n} K((t - t_j)/h)\,(t - t_j)^l, \quad l = 1,2.$$

Furthermore, K is

$$K(z) = \frac{3}{4}(1 - z^2), \quad z \in [-1,1],$$

the so-called Epanechnikov kernel (*Fan*, 1992).

The optimality of Eq. (7) is the minimax optimality over linear smoothers, which is defined as follows. Under quite weak assumptions for f (*Fan*, 1993), WLR based on the Epanechnikov kernel achieves the minimum value over all linear smoothers of the maximum of $MSE(t)$, and has a minimax efficiency at least 89.6%. In other words, the maximum of $MSE(t)$ of WLR with Epanechnikov kernel is at least 10.4% smaller than the maximum of $MSE(t)$ of any other linear smoother.

4. Examples

The first example demonstrates that non-parametric regression can be a more effective technique to estimate trends than parametric regression. Annual temperature anomalies over Northern Hemisphere from 1854 to 1993 (*Jones et al.*, 1994) are examined by fitting a common linear trend and using WLR. Evidently, linear trend basically describes the warming tendency but with considerable departures from data during several subperiods of the entire period (*Fig. 1*). The trend obtained by WLR fits much better the data. In addition to the good fit, WLR provides a smoother curve than a frequently used, say 10-year, running mean (not shown here). Another curve corresponding to WLR with local bandwidth is also presented in Fig. 1. Difference between trends estimated with global and local bandwidths is not considerable due to the small variance of data. The small variance makes it possible to use small global bandwidth (15 years) resulting in a small bias with small variance. Therefore, a somewhat larger or smaller local bandwidth at a given time t provides almost the same estimate.

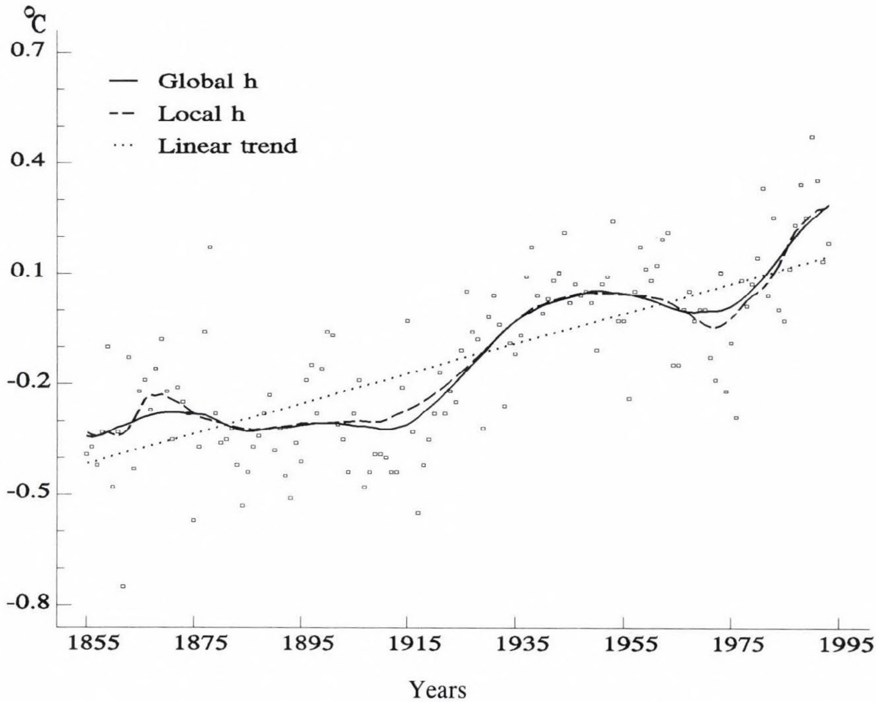

Fig. 1. Trend of annual temperature anomalies over Northern Hemisphere.

The second example represents that quantity Eq. (6) is sometimes a highly complicated function of h. How does the shape of trend functions influence the behavior of Eq. (6)? To illustrate this problem let's consider monthly SOI time series. SOI (Southern Oscillation Index) is defined as the difference between sea surface air pressures of Tahiti and Darwin, and is used to characterize ENSO phenomena. SOI data set has been obtained from NCAR (National Center for Atmospheric Research) for the period from 1868 to 1994. The question to be answered is whether a tendency in strength of ENSO exists. Global minimum of the quantity Eq. (6) has been found at 9 months. This is in a good agreement with the fact that SOI has intensive fluctuations on a 2–6-year scale, which can be reproduced by only a small bandwidth. However, a secondary minimum of Eq. (6) exists at a 36-year value. This bandwidth is related to long-term changes.

Fig. 2 shows that there is a strong decreasing tendency of SOI from the middle of the seventies, which is well reproduced by the curve estimated by WLR. To sum up, the bandwidth to be used depends on the underlying problem, and the selection between global and local minima of Eq. (6) cannot be automatically performed.

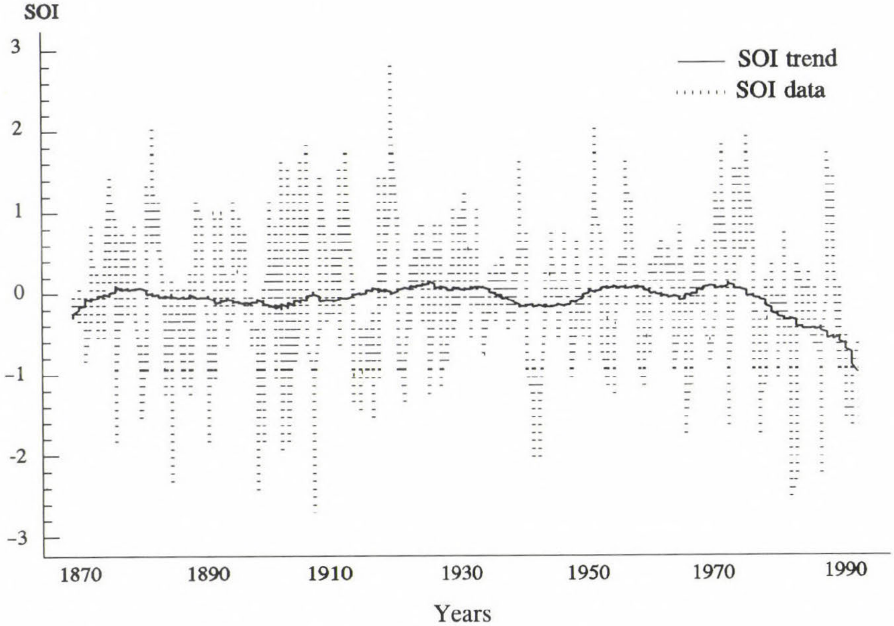

Fig. 2. Trend of SOI estimated with 36-year global bandwidth.

In the first example there was no considerable difference between curves obtained by WLR with global and local bandwidths. The following case illustrates the usefulness of local bandwidths. Central England monthly mean temperature data are available since 1659 (*Manley*, 1974). Trend of these annual mean temperatures has been estimated by WLR using global bandwidth and local bandwidth as suggested by *Staniswalis* (1989). Two important points can be drawn from *Fig. 3*. The first one is that the two curves corresponding to global and local bandwidths are very similar, and both have intensive fluctuations until the XIX century. This is probably due to the low reliability of data originating from this time. Just enough to mention the term "Central England" because location of measurements changed several times during the period of observations. In spite of deep work on data, this period cannot be considered homogeneous (*Malcher* and *Schönwiese*, 1987). The second conclusion is that the trend function estimated with global bandwidth remains highly fluctuating during second part of the whole period, while the curve obtained with local bandwidth is quite smooth. This is because first part of data produces the global bandwidth to be small (6 years), but the local bandwidth adapts quickly to homogeneous part of data and exhibits a smoothness required for "nice" trend functions.

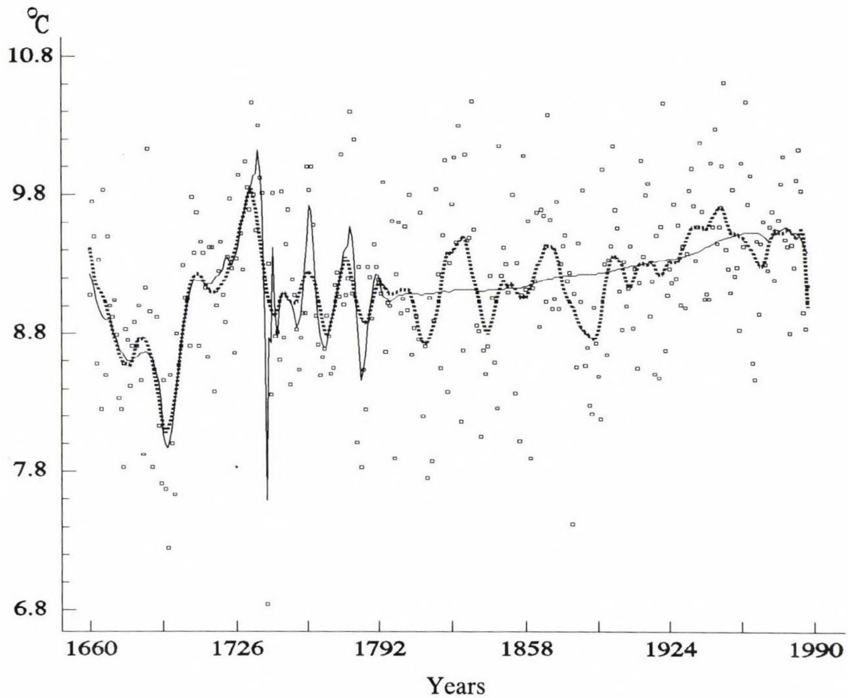

Fig. 3. Trend of Central England annual mean temperatures estimated with local bandwidth (solid line) and global bandwidth (dotted line).

5. Discussion

The paper summarized the most important mathematical details of non-parametric trend estimation, and three examples were shown to illustrate the importance of theoretical questions.

A reviewer suggested to use the expression *signal* instead of *trend* arguing that the deterministic part of climate data sets generally contain a long-term change and a short term interannual variability. This is certainly true, but every deterministic member of any data set may be inserted into a common component, called trend. Following the terminology of statistical literature, we decided to use the term *trend*, because the paper is principally a theoretical one. However, distinction between long-term change and interannual variability is really an important issue as illustrated by our second example. As the trend is a superposition of components with different time scales, the quantity Eq. (6) has various local minima, each of them corresponds to specific components of the trend function.

