Working Paper

On the Effect of Applying Mean Input Data in Long-Term Air Pollution Transport Models

Ralph Lehmann

WP-90-026
June 1990

IIASA

International Institute for Applied Systems Analysis □ A-2361 Laxenburg □ Austria

Telephone: (0 22 36) 715 21 * 0 □ Telex: 079 137 iiasa a □ Telefax: (0 22 36) 71313

On the Effect of Applying Mean Input Data in Long-Term Air Pollution Transport Models

Ralph Lehmann

WP-90-026
June 1990

International Institute for Applied Systems Analysis □ A-2361 Laxenburg □ Austria

Telephone: (0 22 36) 715 21 * 0 □ Telex: 079 137 iiasa a □ Telefax: (0 22 36) 71313

Foreword

The deposition of heavy metals and the resulting risk of toxification of the environment is a problem which has only recently received attention. For the past two years, the Transboundary Air Pollution Project has been developing models for the long range atmospheric transport of heavy metals. The models involve the use of meteorological input and parameters along trajectories. This paper addresses the important question as to whether it is justifiable to use temporally and spatially averaged input data in the application of these models in producing long term average concentrations and deposition, and what kind of errors result from the use of mean values. This type of analysis is a necessary step in the development of heavy metals models.

Bo R. Döös
Leader, Environment Program

Roderick W. Shaw
Leader, Transboundary Air Pollution Project

Acknowledgements

This paper originated during a three-month stay of the author at the International Institute for Applied Systems Analysis (IIASA), Laxenburg (Austria), in 1990. I wish to thank Joseph Alcamo for suggesting the area of investigation and Roderick Shaw for reviewing the results.

Abstract

The effect of applying mean (i.e. temporally and spatially averaged) input data (removal coefficients, wind velocity, travel distance between source and receptor) in a long-term model of the transport of heavy metals (especially *As*) in Europe's atmosphere has been investigated by running modified versions of this model. At 5 receptors considered in this study, the application of mean removal coefficients in the model resulted in an underprediction of concentration values and dry deposition, being approximately 0.75 times the original values, and an overestimation of wet deposition by a factor of approximately 1.5. The assumption of a straight travel of pollutants between a source and some receptor brings about an overestimation of concentration and deposition by a factor of approximately 1.2. The application of a mean wind velocity changed results only slightly. All occurring effects have also been investigated theoretically, so that assertions and formulae which are independent of the special model under consideration have been obtained.

Contents

On the Effect of Applying Mean Input Data in Long-Term Air Pollution Transport Models

Ralph Lehmann

1 Introduction

In order to simulate the transport of air pollutants, a large variety of models has been developed in recent years. They cover a wide range of spatial and temporal scales (from the local scale of hundreds of metres to the global scale; from hourly values to long-term, e.g. yearly, averages) and represent very different scales of model complexity (from highly simplified linear models to nonlinear models with a rather detailed description of physical and chemical processes). In particular, if a first estimate for the long-term spatial distribution of certain air pollutants is required, simple models, often called "climatologic models", are applied (e.g. Klug and Erbshäußer, 1988, for SO_2; Derwent, 1987, for NO_x; Alcamo et al., 1990, for heavy metals). They need only a limited meteorological input. Usual assumptions involved in such models are:

1. The transport from some source to a certain receptor occurs on a straight trajectory by a mean value of the wind speed. The frequency of the occurrence of the corresponding wind direction is obtained from so-called 'wind roses'.

2. The pollutant is perfectly vertically mixed within the planetary boundary layer, for which a mean mixing height is assumed. Horizontal diffusion is neglected.

3. The chemical conversion and physical removal processes are assumed to be linear, i.e. the conversion or removal (per time unit) is proportional to the actual concentration, so that it can be described by linear differential equations.

4. Mean (temporally and spatially averaged) coefficients are applied for chemical reactions and for dry and wet deposition (i.e. "constant drizzle").

As a consequence of 3) and 4), the pollutant concentration along the trajectory can be described by exponential functions. If only decay and removal processes occur, one exponential expression per pollutant is sufficient; an example which includes also the production of a secondary pollutant can be found, e.g., in Derwent (1987).

Generally, it is believed that the application of mean input data (wind speed, removal coefficients etc.) is justified, at least to a certain extent, because effects produced by variations of these data "average out" in the long term. In the present paper we investigate how efficiently this "averaging out" works and which systematic errors can occur. Some theoretical results (Section 4) are accompanied by a "real model" example (Section 5): A trajectory model of the transport of arsenic in Europe's atmosphere (Alcamo et al., 1990) is applied to assess the effects of the simplifications involved in the development of a simple climatological model, especially the effect of averaged removal coefficients. The corresponding model is introduced in Section 2; a discussion of the basic effects resulting from averaged input data is contained in Section 3.

2 Model Description

Let us first consider the vertically and cross-wind integrated pollutant concentration (in $g\ m^{-1}$) along a trajectory that starts at a point source of emission intensity Q $[g\ s^{-1}]$ (at a time $t = 0$). Suppose that the pollutant under consideration undergoes linear removal processes, described by a removal coefficient $K(t)$, which may vary with the travel time t along the trajectory. Then its concentration along the trajectory (i.e. in a Lagrangian sense) is given by

$$c(t) = \frac{Q}{u(t)} e^{-\mathcal{K}(t)}, \tag{1}$$

$$\text{where } \mathcal{K}(t) = \int_0^t K(\tau) d\tau, \tag{2}$$

$u(t)$ = wind speed on the trajectory at travel time t.

This formula reflects the exponential decay of the concentration as well as the dilution of the pollutant parallel to the trajectory, which is indirectly proportional to the actual wind speed; for a derivation see Appendix A.

If $u(t)$ is constant ($= u$), we can simplify (1) by setting $c_0 := \frac{Q}{u}$:

$$c(t) = c_0 \cdot e^{-\mathcal{K}(t)}, \tag{3}$$

Usually, K includes the removal by dry and wet deposition and chemical transformations:

$$K = K_{dry} + K_{wet} + K_{chem},$$

$$\text{where } K_{dry} = \frac{v_d}{h_{mix}},$$

$$K_{wet} = \frac{W \cdot P}{h_{mix}},$$

v_d = dry deposition velocity,

h_{mix} = mixing height,

W = scavenging ratio (ratio of the pollutant concentration in precipitation versus its concentration in air),

P = precipitation intensity (in $mm\ s^{-1}$).

In the present paper we omit the consideration of chemical transformations since arsenic in the atmosphere is assumed to be chemically stable.

Dry and wet deposition are obtained from

$$d_{dry} = K_{dry}(t) \cdot c(t),$$
$$d_{wet} = K_{wet}(t) \cdot c(t).$$

The trajectory model under consideration (Alcamo et al., 1990) is based on equations of the form (1). (The factor $\frac{1}{u(t)}$ is not explicitly included in the trajectory model; but a corresponding factor appears "automatically", because the time that an air parcel stays over a certain emitter region [and during which it can take up pollutants] is indirectly proportional to the velocity $u(0)$ of that air parcel. A subsequent "compressing" and "stretching" of an air parcel due to changes in the wind speed, which is reflected in the appearance of $\frac{1}{u(t)}$ in (1) instead of $\frac{1}{u(0)}$, is not included in the model.)