An important message of the third example is the usefulness of local bandwidths. Fig. 3 shows that local bandwidth can deliver a much more realistic estimate of the trend function when data set has inhomogeneities. We mention that a modified and further developed version of non-parametric trend estimation is able to detect data inhomogeneities (*Muller*, 1992). The principle of the technique is that two estimates are defined at each time t such that the first one uses data preceding t, while the second one uses only succeeding data. After some normalization, the maximum of the difference between these two estimates can be applied as a test statistic to detect change point in a time series. This test shows strong inhomogeneities in Central England temperature data set until the end of XVIII century.

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Two statistical methods of long-time prognoses

László István Orbán

H-8200 Veszprém, Török Ignác u. 12/a, Hungary

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Abstract—In this paper the expected mean values of meteorological elements at Cluj-Napoca (Kolozsvár; Romania) are estimated by two statistical methods. The first method is based on the application of the harmonic analysis. The study is performed for 90-year time series of mean values of temperature and precipitation. The second method is correlation analysis carried out between mean temperatures and precipitation amounts of given month and the preceding months for the same 90-year data set. The methods gave acceptable prognoses for the monthly mean temperatures, but have not been proved to be applicable for precipitation forecasting yet.

Key-words: statistical forecast, harmonics, correlation analysis, long data series.

1. Introduction

Long-range forecasting frequently applies statistical methods because of the complexity of the hydrodynamical differential equations and the insufficient density of the meteorological observing network, mostly over the oceans. In this study, first of all, the estimation of the monthly mean values of temperature at Cluj-Napoca are attempted by two statistical methods. The first approach is the application of the harmonic analysis in which long time series (annual, monthly values) of meteorological elements are decomposed into harmonic components. The dominant harmonics with the greatest amplitudes are selected, and the phase angles of those are defined from the registered data of the previous period. The prognosis is based on the extrapolation of the obtained phase angles. The dominant harmonics are probability representatives of some physical process. (E.g. the harmonic with a period of 11.2 years expresses the period of sun activity.)

An application of this method to forecast the annual precipitation amount at Cluj-Napoca has already been described in journal Időjárás (*Orbán* and *Pap*, 1968). The prognosis used precipitation data of the three preceding years by defining the phase angles of the three harmonics. That time, due to the lack of computer, a nomogram technique was presented to give an approximate solution of the trigonometric equation.

The second approach is the correlation analysis. In this case the forecast is made by using the correlation coefficients between the values of meteorological characteristics for a given term (e.g. year, season, month) and those for preceding terms. The regression coefficients are determined with the method of minimum squares.

For developing the methods, the computer assessment was provided by the Agronomic Institute of Cluj-Napoca.

2.1 Harmonic analysis

In 1992 we analysed the monthly values of the temperature and the precipitation for the previous 90 years (1080 months) on the basis of the data gained at Cluj-Napoca with harmonic analysis. *Fig. 1* shows frequencies and periods of amplitudes of the dominant harmonics (*Bozac,* 1993). (The values marked with asterisks represent the drawn values of the near harmonics.) It is worth observing that the harmonics corresponding to the sun activity (n = 8; T = 135 months) have an insignificant amplitude in the analysis of the monthly values (amplitude of the precipitation is not even indicated for its small value).

First, it was attempted to find a computer assisted method for the forecasting of the monthly mean temperature on the basis of the harmonics with the biggest amplitude (these are periods of 12; 1.1; 1.08; 1.005 months on Fig. 1). Then the mean temperature of the respective month can be approached with the equation:

$$y(t) = \sum_{k=1}^{4} x_k \sin(\theta t n_k + i_k) + t_m. \tag{1}$$

Here (t) denotes the respective month, x_k, n_k and i_k are the amplitude, the frequency and the phase angle of the corresponding harmonic respectively, while t_m is the annual mean temperature (t_m = 8.4°C for Cluj-Napoca). The total period (T = 1080) of the taken interval determines the θ value:

$$\theta = 2\pi/T \approx 0.005817764 \text{ rad/month.}$$

Considering the mean temperature of four consecutive months $y(0)$, $y(1)$, $y(2)$ and $y(3)$, by applying Eq. (1), we get the following equation system:

$$y(0) = \sum_{k=1}^{4} x_k \sin(i_k) + t_m, \tag{2}$$

$$y(1) = \sum_{k=1}^{4} x_k \sin(\theta n_k + i_k) + t_m, \tag{3}$$

$$y(2) = \sum_{k=1}^{4} x_k \sin(2\theta n_k + i_k) + t_m, \tag{4}$$

$$y(3) = \sum_{k=1}^{4} x_k \sin(3\theta\, n_k + i_k) + t_m. \tag{5}$$

This equation system contains twelve unknown factors. The i_k phase angle takes its values from the interval of $[0,\ 2\pi]$, while the x_k amplitudes and n_k frequencies differ only a little bit from the basic values of the harmonic analysis. We can find an exact or approximate solution of Eqs. (2) to (5). The expected mean temperature of the next month is given by

$$y(4) = \sum_{k=1}^{4} x_k \sin(4\theta\, n_k + i_k) + t_m. \tag{6}$$

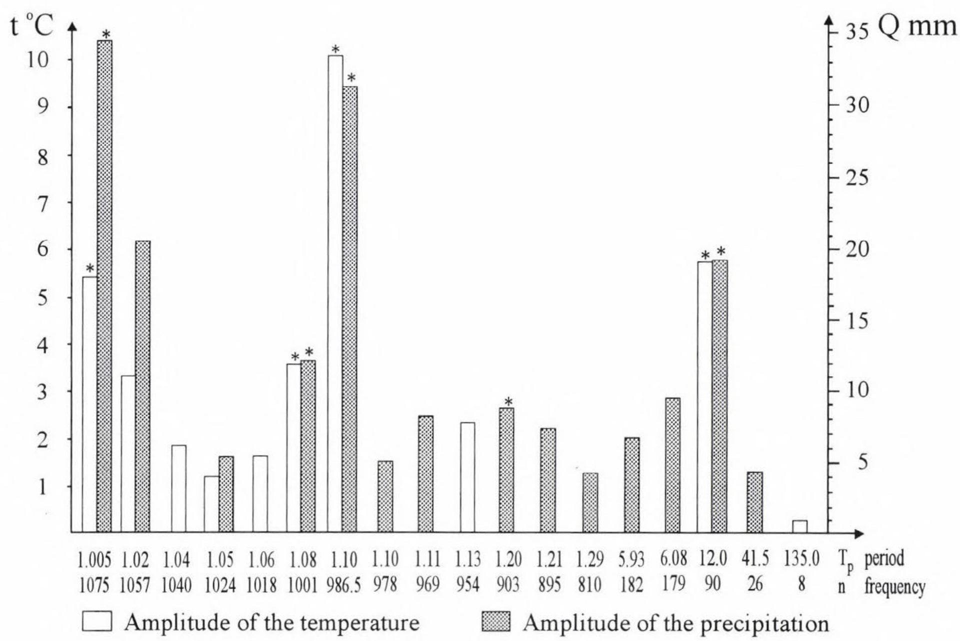

Fig. 1. The amplitudes of the dominant harmonics.

In the computer procedure we had to introduce the following expressions:

$$A_0 = \sum_{k=1}^{4} x_k \sin(i_k) + t_m - y(0), \tag{7}$$

$$A_1 = \sum_{k=1}^{4} x_k \sin(\theta n_k + i_k) + t_m - y(1), \tag{8}$$

$$A_2 = \sum_{k=1}^{4} x_k \sin(2\theta n_k + i_k) + t_m - y(2), \tag{9}$$

$$A_3 = \sum_{k=1}^{4} x_k \sin(3\theta n_k + i_k) + t_m - y(3), \tag{10}$$

$$E = |A_0| + |A_1| + |A_2| + |A_3|. \tag{11}$$

The computer calculates the possible values step by step using the developed software, and we have the best approximate solution when the value of E is minimal. When $E = 0$, the obtained factors satisfy the equation system Eqs. (2) to (5), because the sum of the absolute values can only be zero if each element equals to zero ($A_0 = A_1 = A_2 = A_3 = 0$), and then the computational equation system (Eqs. (7) to (10)) is equivalent to Eqs. (2) to (5).

However, it should be mentioned that the extent of the steps greatly influences the number of trials. If m represents the trial number for the unknown factor, then m^{12} calculations must be done because a given value has to be combined with each other value. Therefore, suitable precision by less calculation can only be achieved by arranging the operations into cycles. In cadre of a certain cycle the m value must be determined in accordance with the speed of the computer. The values obtained in the preceding cycle will provide the basic values for the succeeding cycle. In the following cycle narrower intervals and smaller steps are to be used, retaining the m value for the steps.

On the basis of this conception we made a prognosis for each month of the year of 1903 by using the mean temperatures of the preceding four months in Cluj-Napoca. We chose the year 1903 because the December of 1902 had been extremely cold (–7.1°C mean temperature) and wanted to see the effect of such extreme values in the application of our method. *Table 1* shows the differences between the realised and the calculated monthly mean temperatures. It is noteworthy that the differences do not exceed 1°C in the case of eight months, but there is such a month (April) when the deviation reaches 7°C.

This example illustrates the efficiency of the above procedure well. Improvement of the procedure can be achieved in different ways. Using a faster computer results bigger *m* steps and thus a more precise solution. Increasing the number of the selected harmonics and months would probably lead to better results but the number of operations would increase too. The accuracy of the prognosis may be increased by taking into account the correlation coefficients between the respective months.

Table 1. Computer prognosis for monthly mean temperatures of 1903, °C

	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
Predicted	-3.5	-0.7	6.0	14.5	15.1	17.0	16.5	15.7	15.4	9.2	4.4	1.0
Realised	-4.8	-0.6	6.0	7.5	14.1	17.3	19.2	17.8	14.8	9.8	4.6	0.3
Difference	-1.3	0.1	0.0	-7.0	-1.0	0.3	2.7	2.1	-0.6	0.6	0.2	-0.7

In the case of the temperature and other continual elements, not only the mean value but also the scattering of the values may be of interest. This characteristic can be determined by carrying out the harmonic analysis of the dispersion of the given element, and then we can adopt the above method for dispersion prognosis.

The application of the above method for the forecasting of the monthly precipitation amount is under way. The tests have shown that at least five harmonics should be selected. Acceptable prognosis of the precipitation can only be obtained if we select five months of the preceding period with two-month turns. For example, when predicting the precipitation in December we have to put in the software the precipitation amounts of the previous October, August, June, April and February. The possible correlation between the months can be taken into account by substituting for the *E* value of Eq. (11) with the relationship:

$$E = c_1|A_1| + c_2|A_2| + c_3|A_3| + c_4|A_4| + c_5|A_5|. \tag{12}$$

Here the coefficients c_1, c_2, c_3, c_4 and c_5 are proportional to the suitable correlation coefficients.

To characterise the distribution of the monthly precipitation we have to forecast the number of days with precipitation in a month and then to apply the above method for this quantity.

2.2. *Correlation analysis*

The second statistical forecasting method is based on the correlation analysis. On the computer of the Agronomic Institute of Cluj-Napoca we calculated the correlation coefficients between the given month and the preceding months of the previous year for both the monthly mean temperature and the monthly precipitation. The calculations were made with the data gained at Cluj-Napoca during the years of 1901 and 1991. *Table 2* contains the correlation coefficients between the monthly temperatures. Since much lower correlation coefficients occurred in the study of the precipitation, they are not included in our recent paper.

Table 2. The correlation coefficients of the monthly mean temperatures based on the data gained in (Cluj-Napoca)

	The preceding year											
	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
												*
J	-0.0	-0.03	0.15	0.01	-0.19	0.03	0.04	-0.15	0.07	0.06	-0.09	0.23
	***											
F	0.35	-0.05	0.08	0.07	-0.01	0.0	0.08	0.04	0.17	-0.05	0.03	0.12
		**										
M	0.13	0.28	0.06	-0.12	-0.03	-0.19	-0.05	-0.15	-0.09	0.07	-0.11	0.20
								*				
A	0.15	0.19	0.15	0.06	0.12	-0.09	0.09	0.21	0.07	-0.10	0.17	0.07
							**					
M	-0.02	0.07	-0.03	0.11	0.07	0.02	0.27	0.04	0.01	-0.15	0.03	-0.08
	*				*						*	*
J	-0.21	0.02	0.12	0.14	0.26	0.03	0.13	0.19	-0.06	-0.12	0.24	0.22
						**	**	***	*			
J	-0.07	-0.01	-0.04	0.20	0.17	0.29	0.32	0.43	0.21	0.07	0.11	0.12
				*	*		***	*				
A	-0.03	-0.04	-0.07	0.25	0.22	0.18	0.45	0.24	0.18	-0.11	0.15	0.04
	**	*				*	*	**				
S	-0.30	-0.22	0.0	0.19	0.17	0.25	0.25	0.29	0.0	-0.03	-0.01	-0.17
			*									
O	-0.08	-0.13	-0.23	-0.11	-0.11	-0.11	-0.12	-0.05	0.11	0.01	0.09	0.03
												*
N	-0.15	0.10	0.07	0.12	-0.01	-0.09	0.01	0.02	0.03	0.13	0.15	-0.23
D	-0.08	-0.10	0.09	-0.05	0.06	0.07	-0.03	-0.09	0.0	0.01	0.17	0.0
	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
	The same year											

* Asterisk are above the marked values

The values marked with asterisks indicate significant correlation with the threshold of reliability 5% (*), 1% (**) and 0.1% (***).

Using these correlations we have defined the following forecasting formula:

$$y_i = e_i + \sum_{k=1}^{4} (a_{ik} x_k^2 + b_{ik} x_k). \tag{13}$$

Here y_i is the expected monthly mean temperature, while x_k indicates the registered mean temperatures of the preceding months for which the correlation coefficient are relatively high and significant. The e_i, a_{ik} and b_{ik} values were determined with the method of minimum squares on the basis of the data of the registered 1080 months. Four months proved to be satisfactory for an acceptable prognosis. The study of this method is going to be published by the Agronomic Institute of Cluj-Napoca.

We can state that this simple statistical forecasting method gives good prognosis for monthly mean temperatures, but is not applicable for monthly precipitation prediction. For the improvement of the method it is advisable to use data gained at different meteorological stations which are located at various distances around the cardinal points and to correlate one element to other elements.

It has to be admitted that the above two methods can only be carried out with the help of powerful computers, but the quantity of the calculations is still much fewer than that of the methods based on hydrodynamical equations.

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Risk and hazard assessment for accidental chlorine release using dispersion modeling

Diganta Bhusan Das, Sandeep Kumar Tetarwal and **Raj Sharma**[1]

Department of Chemical Engineering, Malaviya Regional Engineering College, Jaipur-302017, Rajasthan, India; E-mail: rajsh@recjai.ernet.in

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Abstract—Dispersion modeling for risk and hazard assessment for disaster management planning has been a rapidly advancing area. In the present study, a heavy gas model was used to simulate the dispersion of negatively buoyant and highly toxic chlorine to illustrate the use of dispersion modeling for risk and hazard studies in Matsya Industrial Area in the Alwar district of Rajasthan, India, which handles about 400 tons of chlorine every day. The consequences of a possible accidental release of 2 to 100 tons of chlorine were examined. Conservative estimates (e.g. in case of low wind velocity and Pasquill stability class "A") of ground level concentrations (GLC) as a function of distance and time (e.g. concentration of 1.39×10^{-11} g/m^3 at 600 m and after a time interval of 300 sec in case of 2 tons release) were obtained in the present work to be used for risk assessment studies. Hazardous zones were also identified by calculating the safe downwind distances and critical time intervals based on different threshold concentrations of chlorine (e.g. 3.5 km and 54.5 minutes for 2 tons release when critical concentration is STEL (1 ppm)). Effects of topography were incorporated by simulating for different ratios of friction velocity to wind velocity ranging from 0.2 to 0.9, and it was observed that for increasing ratios the ground level concentrations decreased significantly. Polar isopleths based on actual average wind velocities in the area were generated to examine the simultaneous effect of wind direction along with velocity on air quality. The probabilities of percentage of deaths and injuries in a given population were calculated by probit analysis; while area of lethal dosage was determined in order to estimate percentages of population affected in the area based on population density.

It was demonstrated that dispersion modeling is an effective tool for risk assessment studies based on which disaster management plans can be evolved. This would help in either minimising the number of accidents or the losses to human health and life in the event of a catastrophic accident in the area.

Key-words: risk and hazard assessment, chlorine, accidental release, Matsya Industrial Area in Alwar (India), toxic load, concentration profile, safe distance, dose-effect relationship, polar isopleth, probit analysis, area of lethal dosage.

[1] Corresponding author

1. Introduction

Even though the chemical plants are perhaps the safest of all manufacturing facilities (*Crowl* and *Louvar*, 1990), public concern about safety is a result of the potential of this industry to cause significant damage to human health and life. The methyl isocyanate (MIC) disaster of Bhopal (India) in 1984 is a case in point where thousands lost their lives and tens of thousands were affected. Accidents do and continue to occur in spite of the best safety measures, however with disaster management planning the magnitude of the effects of such accidents on human life can be minimised. To develop a disaster management plan, it is first necessary to identify the hazardous zone in case of an accidental release of toxic chemicals. Dispersion modeling for identifying these hazardous zones has been a rapidly advancing area and is also applied for emergency planning, regulatory decision making and risk assessment studies for industrial operations. If properly applied, dispersion models should also improve the quality of design and layout studies (*Bennet,* 1982; *Schreurs* and *Mewis*, 1987).

Kolluru (1991) and *Auger* (1995) have discussed the different components of risk and hazard assessment and the formation of disaster management plan for chemical process industries. This work focuses on the use of dispersion modeling as a route to disaster management planning in case of an accidental release of chlorine. As a case study, an attempt has been made to estimate or quantify the hazards owing to an accidental release of chlorine in the Matsya Industrial Area (M.I.A.), Alwar (India), where large (about 400 tons) quantities of chlorine are handled every day. Chlorine is very toxic and, being heavier than air, disperses at the ground level making it extremely hazardous to human life and health. This work should serve as an aid for evolving a disaster management plan in the event of an accidental chlorine release in M.I.A.

The Matsya Industrial Area in Alwar district of Rajasthan, India was selected as the area of study. The district of Alwar is situated in the North-East of Rajasthan between the 27.4° and 28.4° North latitudes and the 76.7° and 77.13° East longitudes at a distance of 140 km in South-West direction of Delhi and of 160 km in North-East direction of Jaipur. Sariska National Park and Wild Life Sanctuary is about 45 km from the industrial area. M.I.A. has a very large chlorine plant (capacity of 90 tons per day (TPD)) and some 35 smaller units manufacturing chlorinated paraffin wax (CPW) with a chlorine consumption of 5 to 10 TPD. On an average about 400 TPD of chlorine is handled in this area. The ambient concentrations are 6.62 $\mu g/m^3$ for SO_2 and 7.87 $\mu g/m^3$ for NO_x (*RPCB*, 1995) indicating a low level of air pollution in the area, however no information on chlorine in ambient air is available. The four seasons in the district are Summer (March–June), Monsoon (July–September), post-Monsoon (September–November) and Winter (December–February). The sky is mostly cloudy in Monsoon, whereas thin cloud cover may be found sometimes in Summer and post-Monsoon seasons. The sky is generally clear (no clouds) in winter. The maximum temperature recorded so far is 48°C and the lowest value has touched the freezing point. The average temperature is 26°C, the average rainfall is 590 mm in the area. Maximum values of relative humidity occur during the Monsoon season (75%–90%), for the rest of the year the air is dry. For most of the year wind speed in Alwar is less than 2 m/s from North-West direction.