The model is a version of the EMEP-MSC/W model (Eliassen and Saltbones, 1983; Eliassen et al., 1988) adapted to the transport of heavy metals (As, Pb, Cd, Zn). Here we consider only the As version. For a receptor of interest, $96-h$ back trajectories arriving every 6 h (i.e. 4 per day) are determined (using the 925 hPa wind speed and time steps of 2 h). In order to obtain long-term concentration and deposition values, the results of several trajectory runs are used to compute mean concentrations and depositions, e.g. all trajectories arriving at a receptor within one year are evaluated in order to calculate the yearly mean concentration and deposition at that receptor.

3 Basic Effects

3.1 Concentration

It can be seen from the short model descriptions in Sections 1 and 2 that one of the main differences between a climatologic and a trajectory model consists in the averaging operation that is applied to obtain mean climatologic concentration and deposition values: In the trajectory model, *first* computations with the "real" meteorological input are carried out, and *then* the results of several runs are averaged. On the other hand, in the climatologic model, *first* the meteorological input data are averaged, and *then* a model run is performed (cf. Fig. 1). Clearly,

the second variant requires a smaller amount of computational time and less detailed input data. We are going to investigate, which "price" in terms of accuracy of the results we have to pay for this. To be more specific: Will the climatologic model yield results with a systematic error ("bias"), or can errors be expected to "average out"?

As a first illustration, let us consider a pollutant source and a receptor which are separated by a fixed travel time $\bar{t}$. We are going to investigate the effect of the application of an averaged removal coefficient K, which, for simplicity, is assumed to be constant along each single trajectory and may assume only two values: K_1 for half of all trajectories and K_2 for the remaining trajectories. (Emission Q and wind speed u are assumed to be constant.) Then the trajectory and climatologic model would yield the following results (cf. (3)):

$$c_{traj} = c_0 \cdot \frac{1}{2}\left(e^{-K_1\bar{t}} + e^{-K_2\bar{t}}\right), \tag{4}$$

$$c_{clim} = c_0 \cdot e^{-\frac{1}{2}(K_1+K_2)\bar{t}}. \tag{5}$$

It follows directly from the convexity of the exponential function that

$$c_{clim} < c_{traj}$$

(cf. Fig. 2). That means:

> *The application of an averaged removal coefficient in the climatologic model always results in an underestimation of the concentration. Thus, errors in the computation of the concentrations of pollutants coming from different sources to a certain receptor cannot "average out".*

A slightly different interpretation of (4) and (5) is the following: After rewriting (5) in the form

$$c_{clim} = \sqrt{c_0 \cdot e^{-K_1\bar{t}} \cdot c_0 \cdot e^{-K_2\bar{t}}},$$

we easily observe that the trajectory model computes the *arithmetic mean* of the concentrations (corresponding to different trajectories), whereas the climatologic model yields the *geometric mean* of these concentrations, which is known to be smaller than the arithmetic mean. This effect will therefore be important at receptors where concentrations have a significant temporal variability, and less important where this variability is relatively small (i.e. arithmetic mean concentration $\approx$ geometric mean).

3.2 Deposition

Now let us consider the deposition at a certain receptor. The (local) removal coefficient at that receptor may vary with time, e.g. the wet removal coefficient depends on the intensity of precipitation. Concerning the effect of the application of averaged input data, the following question arises:

Knowing the mean concentration at a receptor, can we compute the mean deposition at that receptor simply by multiplying the mean concentration by the mean removal coefficient?

In order to simplify the subsequent discussion, let us consider only one removal process (wet deposition). We observe the following effect: If there is intense precipitation (i.e. a large removal coefficient) at the receptor, then it probably has been raining also along the trajectory prior to arriving at the receptor, i.e. a large portion of the pollutant has already been deposited along the trajectory and only a small concentration arrives at the receptor. If we interpret deposition $d = \sum_l K_{(l)} \cdot c_{(l)}$ (l = number of trajectory) as a weighted average (average of K weighted by c or average of c weighted by K), then large values of K are associated with small "weights" c (i.e. large values of K are under-represented in the weighted average in comparison to the usual arithmetic average), whereas small values of K are associated with large "weights" c. That is why, the weighted average is lower than the product of the mean K and the mean c. In other words:

Multiplying the mean concentration by the mean (local) removal coefficient, we overestimate the deposition. As, however, the mean concentration is underestimated by applying averaged removal coefficients, it is not clear at this stage, whether the deposition will finally be overestimated or underestimated.

The verbal reasoning of the preceding paragraph can be put on a stronger mathematical basis. Keeping in mind that we consider frequency distributions of input data (e.g. the frequency of the occurrence of a certain precipitation intensity in a certain year or in the "climatologic mean") rather than probability distributions, let us adopt the "language" of probability theory: The mean value of some variable $\mathcal{X}$ (e.g. concentration or removal coefficient) corresponds to its expected value $E(\mathcal{X})$; the coincidence of large or small values of some variable $\mathcal{X}$ (e.g. removal coefficient) with large or small values of a variable $\mathcal{Y}$ (e.g. concentration) is expressed by the covariance $Cov(\mathcal{X}, \mathcal{Y})$.

From the definition of the covariance

$$Cov(\mathcal{X}, \mathcal{Y}) = E(\mathcal{X} \cdot \mathcal{Y}) - (E\mathcal{X}) \cdot (E\mathcal{Y})$$

it is only one step to the equation

$$(E\mathcal{X}) \cdot (E\mathcal{Y}) = E(\mathcal{X} \cdot \mathcal{Y}) - Cov(\mathcal{X}, \mathcal{Y}) \tag{6}$$

or, for our special application,

$$(EK) \cdot (Ec) = E(K \cdot c) - Cov(K, c) \tag{7}$$

As larger (local) removal coefficients K usually coincide with a smaller pollutant concentration arriving (cf. reasoning above), K and c are negatively correlated. Thus, it follows immediately from (7) that

$$(EK) \cdot (Ec) > E(K \cdot c),$$

i.e the multiplication of the mean concentration by the mean removal coefficient results in an overestimation of the mean deposition.

Analogous arguments are applicable for dry deposition: The (local) dry deposition coefficient at a certain receptor is expected to be positively correlated with the dry removal in the vicinity of the receptor (when dry deposition is small due to increased atmospheric stability at night, it is also night-time in the vicinity of the receptor; surface properties like, for instance, snow cover are positively correlated within the surroundings of a receptor). As a result, the (local) dry removal coefficient and the arriving concentration can be expected to be negatively correlated. Therefore, (7) is also applicable to dry deposition.

Until now, we have dealt with wet and dry deposition separately, in order to simplify the problem under consideration. For answering the question, whether such separate investigation of processes would be correct even for quantative estimations, Equation (6) is helpful again. After introducing the abbreviations

$$\begin{aligned} \mathcal{K}_{dry}(t) &= \int_0^t K_{dry}(\tau) d\tau, \\ \mathcal{K}_{wet}(t) &= \int_0^t K_{wet}(\tau) d\tau, \end{aligned}$$

we obtain from (3)

$$\begin{aligned} c(t) &= c_0 \cdot e^{-(\mathcal{K}_{dry}(t) + \mathcal{K}_{wet}(t))} \\ &= c_0 \cdot e^{-\mathcal{K}_{dry}(t)} \cdot e^{\mathcal{K}_{wet}(t))}. \end{aligned}$$

If $\mathcal{K}_{dry}$ and $\mathcal{K}_{wet}$ (and thus also $e^{-\mathcal{K}_{dry}}$ and $e^{-\mathcal{K}_{wet}}$) can be assumed to be statistically independent (which is not unrealistic, even though smaller effects like an enhanced dry deposition of soluble pollutants on wet surfaces occur), then we obtain from (6):

$$E(e^{-\mathcal{K}_{dry}} \cdot e^{\mathcal{K}_{wet}}) = E(e^{-\mathcal{K}_{dry}}) \cdot E(e^{-\mathcal{K}_{wet}}). \tag{8}$$