2. *Background information*

2.1 Available models

It was recognised as early as 1970 that attempting to describe dispersion of heavy gas by adopting the Gaussian model suitable for neutrally or positively buoyant clouds was inherently inadequate. Consequently many new models were proposed using the so-called "Top Hat" or "Slab" and "K-theory" or "Eddy diffusivity" approaches. "Top Hat" models assume that mass transfer occurs by entrainment across the density interface of a cloud with an assumed shape (e.g. cylindrical) and that internal mixing is fast enough for the concentration within the cloud to be uniform, whereas K-theory models numerically integrate suitably simplified equation of mass, momentum and energy conservation in two or three dimensional form (*Blackmore et al.*, 1982). One of the first "Top Hat" approaches to modeling was that of *van Ulden* (1974). Many "Slab" models were proposed soon after (*Germeles* and *Drake*, 1975; *Kaiser* and *Walker*, 1978; *Picknett*, 1981; *Fryer* and *Kaiser*, 1979 (DENZ); *Colenbrander*, 1980; *Cox and Carpenter*, 1980; *Eidsvik*, 1980; *Webber* and *Brighton*, 1984; *Witlox*, 1994a,b,c (HEGADAS-V)). Models that use the K-theory representation for heavy gas dispersion have also been discussed by many authors (e.g. *Schnatz* and *Flothmann*, 1980 (TRANSLOC); *Tauscher* (see: *Blackmore et al.*, 1982); *Su* and *Patnak* (see: *Blackmore et al.*, 1982)). *Blackmore et al.* (1982) reviewed the strengths and weaknesses, mechanistic features, the applicability to differing types and geometries of release, the ease of availability to users and the degree to which calculated results can be compared with field data. However very few work use Indian data. *Singh* (1990a) developed a heavy gas model for Indian meteorological conditions. The model has already been applied for oleum leakage (55 tons) at Sriram Fertilisers and Chemicals Ltd. in New Delhi, India, for Bhopal gas tragedy (MIC release of 40 tons) in 1984 and for the 2 tons chlorine spill at Chembur, Bombay, India, in 1985 with good results (*Singh*, 1990b). The model incorporates all the basic characteristics of heavy gas dispersion except that the local fluctuations of concentration are not predicted and dry deposition of pollutants as well as possible flashing of two-phase flow are not taken into account. The most attractive feature of this model is the ease of computational effort while providing quick and reasonable estimates of hazards which can then be used for emergency preparedness. This model was therefore adopted in the present study.

2.2 Atmospheric stability

The concept of atmospheric stability is very important for the evaluation of assimilative and supporting capacity of the atmosphere and is used extensively in dispersion modeling. Pasquill stability criteria (*Pasquill*, 1974), as adopted by the Bureau of Indian Standards, were used for identifying the atmospheric stability class prevailing in M.I.A. Surface wind speed, intensity of solar radiation and night time sky cover are the prime factors defining the atmospheric stability classes.

2.3 Meteorology

Transportation and dilution of pollutants in the atmosphere depend strongly on the prevailing meteorological conditions (wind characteristics and strength, atmospheric stability), the topography of the location, release mechanisms, the height at which the pollutants are released and various reactions occurring in the atmosphere. As suggested by *Jindal* and *Agarwal* (1995) stability class "A" (the Pasquill stability class "A" is an extremely unstable situation when the surface wind speed is less than 2 m/s and there is strong solar radiation from the clear sky) was adopted in the present study. Wind roses were not available for M.I.A., therefore wind roses prepared by the Rajasthan State Pollution Control Board (RPCB), Jaipur, for the district of Alwar were used to represent the wind distribution in the area. As evident from the wind rose (*Fig. 1*), most of the year the wind blows from the South-East direction. During winter, which gives the worst case of stability in Alwar (*Jindal* and *Agarwal*, 1995), the maximum and minimum wind velocities as calculated from the wind roses were 2.91 m/s due West and 1.21 m/s due South. *Jindal* and *Agarwal* (1995) however suggested a wind velocity of 1.0 m/s for predicting the "worst scenarios" in the area. It was argued that if the wind velocity is high, even though the pollutants would disperse more and they would cover bigger area, people would be exposed to the toxic cloud for a shorter duration; whereas in case of low wind velocity, the cloud would move slowly and cover less area, but the time of exposure would be high. Therefore if the toxicity is a direct function of exposure time, a particular dose on the same area would be lethal when the wind velocity is low. Hence a wind velocity of 1.0 m/s as a worst case scenario was used in the simulation. In the present study chlorine release was assumed to take place because of catastrophic failure of a pressurised tank.

The height of release, which is also important for describing the dispersion phenomenon, was taken to be at ground level.

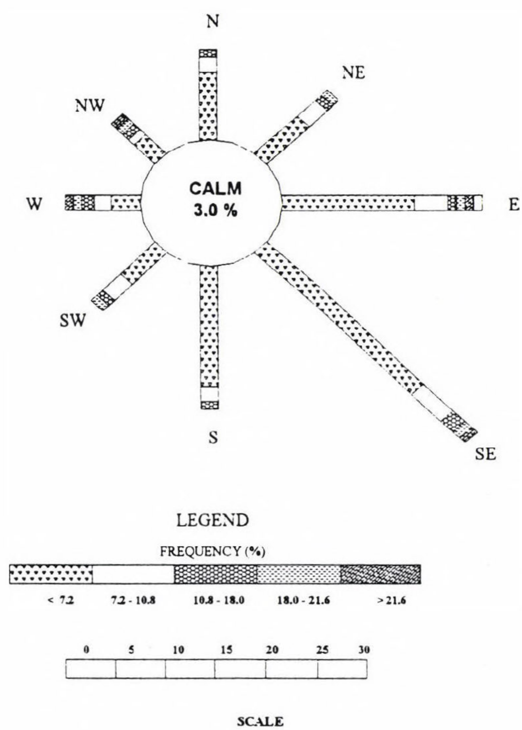

Fig. 1. Wind rose diagram of Alwar district in Winter (*RPCB,* 1995).

3. Basic model/equations used

3.1 Heavy gas model

Generally, the procedure for estimating the consequence of an accident has three components (*Mohan*, 1987):

(i) the mathematical description of the process which forms a cloud; this is necessary to provide the input parameters for the dispersion calculation for any particular event,

(ii) the mathematical description of the dispersion process which provides the concentration of a gas as a function of space and time, and

(iii) the description, in quantitative terms, of how a possible accident injures the population and damages the environment as a result of exposure to the concentration-time history derived from (ii).

In the present work the heavy gas model proposed by *Singh* (1990a) was adopted for estimating the consequences of a possible chlorine accident in M.I.A., Alwar. This model takes into account the first two components above by incorporating all the basic characteristics of heavy gas dispersion except that it does not predict the local mean fluctuations of concentration, ignores deposition and chemistry, and does not consider the possible two phase flashing flow of pollutants. The model was adopted because for all practical purposes it is possible to ignore variations in local concentrations and the presence of liquid gas in the toxic cloud. *Table 1* presents the model equations used in this study.

3.2 Simulator architecture

A computer simulator based on the equations in Table 1 was developed for predicting the concentration of chlorine as a function of distance and time under different meteorological conditions and of the released amounts. *Singh* (1990a) did not provide any value for the air entrainment co-efficient or the ratio of friction velocity to wind velocity. Therefore values suggested by *Eidsvik* (1980) and *Taylor* (1994) were used in the simulator. While simulating for risk estimation, the thermal effects of cloud heating due to temperature difference between the ground and air borne vapor were not considered because heating is important only when the cloud is considerably cold, e.g. in case of an accidental release of pressurised ammonia, therefore the equations describing the cloud heating have not been included in Table 1.

3.3 Probit analysis and area of lethal dosage

A large toxic release may give rise to the following effects on human life: (a) lethal injury (death), (b) non-lethal injury and (c) irritation depending on the breathing rate, functional capacity (e.g. oxygen uptake, muscle strength in case of paralysing agents), exposure to a given dose of the chemical and time of exposure or concentration-time profile (dosage), and most importantly individual susceptibility. In order to estimate the effects of the toxic release, it is therefore necessary to know the relationships between the concentration profile and the degree of injury. The degree of variation in dose-response can be presented in the form of a Gaussian response curve known in the terminology of toxicology and risk assessment as probit curve (*Taylor*, 1994). This dosage-response relation has been expressed by *Eisenberg* (see: *Crowl* and *Louvar*, 1990) as a probit equation. The probit equations for chlorine deaths and injuries are presented in *Table 2*.

Table 1. Model equations used in the simulation (*Singh*, 1990a)

Phenomena	Eq. No.	Parameter	Equation	Constant	Assumption	Remarks
Gravity slumping	1	Velocity at the edge of the cloud	$dr/dt = C\sqrt{\{(\rho_g - \rho_a)\, gh/\rho_g\}}$	C = 1.3* with entrainment and turbulence; C = 1.0** without entrainment and turbulence	The toxic cloud has cylindrical shape	C = 1.3 has been used in the present work
	2	Volume of puff	$V = \Pi r^2 h$		Radius and height are equal at time = 0 s	Volume and density are constant for no entrainment of air
Entrain-ment of air	3	Entrainment at the edges and top of the cloud	$dM_a/dt = \rho_a(\Pi r^2)U_e + 2\rho_a(\Pi rh)\alpha^* \times (dr/dt)$		Amount of air entrained initially is twenty times that of the mass of toxic material	
	4	Entrainment velocity	$U_e = \alpha' U_1 R_i^{-1}$	$\alpha' = 0.5$		U_e is proportional to the difference between the local wind velocity and the velocity at the top surface, which also indicates presence of mechanical turbulence
	5	Richardson Number	$R_i = (gl_s/U_1^2)\Delta\rho/\rho_g$			It shows that entrainment velocity is dependent on the Richardson Number
	6	Entrainment at the top surface	$dM_a/dt = \rho_a\,(\Pi r^2)\,U_e$			Derived from Eq. 3. Entrainment at the edges is ignored because it is not important during gravity slumping
Conc. within the puff	7	Density of mixture at constant temperature	$\rho = (M_a+M_g)/((M_a/\rho_a) + (M_g/\rho_g))$			
	8	Instantaneous concentration	$C(x,y,z,t) = M_g G(x,y,z,t)/(\sqrt{2})\Pi^{3/2}\sigma_y^2\sigma_z$		Toxic mass has a Gaussian distribution	(x,y,z,t) gives the position of the puff in Cartesian coordinate at time *t*
	9	Function G	$G(x,y,z,t) = \exp[-\{y^2 + (x - x(t))^2\}/2\sigma_y^2 - z^2/2\sigma_z^2]$			
	10	Standard deviation in lateral direction	$\sigma_y = r/2.14$			
Post transition period	11	Standard deviation in vertical direction	$\sigma_z = h/2.14$			
	12	Radius of the cloud				
	13	Height of the cloud	$r = r_T + 2U_{fr}(t - t_T)$	$\alpha'' = 0.4$	$U_{fr} / U = 0.25$	
	14	Position of the puff centre	$h = h_T + \alpha'' U_{fr}\,(t - t_T)$ $x(t) = \int_0^t U(t')dt$			When the model is in heavy gas dispersion mode, the cloud point is based on its front velocity. When ambient air turbulence takes over, the cloud point is given by the ambient air

* Ref.: *Eidsvik,* 1980 ** Ref.: *Singh,* 1990a

The area covered by lethal concentration may be used to determine the percentages of population affected in a given area. The area of lethal dosage is a function of wind speed and stability condition and is calculated by the equation given in Table 2.