For the mean coefficients $\bar{\mathcal{K}}_{dry}$ and $\bar{\mathcal{K}}_{wet}$ we obtain

$$e^{-\bar{\mathcal{K}}_{dry} + \bar{\mathcal{K}}_{wet}} = e^{-\bar{\mathcal{K}}_{dry}} \cdot e^{\bar{\mathcal{K}}_{wet}}$$

and thus from (8)

$$\frac{e^{-\bar{\mathcal{K}}_{dry} + \bar{\mathcal{K}}_{wet}}}{E(e^{-\mathcal{K}_{dry} + \bar{\mathcal{K}}_{wet}})} = \frac{e^{-\bar{\mathcal{K}}_{dry}}}{E(e^{\mathcal{K}_{dry}})} \cdot \frac{e^{-\bar{\mathcal{K}}_{wet}}}{E(e^{\mathcal{K}_{wet}})},$$

i.e. the simultaneous effect of applying averaged values for dry *and* wet removal coefficients can be estimated by simply multiplying their individual effects. For an illustration see Table 5 (which, however, includes the effect of applying an averaged mixing height separately): The product of

the numbers in the first three rows (application of single averaged parameters) differs from the corresponding elements in the fourth row (application of simultaneously averaged parameters) by less than 10%.

3.3 The Effect of Averaging Wind Speed and Travel Distance

If (1), (2) are applied within a climatologic model, averaged values are used not only for the removal coefficient K, but also for the wind speed $u(t)$ at the receptor and the travel time t. The variance of the travel time t results from two effects: Variation of the travel distance between a (fixed) source and receptor (caused by the varying curvature of the trajectories) and variation of the (mean) wind speed along different trajectories.

Let us first consider the effect of an averaged travel distance between some source and a certain receptor. In order to separate effects, we assume that u and K are constant. Then we obtain from (1)-(2) after expressing the travel time t by the travel distance s:

$$c(s) = \frac{Q}{u} \cdot e^{-\frac{K}{u} \cdot s} \tag{9}$$

We observe that s enters this formula "in the same way" as K does. That is why all considerations concerning K are also applicable to s, i.e. the application of a mean travel distance will produce an underestimation of concentrations, and of depositions as well (because of the proportionality $d = K \cdot c$, which is independent of s).

If, however, instead of the mean travel distance, the straight-line travel distance s_{min} between source and receptor is applied (what is usually done), then s is underestimated (or, at least, not overestimated) for each trajectory, which results in an overestimation of the concentration arriving (and deposition, too).

Now let us consider the effect of applying an averaged wind speed. In order to separate effects, we assume that K and the travel distance s are constant, so that we obtain from (1)-(2) analogously to (9):

$$c(s) = \frac{Q}{u(s)} \cdot e^{-\int_0^s \frac{K}{u(\rho)} d\rho} \tag{10}$$

Here $\frac{1}{u}$ enters the formula in the same way as the removal coefficient K enters the formula for the deposition

$$d(t) = K(t) \cdot c_0 \cdot e^{-\int_0^t K(\tau) d\tau}$$

(with the only difference that, when averaging u, we apply $\frac{1}{\bar{u}}$ instead of $\left(\frac{1}{u}\right)$, where the bar $\bar{}$ denotes averaging over several arriving trajectories). We are going to explain this in more detail: As can be seen from (10), u influences the concentration c at the receptor in two ways: It determines the travel time and thus the removal of the pollutant along the trajectory; and it influences the spreading of an air parcel parallel to the trajectory (which is reflected by the factor $\frac{1}{u(s)}$). The application of the mean value $\left(\frac{1}{u}\right)$ instead of $\int_0^s \frac{1}{u(\rho)} d\rho$ would result in an

underestimation of the averaged (long-term) value of $c(s)$ (the reasoning from Section 3.1 can be applied directly in this case). However, this underestimation is partly compensated because of the use of $\frac{1}{\bar{u}}$ instead of $\left(\frac{1}{u}\right)$ (because of $-\frac{1}{\bar{u}} > -\left(\frac{1}{u}\right)$, which follows from the fact that the harmonic mean $1/\left(\frac{1}{u}\right)$ is less than the arithmetic mean $\bar{u}$) .

Moreover, the factor $\frac{1}{u(s)}$ in front of the exponential expression results in a further compensation of the underestimation (or even in an overestimation). In order to explain this, we can argue in total analogy to Section 3.2: The wind speed $u(s)$ at the receptor is positively correlated with the wind speed in the vicinity of the receptor (at least). That is why $\frac{1}{u(s)}$ will be positively correlated to $\frac{1}{u}$ in the vicinity of the receptor (at least) and, as a consequence, also with $\int_0^s \frac{1}{u(\rho)} d\rho$. That means that a rather strong removal along the trajectory (resulting from a small wind speed and thus a large travel time) will most probably be associated with a weak spreading parallel to the trajectory (resulting from a small wind speed $u(s)$) . In total analogy to the reasoning in Section 3.2, this negative correlation between removal along the trajetory and spreading (or dilution) parallel to it results in a compensation of underestimation (or even overestimation) of the mean concentration (and also deposition, which is proportional to c, independently of u).

4 Analytical Formulae

4.1 Concentration

Let us consider a certain receptor and some (fixed) pollutant source again. The concentration along one trajectory connecting these two points can be computed by (3) (omitting the argument t here):

$$c = c_0 \cdot e^{-\mathcal{K}},$$

where

c_0 = concentration at the source
(we have implicitely assumed a constant wind speed),

c = concentration at the receptor,

$\mathcal{K}$ = integrated (along the trajectory) removal coefficient.

Now assume that $\mathcal{K}$ can assume not only two discrete values like in Section 3.1, but a continuous distribution of the frequency of the occurrence of $\mathcal{K}$ values is given. Thus, the factor of the underestimation of the mean concentration $E\left(c_0 \cdot e^{-\mathcal{K}}\right)$ by applying a mean removal coefficient $E(\mathcal{K})$ in the computations depends on the form of the frequency distribution for $\mathcal{K}$ (as well as on the variance of $\mathcal{K}$). The variance of

$$\mathcal{K} = \int_0^t \frac{v_d}{h_{mix}} + \frac{W \cdot P}{h_{mix}} d\tau$$

is caused by variations in the dry deposition velocity v_d, the precipitation intensity P, the mixing height h_{mix}, and the travel time t (variations from one trajectory to another).

Analytical results for three simple, but frequently used, distributions are given in Table 1. They have been obtained by straight-forward integration.

The result for the normal distribution should be applied carefully (i.e. only for small σ^2), since the assumption of normally distributed $\mathcal{K}$ values is unrealistic in the sense that negative $\mathcal{K}$ are allowed to occur, which correspond to an exponential *increase* of the concentration in $c = c_0 \cdot e^{-\mathcal{K}}$.

The gamma distribution is rather realistic, for instance, for describing the frequency of precipitation amounts, especially if integrated (along trajectories) amounts are considered (cf. Fig. 3).

In order to give an example for the application of the theoretical formulae, let us assume that we are interested in estimating the effect of the application of averaged precipitation data. Then $\mathcal{K}$ represents wet removal. (Remember that, according to Section 3.2, wet and dry removal can be treated separately). In order to exclude the effect of varying mixing heights h_{mix} and travel times t, we assume a fixed mixing height (900 m) and consider concentration versus travel time t rather than versus travel distance. If we want to apply a formula from Table 1, we have to estimate the variance σ^2 of the integrated (along trajectories) removal coefficient $\mathcal{K} = \int_0^t K(\tau)d\tau$. This is rather difficult, because σ^2 depends on the covariance between the coefficients $K(\tau)$ which are met on the trajectory:

$$\begin{aligned}
\sigma^2 &= E(\mathcal{K} - \bar{\mathcal{K}})^2 \text{ with } \bar{\mathcal{K}} = E(\mathcal{K}) \\
&= E\left(\int_0^t K(\tau)d\tau - \int_0^t \bar{K}d\tau\right)^2 \text{ with } \bar{K} = E(K) \\
&= E\left(\int_0^t (K(\tau) - \bar{K})d\tau\right)^2 \\
&= E\left(\int_0^t (K(\tau) - \bar{K})d\tau\right) \cdot \left(\int_0^t (K(\tau') - \bar{K})d\tau'\right) \\
&= E\int_0^t \int_0^t (K(\tau) - \bar{K}) \cdot (K(\tau') - \bar{K})d\tau d\tau' \\
&= \int_0^t \int_0^t E(K(\tau) - \bar{K}) \cdot (K(\tau') - \bar{K})d\tau d\tau' \\
&= \int_0^t \int_0^t Cov(K(\tau), K(\tau'))d\tau d\tau'
\end{aligned}$$

In the following we shall present results for two extreme cases:

1. *"Full information on σ^2":* Here this information is obtained by analysing the precipitation data on the 1460 trajectories arriving at the receptors under consideration during one

year (1985). However, in usual circumstances, such information would probably not be available.

2. *"No information on σ^2":* Suppose that we know only the mean value $\bar{\mathcal{K}}$ and do not have any information about the variance σ^2 of $\mathcal{K}$. The only additional information we have is that $\mathcal{K}$ is non-negative, i.e. may range in the interval $[0, \infty)$. If one wants to select a frequency distribution "using all information which one has, but avoiding any information that one does not have" (Theil and Fiebig, 1984), it is often recommended to apply the "maximum entropy principle" (Tiwari and Hobbie, 1976; Theil and Fiebig, 1984). The essential idea consists in maximizing the entropy of the frequency distribution subject to the constraints expressing the whole information that we have . In the present situation ($\mathcal{K} \in [0, \infty)$, $E(\mathcal{K}) = \bar{\mathcal{K}}$) this results in the exponential distribution (frequency distribution function $f(\mathcal{K}) = \bar{\mathcal{K}} \cdot e^{-\mathcal{K}/\bar{\mathcal{K}}}$) (Kagan et al., 1973; Theil and Fiebig, 1984), which fortunately happens to be a special case of the gamma distribution (with $\sigma = \bar{\mathcal{K}}$).

We have applied the formula in Table 1 for both assumptions on σ^2 and compared the results to the concentration computed by applying a mean wet removal coefficient as well as the "real" mean concentration obtained from calculations along 1460 separate trajectories. The results are contained in Table 2.

It can be seen that the assumption of exponentially distributed wet removal coefficients yields results which are superior to results calculated for "mean drizzle". Even if one would reject them as not being totally exact, a comparison between them and the "mean drizzle" case provides good information about the order of the deviation of the "mean drizzle" case from the "real" case, i.e. the magnitude of the effect (error) of applying an averaged removal coefficient.

If no information about the form of the frequency distribution function for $\mathcal{K}$ is available, one might again exploit the maximum entropy principle or apply an approximation of the following form, which is valid for arbitrary distribution functions:

$$
\begin{aligned}
& E\left(e^{-\mathcal{K}}\right) \\
= \; & E\left(e^{-\bar{\mathcal{K}}} \cdot e^{-(\mathcal{K}-\bar{\mathcal{K}})}\right) \\
= \; & e^{-\bar{\mathcal{K}}} \cdot E\left(e^{-(\mathcal{K}-\bar{\mathcal{K}})}\right) \\
\approx \; & e^{-\bar{\mathcal{K}}} \cdot E\left(1 + (\mathcal{K}-\bar{\mathcal{K}}) + \frac{1}{2}(\mathcal{K}-\bar{\mathcal{K}})^2\right) \text{ (truncated Taylor series)} \\
= \; & e^{-\bar{\mathcal{K}}} \cdot \left(1 + E(\mathcal{K}-\bar{\mathcal{K}}) + \frac{1}{2}E(\mathcal{K}-\bar{\mathcal{K}})^2\right) \\
= \; & e^{-\bar{\mathcal{K}}} \cdot \left(1 + \frac{1}{2}\sigma^2\right) \\
\approx \; & e^{-\bar{\mathcal{K}}} \cdot e^{\frac{1}{2}\sigma^2} \text{ (truncated Taylor series)},
\end{aligned}
$$

which coincides with the result for the normal distribution. It is equivalent to

$$e^{-\bar{\mathcal{K}}} \approx e^{-\frac{1}{2}\sigma^2} \cdot E\left(e^{-\mathcal{K}}\right),$$

i.e. by applying an averaged (integrated) removal coefficient $\bar{\mathcal{K}}$, we underestimate the mean concentration by a factor of approximately $e^{-\frac{1}{2}\sigma^2}$, where σ is the standard deviation of $\mathcal{K}$ (i.e. a measure of the variability of $\mathcal{K}$ from one trajectory to another).

4.2 Deposition

It has been explained in Section 3.2 that the factor of over- or underestimation of the deposition depends on the correlation (or covariance) between the (local) removal coefficient at the receptor and the integrated removal coefficient along the trajectories. It is extremely difficult both to compute this correlation for "real world" cases and to include it in analytical formulae. That is why we present analytical results only for the most simple case: We assume the removal coefficient K to be constant along each trajectory (i.e. "perfect correlation"; it follows $\mathcal{K}(t) = K \cdot t$); but it may vary from trajectory to trajectory. Results for this case are contained in Table 3. Here K may represent either wet or dry or wet + dry removal.

Analogously to Section 4.1, we can derive an approximation to $E\left(K \cdot e^{-\mathcal{K}t}\right)$ which is valid for an arbitrary frequency distribution function:

$$\begin{aligned}
& E\left(K \cdot e^{-\mathcal{K}t}\right) \\
= \; & E\left(K \cdot e^{-\bar{K}t} \cdot e^{-(K-\bar{K})t}\right) \\
= \; & e^{-\bar{K}t} \cdot E\left(K \cdot e^{-(K-\bar{K})t}\right) \\
\approx \; & e^{-\bar{K}t} \cdot E\left((\bar{K}+d)(1-dt+\frac{1}{2}d^2t^2\right) \text{ where } d := K - \bar{K} \\
= \; & e^{-\bar{K}t} \cdot E\left(\bar{K} + (1-\bar{K}t)d + (\frac{1}{2}\bar{K}t^2 - t)d^2 + \frac{1}{2}t^2d^3\right) \\
\approx \; & e^{-\bar{K}t} \cdot \left(\bar{K} + (\frac{1}{2}\bar{K}t^2 - t)\sigma^2\right) \text{ (third order term has been omitted)} \\
= \; & \bar{K} \cdot e^{-\bar{K}t} \cdot \left(1 + \left(\frac{1}{2}\sigma^2t^2 - \frac{\sigma^2 t}{\bar{K}}\right)\right) \\
\approx \; & \bar{K} \cdot e^{-\bar{K}t} \cdot e^{\frac{1}{2}\sigma^2t^2} \cdot e^{-\frac{\sigma^2}{\bar{K}}t},
\end{aligned}$$

i.e.