Table 2. Equations for probit analysis and area of lethal dosage (*Crowl* and *Louvar*, 1990)

Parameter	Eq. No.	Equation	Remarks
Probit equation for deaths	15	$Y = 1.69 \ln \Sigma C^{2.75} t - 17.1$	The relationship applies only to healthy adults and susceptible individuals such as infants, old people & people with advanced pulmonary/cardiovascular disease.
Probit equation for non-lethal injuries	16	$Y = 2.9 \ln C - 2.40$	Non-lethal injury is taken to mean hospitalisation with or without lasting impairment of health.
Area of lethal dosage	17	$A = K' U^{-n'}$	$K' = 1.72$ for Pasquill stability categories A-C. $n' = 1.06$

4. *Results and discussion*

Simulated results are sensitive to variables affecting atmospheric dispersion according to certain physical laws. As discussed earlier, in the present work a worst case scenario study has been conducted for M.I.A., Alwar, India, with implications of an accidental release of 2 to 100 tons of chlorine during the winter season.

4.1 Toxic load

Toxic load is essentially the quotient of the quantity of the released pollutant and the immediately dangerous to life and health (IDLH) value, and is a quantitative assessment of the potential risk associated with the use of that toxic chemical (*Singh*, 1990b). *Fig. 2* shows the toxic loads of chlorine for different accidental releases. Increasing values of toxic load indicate that the potential risk associated with a larger amount of release is higher. It necessitates the plants to use the best safety measures to lower the associated risks. An effective way of decreasing the risk would be to reduce the storing capacity of the plants.

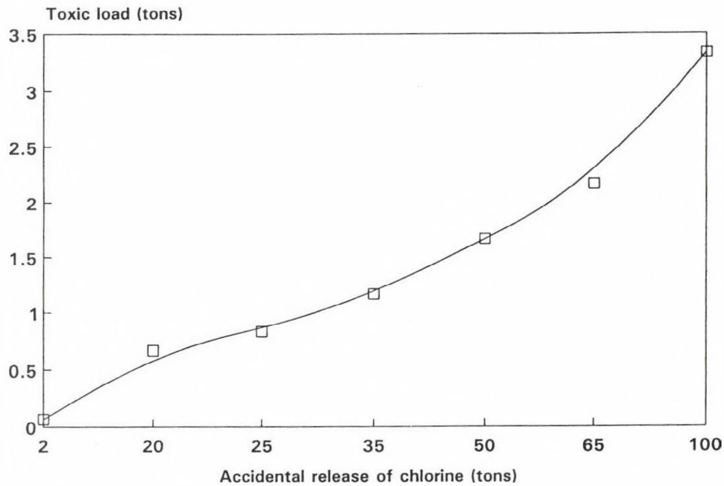

Fig. 2. Toxic load for different chlorine releases.

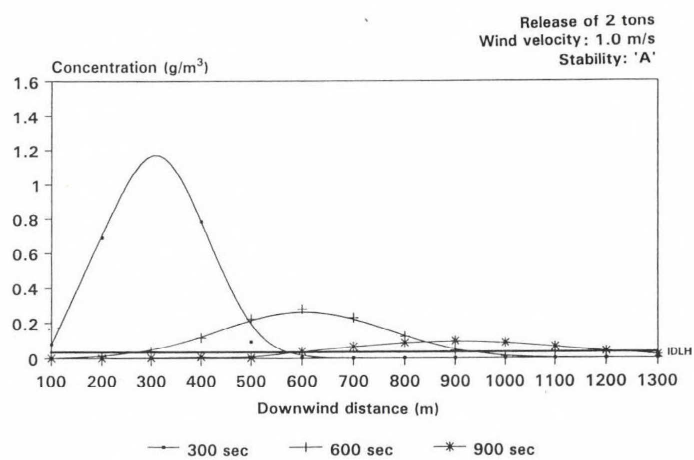

Fig. 3. Concentration profile at different time intervals.

4.2 Concentration profile

In general, at a given time the concentration of chlorine increases with the increase in downwind distance until reaching a maxima and then decreases. *Fig. 3* shows a typical concentration profile for a 2-ton release of chlorine at 300, 600 and 900 second time intervals as a function of downwind distance (100 m to 1300 m). The puff center is the point of the maximum concentration. Hence the peaks of the curves which are the points of highest concentration, represent the points near the puff center. At points away from the puff center concentration is lower as shown in Fig. 3. The points of maximum concentration are 300 m at 300 seconds, 600 m at 600 seconds and 800 m at 900 seconds. The concentration levels are also seen to be above the IDLH value between 100 m and 550 m at time interval of 300 seconds, between 300 m and 900 m for the 600 second interval and between 600 m and 1200 m for the 900 second interval. If the wind dispersed the toxic cloud towards North-West direction, Alwar city and the industrial township would be the most affected area, whilst the effect would be the least in case of a South-Easterly wind direction. Variation of concentration at a particular downwind distance with time is a matter of importance. *Table 3* presents a typical trend of the variation of concentration with time at distance of 900 m from the point of release. Concentration is found below the lethal dose (IDLH) at 300 seconds and 600 seconds and above the IDLH value at 900 seconds.

Table 3. Variation of concentration with time at a fixed distance (ref. Fig. 6)

	Time interval, sec		
	300	600	900
Downwind distance, m	900	900	900
Concentration, g/m^3	1.39×10^{-11}	4.29×10^{-2}	9.59×10^{-2}

4.3 Safe downwind distance

For the purpose of hazard assessment, the main objective of dispersion calculation here is to determine the potential for damage at various points in the vicinity of an accidental release. At some distance from the release the concentration experienced would be below that likely to constitute a significant hazard; but within that range the proper specification and identification of hazards are a matter of great importance. Therefore safe downwind distances for different threshold concentrations (see Appendix for definitions) and different amounts of chlorine release were calculated. The simulated results are presented in *Table 4* and a typical trend is shown in *Fig. 4*. These results would find use while identifying the hazardous zones in case of an accidental chlorine release. For example the safe IDLH distance for 50 tons release is 4.9 km. The effect is estimated to last for a time period of 57 minutes.

Table 4. Safe downwind distances based on different threshold concentration of chlorine

Based on STEL concentration (1 ppm)							
Amount released (tons)	2	20	25	35	50	75	100
Safe distance (km)	3.5	7.9	8.6	11.1	12.9	14.6	-
Time interval (minute)	54.5	118.3	127.5	142.8	161.0	184.7	-
Based on NJATC (14 ppm)							
Amount released (tons)	2	20	25	35	50	75	100
Safe distance (km)	1.6	3.8	4.2	4.8	5.5	6.6	7.5
Time interval (minute)	22.8	50	53.8	60.5	68.3	78.3	86.5
Based on ERPG-3 concentration (20 ppm)							
Amount released (tons)	2	20	25	35	50	75	100
Safe distance (km)	1.41	3.5	3.8	4.4	5.1	6.1	7.0
Time interval (minute)	20.3	44.5	48.0	53.8	60.8	70	77.2
Based on LC-50 (10 min exposure) concentration							
Amount released (tons)	2	20	25	35	50	75	100
Safe distance (km)	0.7	1.8	2.0	2.4	2.9	3.6	4.1
Time interval (minute)	7.7	17.3	18.3	21.2	24.2	27.8	30.8
Based on IDLH concentration (30 ppm)							
Amount released (tons)	2	20	25	35	50	75	100
Safe distance (km)	1.32	3.3	3.6	4.2	4.9	5.8	6.5
Time interval (minute)	18.8	41.3	43	50	56.5	65	71.8

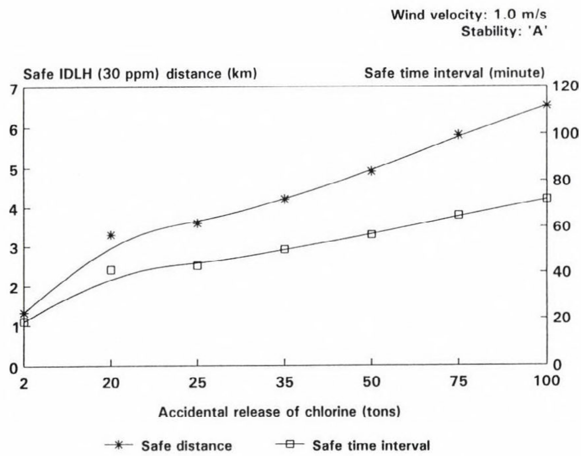

Fig. 4. Safe IDLH distance and time interval for different chlorine release.

4.4 Dose-effect relationships

To determine the chronic (long term) health effect of any specific chlorine dose on human beings in case of an accident so that immediate remedial action may be taken up, or to ascertain the extent of medical help that might be needed in the event of an emergency, it would be necessary to estimate the exposure of the population and to identify a safe range. *Table 5* presents the results simulated for a few "critical" concentrations. The estimated safe downwind distances and time intervals for different amounts of chlorine release when the concentration of interest is 3 ppm are presented in *Fig. 5*. A dose of 3 ppm for a 30-minute exposure time can be tolerated without any subjective feeling of malaise. If 2 tons of chlorine are released, a concentration of this intensity would be experienced up to a downwind distance of 2.5 km and for a period of 13.5 minutes; whereas, in case of 100 tons release, the critical distance and time interval would be 11 km and approximately 52 minutes respectively.

4.5 Effect of topography

Hills, buildings and other obstacles may cause additional turbulence. Typical effects of additional turbulence on concentration profiles for the releases of 20, 50, 65 and 100 tons of chlorine at a distance of 1.0 km and a time interval of 600 seconds are presented in *Fig. 6*. It is apparent that with the increase of the ratio of friction velocity (U_{fr}) to wind velocity (U), the concentration decreases significantly. This trend is observed because the additional turbulence produces a well-mixed plume. Since the concentration is a function of degree of mixing, a well-mixed plume gives lower concentration. Nevertheless except for 20 tons release at the U_{fr}/U ratio of 0.7 or greater, the pollutant levels would be high and dangerous.

4.6 Polar isopleths

Polar isopleths for examining the simultaneous effect of wind direction and velocity on air quality were prepared. Two types of polar isopleths are generally reported (*Bower* and *Sullivan*, 1981): wind isopleths and pollution isopleths, which are generalisations of wind and pollution roses, respectively. *Table 6* presents the average wind speeds for eight directions during the Winter season in Alwar district. *Fig. 7* shows the wind isopleth compiled from the wind data where average wind velocities are plotted against wind directions. Maximum and minimum wind speeds were found to be 2.91 m/s and 1.21 m/s in West and South directions, respectively. Pollution isopleths were prepared on the basis of actual wind velocities (Table 6) to give the dispersion of the pollutants. A pollution isopleth is presented in *Fig. 8*. The isopleth shows how chlorine would disperse with time to give a constant concentration of 90 ppm in case of 50 tons release.

Table 5. Dose-effect relationships for different chlorine release

Concentration: 3 ppm. Effect: can be tolerated without any feeling of malaise. Ex. time: 30 minute					
Amount released (ton)	2	20	35	50	100
Time interval (minute)	13.5	29.7	36	46.3	51.8
Downwind distance (km)	2.5	5.9	7.3	9.2	11
Concentration: 5 ppm. Effect: mild irritation to upper respiratory tract. Ex. time: 30 minute					
Amount released (ton)	2	20	35	50	100
Time interval (minute)	11.5	25.3	30.8	34.8	44.5
Downwind distance (km)	2.1	5.1	6.3	7.3	9.5
Concentration: 15 ppm. Effect: severe coughing, running of nose. Ex. time: 30 minute					
Amount released (ton)	2	20	35	50	100
Time interval (minute)	8	18.2	22.2	25	32
Downwind distance (km)	1.6	3.6	4.7	5.6	7.6
Concentration: 30 ppm. Effect: nausea, vomiting, over pressure feeling, shortness of breath, fits of coughing. Ex. time 30 minute					
Amount released (ton)	2	20	35	50	100
Time interval (minute)	6.5	14.8	18	20.5	26.3
Downwind distance (km)	1.3	3.53	4.1	4.7	6.4
Concentration: 40 ppm. Effect: toxic tracheobronchitis. Ex. time: 30 minute					
Amount released (ton)	2	20	35	50	100
Time interval (minute)	6	13.5	16.7	18.8	24.3
Downwind distance (km)	1.2	3.0	3.75	4.5	6.1

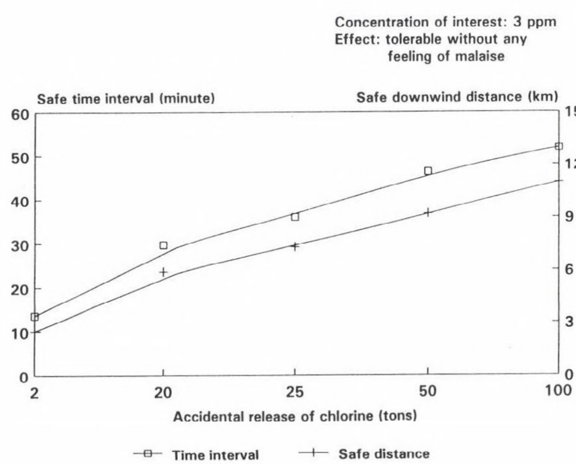

Fig. 5. Safe distance and time interval for different chlorine releases.

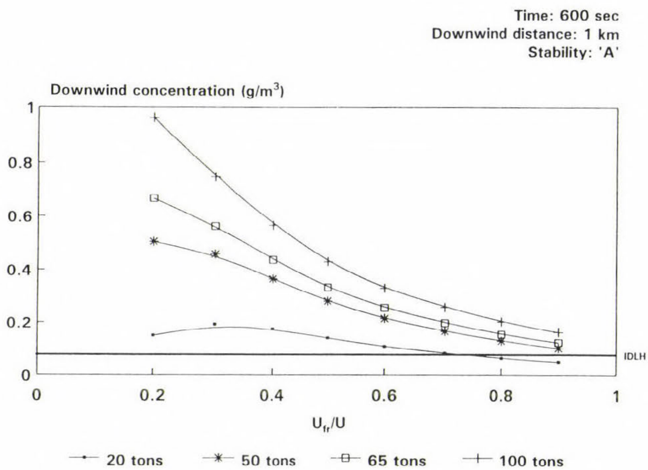

Fig. 6. Effect of topography.

Table 6. Wind directions and average velocities in winter at Alwar district of Rajasthan

Direction	North	N-East	East	S-East	South	S-West	N-West	West
Velocity, m/s	1.46	1.91	1.82	2.02	1.21	2.08	2.75	2.91

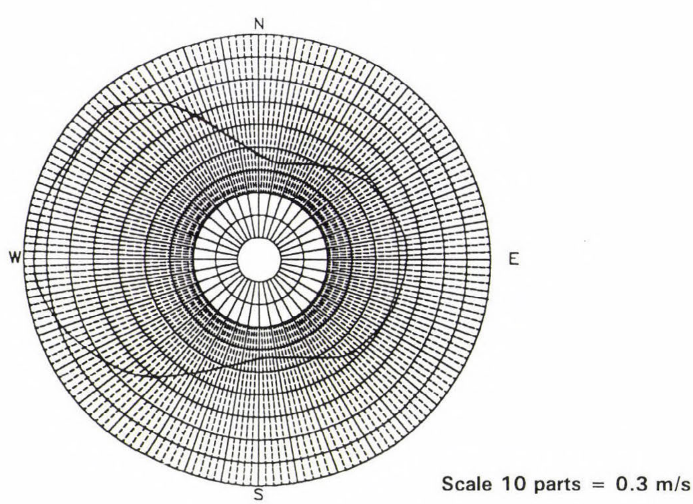

Fig. 7. Wind isopleth for Alwar district, India.

Variation of concentration around the source at a particular time interval of release is also a matter of importance. In order to illustrate this variation *Fig. 9* presents different concentrations of chlorine at 600 seconds for 50 tons release.

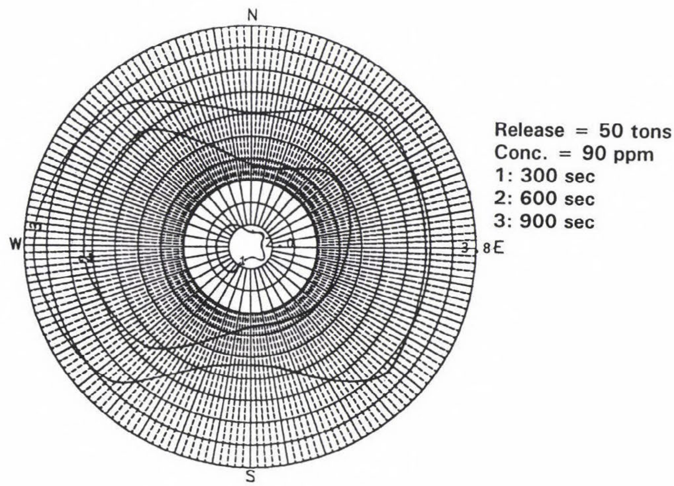

Fig. 8. Pollution isopleth for a concentration of 90 ppm at varying time interval in case of 50 tons release.

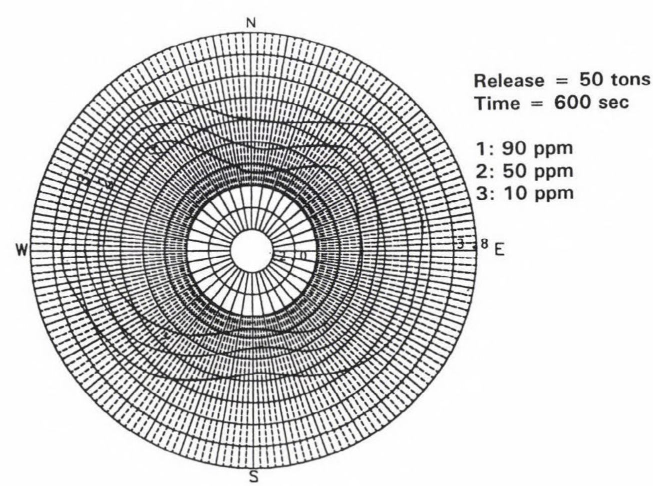

Fig. 9. Pollution isopleth for varying concentration at 600 seconds in case of 50 tons release.

4.7 Probit analysis

Probit analysis for lethality of chlorine has been applied to healthy adults including susceptible individuals such as infants, old people and people with advanced pulmonary/cardiovascular disease. The response of the subjects to a given dose is assumed to vary in a Gaussian manner (*Taylor*, 1994). These variations are presented by two cumulative distribution plots (probit curves), which indicate the probabilities of percentage of deaths (*Fig. 10*) or injuries (*Fig. 11*) of a given population for a specific dose or exposure time. Fig. 10 shows that the time of exposure causing 100% deaths for given concentration decreases with increasing concentrations and it is in the range of 4.3 hours for 30 ppm to 3.2 minutes for 150 ppm; similarly for given time intervals the concentrations for 100% death ranged from 85.0 ppm for 15 minutes to 51.0 ppm for 60 minutes. For non lethal injuries with or without lasting impairment of health, a concentration of 37.5 ppm will affect 100% of the population (*Fig. 11*).