$$\bar{K} \cdot e^{-\bar{K}t} \approx e^{-\frac{1}{2}\sigma^2t^2} \cdot e^{\frac{\sigma^2}{\bar{K}}t} \cdot E\left(K \cdot e^{-Kt}\right).$$

That means that, based on the concentration computed with a mean removal coefficient, the deposition is overstimated by a factor of approximately $e^{\frac{\sigma^2}{\bar{K}}t}$. The combination of both effects (underestimation of concentration, overestimation of deposition based on that concentration) results in an overestimation of the deposition when (approximately)

$$\frac{\sigma^2}{\bar{K}}t > \frac{1}{2}\sigma^2t^2, \text{ i.e. } \bar{K}t < 2,$$

and in an underestimation of the deposition when (approximately) $\bar{K}t > 2$. It is interesting to note that the (perhaps difficult to obtain) variance σ^2 does not appear in this estimate so that the latter one can be applied easily. However, one should keep in mind the assumptions and approximations involved in its derivation. In the case of wet deposition at Haapasaari (Finland) ($W = 350000$ for As, $\bar{R} = 0.3\ mm/6\ h$ on trajectories to Haapasaari, h_{mix} assumed to be 900 m) we obtain that the deposition is overestimated for travel times

$$t < \frac{2}{\bar{K}} \approx 103\ h,$$

which is, at least, not in contradiction with the results of Table 4.

The data in Table 4 indicate that, by applying a mean wet removal coefficient, we may overestimate the wet deposition at some receptor originating from emissions of a certain source by a factor of more than 2. The reason is the rather strong negative correlation between the removal coefficient at the receptor and the arriving pollutant concentration (cf. Equation (7)), which may be explained in words as follows (cf. also Section 3.2): If wet removal occurs at the receptor, then only a rather small concentration arrives because of the wet removal along the way to the receptor. This arriving concentration (in the case of precipitation at the receptor) is overestimated if a "constant drizzle" removal coefficient (which is relatively small because it distributes the precipitation uniformly over dry and wet periods) is applied. That is, even though for "real" precipitation as well as "constant drizzle" the same precipitation amounts are assumed, the wet deposition differs considerably, since rain falling in the "constant drizzle" case encounters a higher pollutant concentration at the receptor.

4.3 Possible Applications

Analytic formulae of the type presented in the Sections 4.1 and 4.2 may be applied to estimate the effect of averaged removal parameters in a simple air pollution transport model. *If* accurate information about the frequency distribution of the removal parameters is available (including rather exact knowledge of the standard deviation), they can even be applied to correct results obtained by a simple model.

If one is forced to draw the conclusion that the application of an averaged removal parameter does not yield sufficiently accurate results, one may prefer the following approach: The range of possible values of that parameter is divided into several subranges. Then model runs are performed for one parameter value out of each subrange, and, finally, the model results are superimposed (in the sense of a weighted average) according to the frequency of occurrence of each subrange. In this case error estimates as presented in Sections 4.1 and 4.2 can provide a tool for determining the necessary number of subranges. Concerning the form of the frequency distribution, it would be rather "natural" to assume a uniformly distributed parameter within each subrange, since, given only lower and upper bounds of a random variable, the maximum

entropy distribution coincides with the uniform distribution (Kagan et al., 1973; Theil and Fiebig, 1984).

5 Model Results

In order to estimate the magnitude of the effects of applying averaged input data (removal coefficients, wind speed, travel distance), we *simulated* these effects by running the trajectory model (cf. Section 2) with averaged input data and comparing the results to the original output of the trajectory model. This was done for all trajectories arriving at 5 receptors (Haapasaari, Finland; Arup, Sweden; Aspervatn, Sweden; Kiel, F.R.G.; Kecskemet, Hungary) in 1985. We are going to report mean (= average of the results for the 5 receptors) factors of over- or underestimation of the computational results.

In practice, there are (at least) three levels of averaging input data:

1. *Temporal averaging (T):* Suppose we know long-term averages of input data for all grid elements. Then we can apply them immediately in the model calculations (without any spatial averaging). The theoretical investigations in Section 4 refer to this case.

2. *Temporal and partially spatial averaging (T/pS):* Suppose we are interested in computing concentrations and depositions at a certain receptor, for which we know long-term averages of input data; but for the rest of the model area only one (temporal and spatial) average is known. Then we have to apply the latter average to calculate the removal along trajectories, but we can use the "local" information at the receptor under consideration to convert the arriving concentration into deposition values, thus taking into account local features like, e.g., orographically induced precipitation. This level of averaging of input data has been applied in the climatologic model of Alcamo et al. (1990). It does not guarantee a strict mass conservation, since the local deposition ($K(s) \cdot c(s)$) does not coincide with the local removal computed with a mean removal coefficient ($\bar{K} \cdot c(s)$). For example, the local wet removal at a receptor with large precipitation amounts may be rather high, whereas the concentration along the trajectory after passing that "wet" receptor is not affected by it (in the computations), because the mean removal coefficient is applied. However, the kind of averaging input data considered here does *not* introduce a systematic *bias* (if applied to several sources and receptors), cf. Appendix B.

3. *Temporal and spatial averaging (T/S):* Suppose we know only one (temporal and spatial) average of the input data. Then we have to apply this throughout the model calculations. In this case, the effects of temporal averaging combine with the effects of spatial averaging, so that at a certain receptor either an underestimation or overestimation of the concentration and deposition can occur. If, however, we are interested in the spatially (and

temporally) averaged model output, e.g. the total deposition within the model region, then the results of Section 4 can be applied directly (after replacing temporal averages by temporal and spatial averages). This can be reformulated in another way: If we are interested in the *expected value* of over- or underestimation at receptors within the model region, then the results of Section 4 can be employed again.

In the present study, we used only the input data corresponding to the $5 \cdot 1460$ trajectories mentioned above, so that, e.g., temporal averaging means computing the average of the input data at a certain grid element over all time periods in which it is met by one of those trajectories. This kind of "limited averaging" excludes the introduction of a bias resulting from the use of data corresponding to grid elements (or time periods) which are not covered by the trajectories under consideration; in other words: The "averaged-input" trajectory model and the original one operate with exactly the same input, which is then treated differently (cf. Fig. 1).

The simulation results for different input parameters as well as for different levels of averaging are displayed in Table 5. In each run the parameters indicated have been averaged, whereas the remaining were applied in their original form. The effects (of the temporal averaging) predicted in Section 3 (underestimation of concentration, possible overestimation of d_{wet} for averaged precipitation intensity and of d_{dry} for averaged dry deposition velocity) are correctly reflected by the results in Table 5. The cases "T/pS" and "T/S", though including also spatial averaging, yielded rather similar outcomes. The only major difference occurs for an averaged dry deposition velocity, for which the spatial averaging results in an underestimation of the deposition over land (and hence increased concentration), but a severe overestimation of deposition over sea (at the grid elements of two of the five stations under consideration, Arup and Aspervatn, a dry deposition velocity corresponding to the sea surface has been applied in the present model!).

In order to simulate the effect of applying an averaged or straight-line travel distance between a source and a receptor, we "stretched" or "shortened" all trajectories linking a source-receptor pair, so that they had the prescribed (averaged or straight-line) length (which is equivalent to applying the original length of the trajectories with an altered travel time). The simulation results are contained in Table 6. They confirm the effects predicted in Section 3.3: Underestimation of concentration and deposition in the case of the application of a mean travel distance (i.e. mean length of all curved trajectories linking a source and a receptor) and overestimation in the case of the application of the straight line distance.

The estimation of the effect of averaged input parameters on so-called transfer coefficients, i.e. the pollutant concentration (or deposition) at a certain receptor coming from one specified source, remained outside the scope of the present simulation (though the theoretical results of Section 4 are directly applicable to this case). However, as the deposition (or concentration) at a receptor is equal to the emission-weighted mean of transfer coefficients, e.g.

$$d_k = \sum_i e_i \cdot c_{ik} \ ,$$

where d_k = deposition at receptor k,

e_i = emission at source i,

c_{ik} = transfer coefficient from source i to receptor k,

the results presented can be interpreted as being the weighted mean of results obtained for transfer coeffients.

6 Conclusions

For the heavy metals transport model and the 5 European receptors under consideration, we can conclude that the averaging of input data describing removal processes (precipitation intensity P, dry deposition velocity v_d, mixing height h_{mix}) results in an underestimation of concentration and dry deposition by a factor of approximately 0.75 (i.e. the prediction amounts to 75% of the original value) and an overestimation of the wet deposition by a factor of approximately 1.5.

In the application of an averaged wind speed, there are overestimating as well as underestimating effects, which nearly cancel out.

The use of a mean (curved) travel distance between a source and a receptor brings about an underestimation of concentration and deposition by a factor of ≈ 0.9; whereas using the straight line distance, we overestimate concentration and deposition by a factor of ≈ 1.2.

(For a theoretical explanation of all effects see Section 3.)

These results indicate that the errors introduced by applying averaged input data in a long-term air pollution transport model are within the error bounds that are generally accepted (e.g. deviation from measurements within a factor of 2) and comparable to the errors due to uncertainties in the input parameters (e.g., doubling the dry deposition velocity and the washout ratio in our present example would decrease concentrations by a factor of 0.67). However, if one attempts to include more sophisticated descriptions of physical and chemical processes in a simple long-term model, one should keep in mind that errors of the order of a factor of 1.5 are already implicit in the simple model structure (i.e. the application of averaged input parameters). A possible remedy could consist in dividing the range of each parameter into several subranges and running the model for one parameter value per subrange. However, this procedure would require information about the frequency of the occurrence of each subrange and about correlations between different parameters, which may be difficult to obtain.

Appendix A: Derivation of the Basic Transport Equation

In this appendix the basic transport equation (1) will be derived. We consider the vertically and cross-wind integrated pollutant concentration (in $g\ m^{-1}$) along a trajectory. If this trajectory starts at a point source of emission intensity Q $[g\ s^{-1}]$, the initial concentration $\tilde{c}_0$ is obtained by setting the pollutant flux (through a plane which is perpendicular to the trajectory and situated in the immediate vicinity of the source) $F = u_0 \cdot \tilde{c}_0$ equal to the emission intensity Q:

$$\begin{aligned} u_0 \cdot \tilde{c}_0 &= Q \\ \tilde{c}_0 &= \frac{Q}{u_0} \end{aligned}$$

where u_0 is the wind speed at the source.

Assume, for simplicity, that the wind field is temporally constant, so that the form of the the trajectory does not change with time. Then we can define a (perhaps curved) coordinate system along the trajectory and describe the fate of the pollutant along the trajectory by the Eulerian equation

$$\frac{\partial c}{\partial t} + \frac{\partial}{\partial s}(uc) = -Kc, \tag{11}$$

where

$$\begin{aligned} s &= \text{path length along the trajectory,} \\ c(s,t) &= \text{pollutant concentration,} \\ u(s) &= \text{wind speed along the trajectory,} \\ K(s) &= \text{removal coefficient.} \end{aligned}$$

If we follow a certain air parcel on its way along the trajectory, we have to consider the pollutant concentration and the input data in a Lagrangian sense (indicated by $\tilde{}$ over the variables):

$$\tilde{c}(t) := c(\tilde{s}(t), t), \tag{12}$$

$$\tilde{u}(t) := u(\tilde{s}(t)), \tag{13}$$

$$\tilde{K}(t) := K(\tilde{s}(t)), \tag{14}$$

where $\tilde{s}(t)$ is the position of the air parcel at time t, which is determind by

$$\frac{d}{dt}\tilde{s}(t) = u(\tilde{s}(t)). \tag{15}$$

Remark: The assumption of a temporally costant form of the trajectory can be relaxed if we consider (11) only on a domain $\{(s,t)|s \in (\tilde{s}(t)-\epsilon, \tilde{s}(t)+\epsilon), t \in [t_0, t_{max}]\}$ with an arbitrarily small ϵ, (instead of $\{(s,t)|s \in [s_{min}, s_{max}], t \in [t_0, t_{max}]\}$), which is sufficient for "following" the air parcel under consideration.

In order to simplify the notation, in the following we omit the arguments of the temporally or spatially dependent variables, which are $(\tilde{s}(t),t)$ for c, $\tilde{s}(t)$ for u and K, and t for $\tilde{c}$, $\tilde{u}$, and $\tilde{K}$.

Differentiating (12), we obtain

$$\begin{aligned} \frac{d\tilde{c}}{dt} &= \frac{d\tilde{s}}{dt} \cdot \frac{\partial c}{\partial s} + \frac{\partial c}{\partial t} \\ &= u \cdot \frac{\partial c}{\partial s} + \frac{\partial c}{\partial t} \\ &= -\frac{\partial u}{\partial s} \cdot c - Kc \end{aligned} \tag{16}$$

because of (11) (after differentiating the product term on the left-hand side).

Taking into account that

$$\begin{aligned} \frac{d\tilde{u}}{dt} &= \frac{d\tilde{s}}{dt} \cdot \frac{du}{ds} \\ &= \tilde{u} \cdot \frac{du}{ds} \end{aligned}$$

(because of (13) and (15)), we obtain from (16) together with (12) - (14):

$$\frac{d\tilde{c}}{dt} = -\frac{1}{\tilde{u}} \cdot \frac{d\tilde{u}}{dt} \cdot \tilde{c} - \tilde{K}\tilde{c},$$

i.e.

$$\frac{1}{\tilde{c}} \frac{d\tilde{c}}{dt} = -\frac{1}{\tilde{u}} \cdot \frac{d\tilde{u}}{dt} - \tilde{K},$$

which yields after integration

$$\ln \tilde{c} - \ln \tilde{c}_0 = -[\ln \tilde{u} - \ln \tilde{u}_0] - \int_0^t \tilde{K}(\tau) d\tau,$$

i.e.

$$\begin{aligned} \tilde{c} &= \tilde{c}_0 \cdot \frac{\tilde{u}_0}{\tilde{u}} \cdot e^{-\int_0^t \tilde{K}(\tau)d\tau} \\ &= \frac{Q}{u_0} \cdot \frac{\tilde{u}_0}{\tilde{u}} \cdot e^{-\int_0^t \tilde{K}(\tau)d\tau}, \end{aligned}$$

and finally

$$\tilde{c}(t) = \frac{Q}{\tilde{u}(t)} \cdot e^{-\int_0^t \tilde{K}(\tau)d\tau},$$

which we wanted to prove (the tilde ˜ for indicating variables in the Lagrangian sense has been omitted in Section 1).