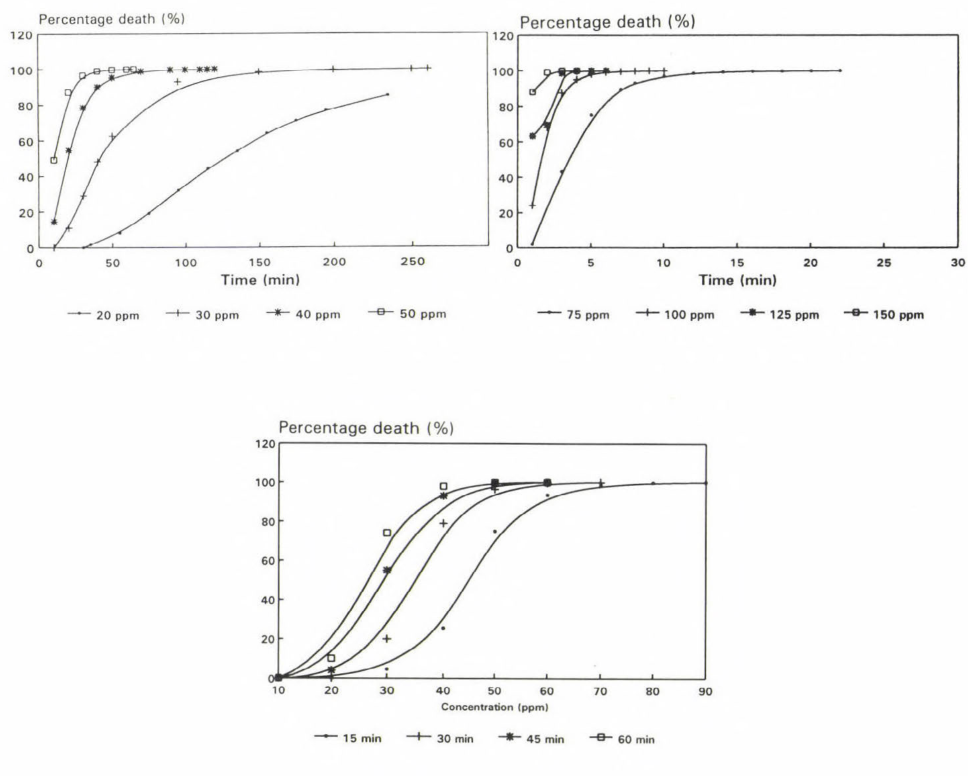

Fig. 10. Probit analysis for chlorine death.

4.8 Area of lethal dosage

Simulated results showed that the area of lethal dosage decreased with the increasing wind velocity (*Fig. 12*). This may be attributed to the fact that as the atmospheric conditions become more turbulent the chlorine disperses over greater area, hence the area of lethal dosage is smaller. For the wind velocity of 1.0 m/s used in the simulator the area of lethal dosage is 1.72 m^2. No information was available on the population density either in the vicinity of M.I.A. or in the industrial township. However, for a population density of 100 persons km^{-2}, the predicted number of people that would be most affected in case of an accident in the area is 172.

5. Summary

The paper has illustrated how a heavy gas model can be utilised for risk assessment in cases where combined effects of gravity slumping and air entrainment should be taken into consideration. The simple analytical form of the model allowed quick estimates of the consequences when the magnitudes of any or all variables like wind velocity, stability, source strength, downwind distance vary. Since the dispersion models assume the current and the future weather and release conditions to be representatives of past measurements, even though there can be a wide range of cases giving many probable outcomes, the worst meteorological conditions (e.g. stability class "A") were considered.

The source term conditions studied for predicting downwind concentrations and other measurements include instantaneous release of chlorine in the range of 2 to 100 tons. Concentration profiles as functions of time and downwind distance were obtained which can be used for various risk assessment studies. One of the estimates that can be obtained from this study is the safe downwind distance (e.g. the safe IDLH distance of 1.32 km in case of 2-ton release). The safe downwind distances can be used for identification of hazardous zone. Dose-effect relationships based on the threshold concentrations of chlorine, developed by various organisations, were established for proper specification of the extent of hazards involved. Topography or surface roughness which has a significant effect on the dispersion process, was considered in the present study for generating concentration profiles. Concentration was found to decrease with the increasing surface roughness at a particular distance. The polar isopleths, which combine the simultaneous effects of direction and velocity, would be very useful to ascertain the exact spreading of the pollutants.

The work presented here lays the foundation of a disaster management plan and demonstrates the use of dispersion modeling as an important tool for risk assessment studies. A future exercise would evolve a disaster management plan based on this work, which would demonstrate the utility of such studies in preventing or minimising accident related damage. This would be useful either in preventing accidents in the M.I.A., Alwar, or for minimising the number of deaths in case of an accidental release of chlorine to the atmosphere.

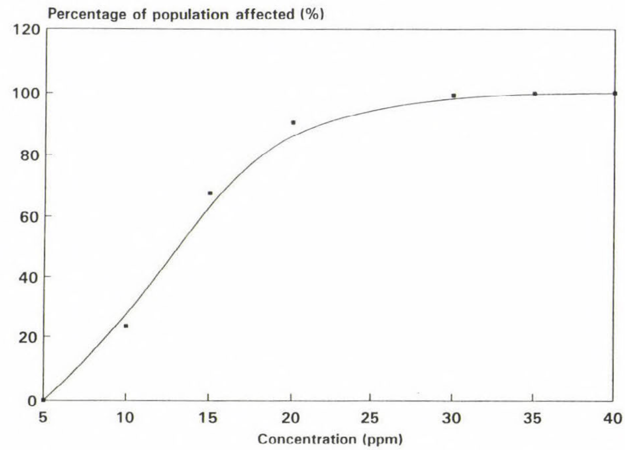

Fig. 11. Probit analysis for chlorine injury.

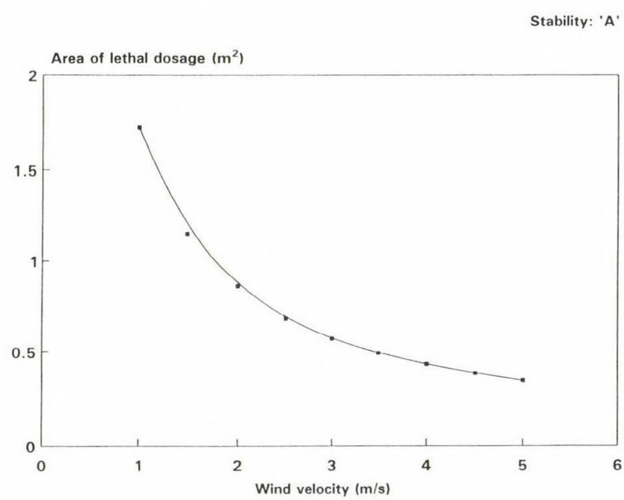

Fig. 12. Area of lethal dosage.

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APPENDIX

Definition of critical concentration of chlorine (*Mallet*, 1993):

(a) STEL (1 ppm):	The occupational short term exposure limit. It is a 15-minute time weighted average concentration that should not be exceeded at any time during the work day.
(b) IDLH (30 ppm):	The Immediately Dangerous to Life and Health concentration. It represents the maximum concentration from which, in the event of respiratory failure, one could escape in 30 minutes without a respirator and without experiencing any escape impairing (e.g. severe eye irritation) or irreversible health effect.
(c) NJATC (14 ppm):	New Jersey Acute Toxicity Concentration — established by the state of New Jersey and used for developing its risk management program regulations.
(d) LC-50:	The concentration believed to be lethal to 50% of humans exposed for the stated period of exposure time.
(e) ERPG-3 (20 ppm):	Emergency Response Planning Guidelines-3. It is the maximum air borne concentration below which it is believed that nearly all individuals would be exposed for up to one hour without experiencing or developing any life threatening effect.

List of symbols

Symbol	*Definition*	*Used in*
A	Area of lethal dosage, m^2	Eq. 17
C	Downwind concentration, g/m^3	Eq. 8
c	Gravity slumping constant, dimensionless	Eq. 1, 2
dr/dt	Rate of change of cloud radius or, cloud front velocity	Eq. 1, 3
g	Acceleration due to gravity, m/s^2	Eq. 1
G	A function	Eq. 8, 9
h	Cloud height, m	Eq. 1, 3, 11
h_T	Height of the cloud at transition time, m	Eq. 13
K′	Constant	Eq. 17
l_s	Turbulence length scale, m	Eq. 5
M_g	Mass of toxic chemical released, ton	Eq. 7
M_a	Mass of air entrained, ton	Eq. 7
n′	Turbulence index, dimensionless	Eq. 17
r	Cloud radius, m	Eq. 2, 3, 6, 10
r_T	Radius of the cloud at the transition time, m	Eq. 12
R_i	Richardson Number	Eq. 4, 5
t	Time, sec	Eq. 12, 13
U	Wind velocity, m/s	Eq. 14
U_e	Entrainment velocity, m/s	Eq. 2, 4, 6
U_{fr}	Friction velocity, m/s	Eq. 12, 13
U_l	Turbulence velocity, m/s	Eq. 4
V	Volume of the cloud, m^3	Eq. 2
x	Downwind direction, m	Eq. 9
x(t)	Puff centre, m	Eq. 9, 14
y	Lateral direction, m	Eq. 9
Y	Probit	Eq. 15, 16
z	Vertical direction, m	Eq. 9
(x,y,z,t)	Position of the puff centre in Cartesian coordinates	Eq. 7

Greek Letters

α'	Co-efficient for top entrainment, dimensionless	Eq. 4
α''	Co-efficient for vertical entrainment, dimensionless	Eq. 13
α^*	Co-efficient for entrainment at the edges, dimensionless	Eq. 3
ρ_a	Density of air, kg/m^3	Eq. 1, 3, 6
ρ_g	Density of toxic cloud, kg/m^3	Eq. 1, 5
σ	Deviations, m	Eq. 8

Subscript

a	Air	Eq. 1, 3, 6, 7
g	Toxic gas	Eq. 1, 5, 7
T	Transition time, sec	Eq. 12, 13
y	Lateral direction, m	Eq. 7, 9
z	Vertical direction, m	Eq. 8, 9

BOOK REVIEWS

J.H. Seinfeld and *S.N. Pandis*: **Atmospheric Chemistry and Physics — From Air Pollution to Climate Change**. John Wiley and Sons, Inc., New York, Chichester, Weinheim, Brisbane, Singapore, Toronto, 1998. XXVII + 1326 pages, tremendous amount of figures, tables, references, appendices and problems to be solved.