Appendix B: The Problem of Mass Conservation in the Case of a Temporal and Partially Spatial Averaging of Input Data

Let us consider the deposition along a straight trajectory, assuming a mean wind speed $\bar{u}$ and a mean removal coefficient $\bar{K}$. Then the concentration of a pollutant along this trajectory can be described by

$$c(s) = \frac{Q}{\bar{u}} \cdot e^{-\frac{\bar{K}}{\bar{u}}s}$$

(cf. (9)).

If the local deposition is computed by applying the local (non-averaged) removal coefficient $K(s)$, it is given by

$$\begin{aligned} d(s) &= K(s) \cdot c(s) \\ &= K(s) \cdot \frac{Q}{\bar{u}} \cdot e^{-\frac{\bar{K}}{\bar{u}}s} \end{aligned}$$

The total deposition along the trajectory amounts to

$$D := \int_0^\infty d(s)\, ds.$$

Strict mass conservation would be satisfied if

$$D = Q. \tag{17}$$

As different removal coefficients have been applied for computing the decay of the concentration along the trajectory ($\bar{K}$) and the local removal ($K(s)$), we cannot expect Equ. (17) to be strictly fulfilled. However, a weaker form of "mass conservation" can be proved: If we assume that the local removal coefficient $K(s)$ is a result of random fluctuations of the removal coefficient around its mean value $\bar{K}$ (i.e. $E(K(s)) = \bar{K}$), then we obtain for the expected value of D:

$$\begin{aligned} E(D) &= E\left(\int_0^\infty d(s)\, ds\right) \\ &= \int_0^\infty E\left(K(s) \cdot \frac{Q}{\bar{u}} \cdot e^{-\frac{\bar{K}}{\bar{u}}s}\right) ds \\ &= \int_0^\infty E\left(K(s)\right) \cdot \frac{Q}{\bar{u}} \cdot e^{-\frac{\bar{K}}{\bar{u}}s}\, ds \\ &= \int_0^\infty \bar{K} \cdot \frac{Q}{\bar{u}} \cdot e^{-\frac{\bar{K}}{\bar{u}}s}\, ds \\ &= Q \end{aligned}$$

References

Alcamo J., Bartnicki J., and Olendrzynski K. (1990) Modeling heavy metals in Europe's atmosphere: A combined trajectory-climatologic approach. *18th NATO-CCMS International Technical Meeting on Air Pollution Modeling*, Vancouver, 1990.

Derwent R.G. (1987b) Combatting the long range transport and deposition of acid nitrogen species in Europe. *AERE Report-R12799*, Harwell Laboratory.

Eliassen A. and Saltbones J. (1983) Modeling of long range transport of sulfur over Europe: a two year model run and some model experiments. *Atmospheric Environment 17*, 1457-1473.

Eliassen A., Hov O., Iversen J., Saltbones J., and Simpson D. (1988) Estimates of airborne transboundary transport of sulphur and nitrogen over Europe. *EMEP/MSC-W Report 1/88*, Oslo.

Kagan A.M., Linnik Y.V., and Rao C.R. (1973) *Characterization Problems in Mathematical Statistics.* John Wiley & Sons, New York.

Klug W. and Erbshäußer B. (1988) Application of the 'FAST'-method to a long term interregional air pollution model. *Water, Air, and Soil Pollution 40*, 51-70.

Theil H. and Fiebig D.G. (1984) *Exploiting Continuity - Maximum Entropy Estimation of Continuous Distributions.* Ballinger, Cambridge.

Tiwari J.L. and Hobbie J.E. (1976) Random differential equations as models of ecosystems - II. Initial conditions and parameter specifications in terms of maximum entropy distributions. *Math. Biosci. 31*, 37-53.

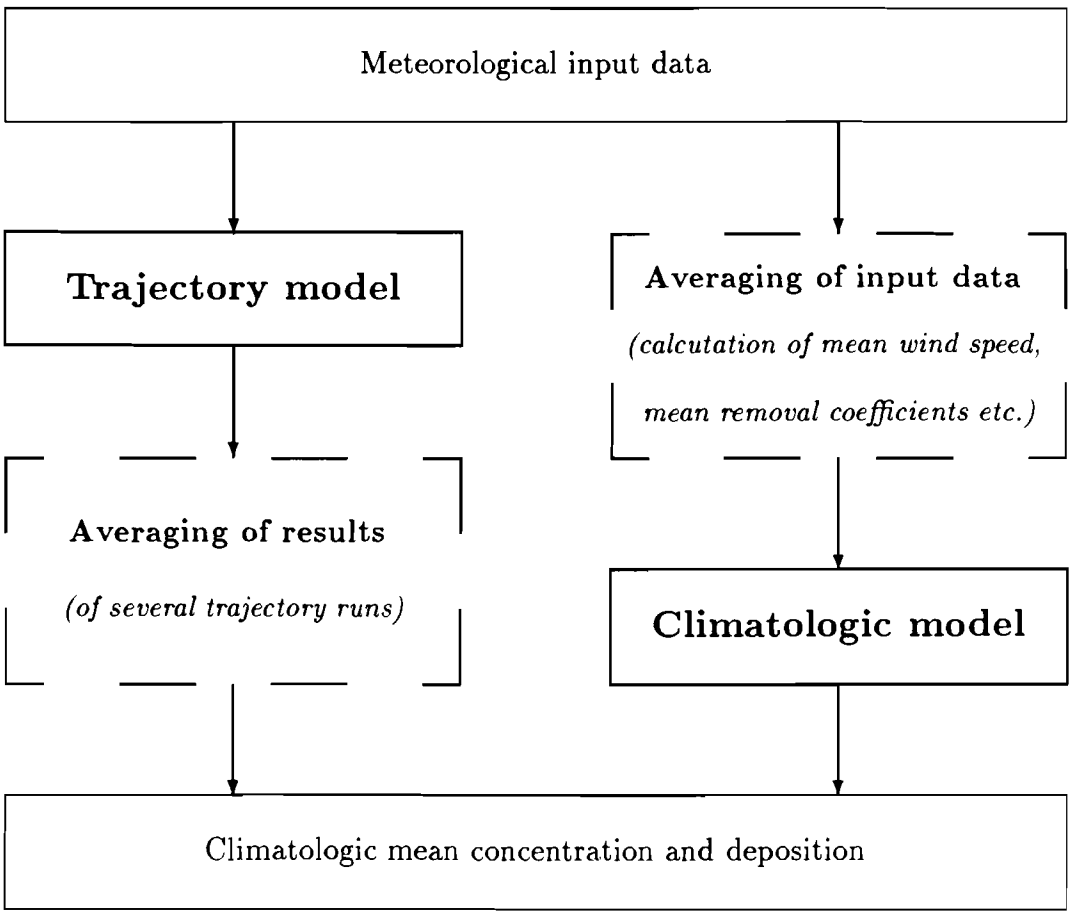

Figure 1: Schematic representation of the data flow for the trajectory model and the climatologic model.