This volume is the thickest book on the atmosphere, the reviewer has ever seen. It deals mostly with the chemistry of the atmosphere, with separate chapters on aerosol and cloud physics and air pollution meteorology. As the authors state in the preface: "The object of this book is to provide a rigorous, comprehensive treatment of the chemistry of the atmosphere, including the formation, growth, dynamics and properties of aerosols, the meteorology of air pollution, the transport, diffusion and removal of species in the atmosphere, the formation and chemistry of clouds, the interaction of atmospheric chemistry and climate, the radiative and climatic effects of gases and particles and the formulation of mathematical chemical/transport models of the atmosphere". This ambitious aim is obtained in 24 chapters. After reviewing the book one can say that, except some cases, the individual chapters are not too long and sophisticated. The length of the volume is due to the great number of chapters discussing the large subject mentioned.

If we took only Chapters 1–7, 13, and 19–24 we would receive a book on atmospheric chemistry as this branch of atmospheric science is generally imagined. The first chapter is an introduction into atmospheric characteristics, the second deals with atmospheric composition, cycles and lifetimes, the third one discusses the bases of photochemistry and reaction kinetics, while the aim of Chapters 4–7 is to give a summary of our present knowledge of stratospheric chemistry, tropospheric chemistry, atmospheric aqueous phase chemistry and aerosol properties, respectively. It is interesting that aerosols consisting of organic materials are not treated together with other particles, their properties are presented in a separate chapter (Chapter 13). It seems that the authors considered the organic particles so important that they devoted a separate chapter to their discussion (this is understandable since they are from California where organic substances were widely studied owing to their importance in photochemical smog). However, this discussion is rather separated from that of other aerosol particles. In Chapters 19–20 dry and wet depositions are presented, while in Chapters 21 and 22 our ideas on the interaction of atmospheric chemistry and climate as well as the radiative effects of atmospheric aerosols are summarized. Finally the authors' goal in the last two chapters is to describe atmospheric chemical and statistical models. The last chapter on statistical models is of particular interest since this subject is rarely occurs in atmospheric chemistry books prepared by European writers.

Chapters 8–12 are rather unique in books discussing the atmospheric physics and chemistry. Chapter 8 are devoted to the dynamics of single aerosol particles. This means that transport processes of single particles, their interaction with the fluid (drag force) and fluid molecules (Brownian motion) as well as their motion caused by external forces (e.g. gravitational settling) are presented. The next chapter ("Thermodynamics of Aerosols") contains the principles of partitioning of atmospheric species between the vapor and particulate phases, while Chapter 10 is a rather detailed discussion of nucleation processes. Then, the mass transfer rates between condensed and gas phases are discussed (Chapter 11). This part of the book on "aerosol physics" is closed by a chapter on the dynamics of aerosol populations including such important subjects as changes in particle size distribution by condensation and evaporation and by coagulation.

Except Chapter 15 on cloud physics, the remaining chapters are composed of material what we generally call air pollution meteorology. Thus, the title of Chapter 14 is "Meteorology of Air Pollution", while Chapter 16 contains the basic principles of micrometeorology. Further, in Chapter 17 and 18 the authors present the bases of atmospheric diffusion and analytical solutions of atmospheric diffusion equations, respectively. In this last chapter they describe, among other things, the Gaussian plume equation which is discussed in a detailed way.

It goes without saying that one can raise questions concerning the structure of this book. It is not questionable, however, that the authors present an excellent material, in agreement with the subtitle of the volume, from air pollution to climate change. This means that, except atmospheric radioactivity, the reader finds practically everything in this book which is important to understand atmospheric environment of our planet. Each chapter is followed by references consisting of a lot of literature published mostly by American research workers. Further, the chapters are closed by problems of different levels "to enable the reader to evaluate his or her understanding of the material" (cited from the authors' preface). In the preface the authors also state that "The book is intended to serve as textbook for a course in atmospheric science that might vary in length from one quarter or semester to a full academic year". However, one fills that the material composing this book seems to be too large even for a course of one year. On the other hand, it is self-evident that parts of this volume can be use in different courses on environmental engineering and science as well as on meteorology and chemistry.

Thus, the reviewer concludes that the present book is a magnificent successor of the previous book of one of the authors (J.H. Seinfeld) entitled *Atmospheric Chemistry and Physics of Air Pollution* published by the same editor in 1986. The former book is completed successfully by material emerged during the last decade. Consequently, the volume can be proposed to university professors and graduate students for their courses as well as to all persons educated in physics and chemistry for making an acquaintance with basic principles of atmospheric science.

E. Mészáros

Wilfried Schröder: **Noctilucent Clouds (Theoretical concepts and observational implications)**. Science Edition, D-28777 Bremen, 1998. 340 pages. Price: $ 20.

This book is a compilation of the works written by the author during the last decades. Since some of them have been published in German, it seems necessary to collect them into an English book, to be available for the readers who look only for English literature.

The articles can be grouped into three main topics:

— Climatology of noctilucent clouds (NLC);

— Mesosphere and NLC;

— History of observations made in Germany.

Noctilucent clouds have been observed since the eighties of the last century. They could be seen and photographed when the Sun is below the horizon by 5–15 degrees. Their average altitude is 83 km, that is they give information about such heights that could only be investigated by meteorological rockets. Their observation zone is the belt between 50 and 70 degrees latitude on both Hemispheres. The NLC are summer phenomena.

Their origin is not understood fully. Also, their relation to the wind speed and temperature is not known perfectly.

A small and enthusiastic group of scientists runs the observations and uses them to study the physics of the mesosphere. Those, who are interested in this topic, please, read this interesting book.

G. Major

NEWS

Congratulations to Professor Ernő Mészáros for the Széchenyi-prize

In March of 1998 the President of the Hungarian Republic awarded Széchenyi-prize (the highest prize for scientist in Hungary) to Professor E. Mészáros, former editor of Időjárás, in recognition of his excellent work in developing of the atmospheric chemistry as new area of atmospheric sciences in Hungary.

Ernő Mészáros began its scientific carrier in 1957 at the Hungarian Meteorological Service. That time his interest was in the cloud physics. Later on he investigated the physical and chemical properties of atmospheric aerosol particles. The prize recognized his activity in developing of the atmospheric chemistry as new area of atmospheric sciences in Hungary.

He has published several papers in high-impact scientific journals. He has always taken part in international scientific activities.

On the occasion of his 60th birthday, the Időjárás published his curriculum vitae (Vol. 100, No. 1-3, 1996).

G. Major

Symposium of the International Society for Photogrammetry and Remote Sensing (ISPRS) in Budapest

The Commission VII of ISPRS held its 1998 year Symposium at the Hungarian Academy of Sciences, 1-4 of September. The basic area of the Commission is resource and environmental monitoring, therefore the meteorologists are involved in the activity. The scientists of the Hungarian Meteorological Service presented a poster and computer demonstration of the activity of the Satellite Research Laboratory.

G. Major

ATMOSPHERIC ENVIRONMENT

an international journal

To promote the distribution of Atmospheric Environment *Időjárás* publishes regularly the contents of this important journal. For further information the interested reader is asked to contact *Prof. P. Brimblecombe*, School for Environmental Sciences, University of East Anglia, Norwich NR4 7TJ, U.K.; E-mail: atmos_env@uea.ac.uk

Volume 32 Number 7 1998

The Transport of Air Pollutants Over Complex Terrain (TRACT) September, 1992

Volume 32 Number 8 1998

Atmospheric Sciences and Applications to Air Quality (ASAAQ)

New Directions

Volume 32 Number 9 1998

Volume 32 Number 10 1998

Volume 32 Number 11 1998

Volume 32 Number 12 1998

The Mediterranean Campaign of Photochemical Tracers — Transport and Chemical Evolution (MEDCAPHOT-TRACE), Athens, Greece 1994–1995

NOTES TO CONTRIBUTORS OF *IDŐJÁRÁS*

The purpose of the journal is to publish papers in any field of meteorology and atmosphere related scientific areas. These may be

- reports on new results of scientific investigations,
- critical review articles summarizing current state of art of a certain topic,
- shorter contributions dealing with a particular question.

Each issue contains "News" and "Book review" sections.

Authors may be of any nationality, but the official language of the journal is English. Papers will be reviewed by unidentified referees.

Manuscripts should be sent to
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in three copies including all illustrations. One set of illustrations has to be of camera ready quality, the other two might be lower quality.

Title part of the paper should contain the concise title, the name(s) of the author(s), the affiliation(s) including postal and E-mail address(es). In case of multiple authors, the cover letter should indicate the corresponding author.

Abstract should follow the title, it contains the purpose, the data and methods as well as the basic conclusion.

Key-words are necessary to help to classify the topic.

The text has to be typed in double spacing with wide margins. Word-processor printing is preferred. The use of SI units are expected. The negative exponent is preferred to solidus. Figures and tables should be consecutively numbered and referred to in the text.

Mathematical formulas are expected to be as simple as possible and numbered in parentheses at the right margin. Non-Latin letters and hand-written symbols should be indicated and explained by making marginal notes in pencil.

Tables should be marked by Arabic numbers and printed in separate sheets together with their captions. Avoid too lengthy or complicated tables.

Figures should be drawn or printed in black and white, without legends, on separate sheets. The legends of figures should be printed as separate list. Good quality laser printings are preferred as master copies.

References: The text citation should contain the name(s) of the author(s) in Italic letter and the year of publication. In case of one author: *Miller* (1989), or if the name of the author cannot be fitted into the text: *(Miller,* 1989); in the case of two authors: *Gamov* and *Cleveland* (1973); if there are more than two authors: *Smith et al.* (1990). When referring to several papers published in the same year by the same author, the year of publication should be followed by letters a,b etc. At the end of the paper the list of references should be arranged alphabetically. For an article: the name(s) of author(s) in Italics, year, title of article, name of journal, volume number (the latter two in Italics) and pages. E.g. *Nathan, K.K.,* 1986: A note on the relationship between photosynthetically active radiation and cloud amount. *Időjárás 90,* 10-13. For a book: the name(s) of author(s), year, title of the book (all in Italics except the year), publisher and place of publication. E.g. *Junge, C. E.,* 1963: *Air Chemistry and Radioactivity.* Academic Press, New York and London.

The final version should be submitted on diskette altogether with one hard copy. Use standard 3.5" or 5.25" DOS formatted diskettes. The preferred word-processors are WordPerfect 5.1 and MS Word 6.0.

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More information: gmajor@met.hu
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