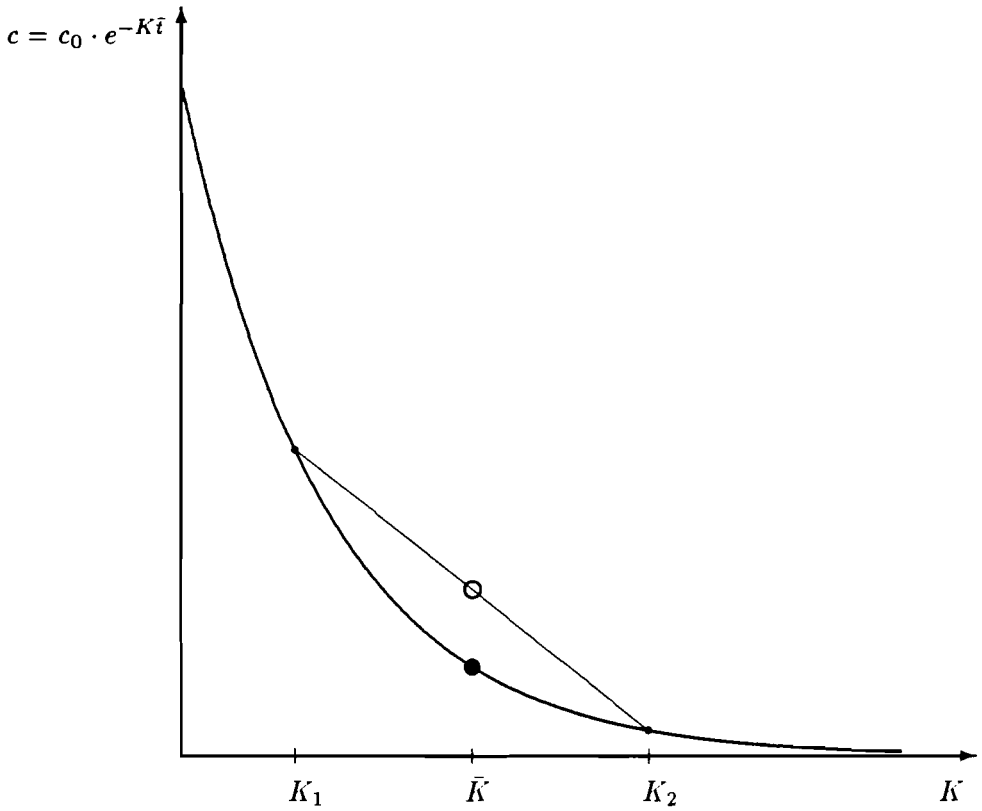

Figure 2: The effect of the application of a mean removal coefficient $\bar{K}$ (in the case of two possible values K_1 and K_2 for K):
Full circle: Concentration computed by applying the mean removal coefficient $\bar{K} = \frac{1}{2}(K_1 + K_2)$;
Blank circle: Average of the concentrations computed by applying the removal coefficients K_1 and K_2 separately.

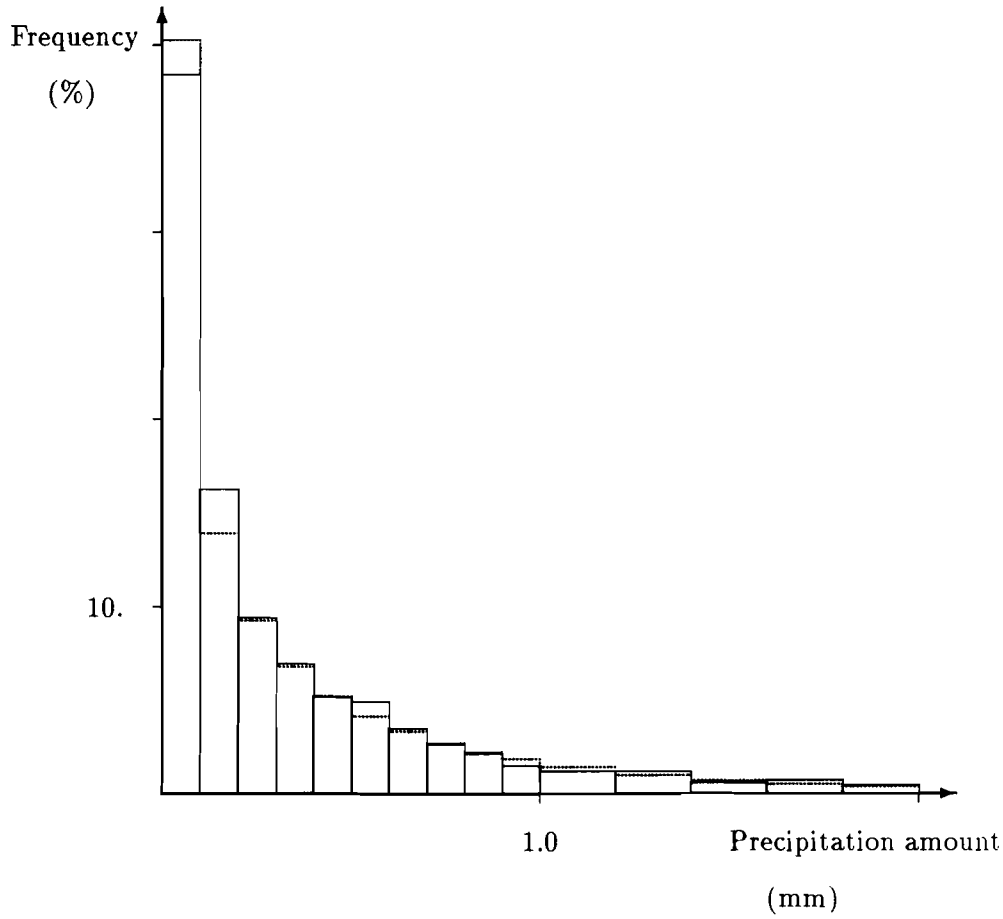

Figure 3: Frequency of the occurrence of precipitation amounts: measured values (solid boxes) versus gamma distribution (with the same mean value and standard deviation; dotted boxes) for integrated precipitation on 48 h-back trajectories arriving at Haapasaari, Finland.

	Uniform distribution	Normal distribution	Gamma distribution
Range	$[\bar{\mathcal{K}} - \Delta, \bar{\mathcal{K}} + \Delta]$	$(-\infty, \infty)$	$[0, \infty)$
Mean	$\bar{\mathcal{K}}$	$\bar{\mathcal{K}}$	$\bar{\mathcal{K}} = \alpha\beta$
Variance	$\sigma = \frac{1}{\sqrt{3}}\Delta$	σ	$\sigma = \sqrt{\alpha}\beta$
Frequency distribution function $f(\mathcal{K})$	$\frac{1}{2\cdot\Delta}$	$\frac{1}{\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{\mathcal{K}-\bar{\mathcal{K}}}{\sigma}\right)^2}$	$\frac{1}{\beta^\alpha \Gamma(\alpha)} \mathcal{K}^{\alpha-1} e^{-\frac{\mathcal{K}}{\beta}}$
$E(e^{-\mathcal{K}})$	$e^{-\bar{\mathcal{K}}} \cdot \frac{e^{\Delta} - e^{-\Delta}}{2\Delta}$	$e^{-\bar{\mathcal{K}}} \cdot e^{\frac{\sigma^2}{2}}$	$\left(\frac{1}{1+\frac{\bar{\mathcal{K}}}{\alpha}}\right)^\alpha$
$\phi = e^{-\bar{\mathcal{K}}} / E(e^{-\mathcal{K}})$	$\frac{2\Delta}{e^{\Delta} - e^{-\Delta}}$	$e^{-\frac{\sigma^2}{2}}$	$e^{-\bar{\mathcal{K}}} \cdot \left(1 + \frac{\bar{\mathcal{K}}}{\alpha}\right)^\alpha$

Table 1: Factor ϕ of the underestimation of the mean concentration in the case of the application of a mean removal coefficient (for three assumptions on the frequency distribution of the removal coefficient).

Travel Travel time (h)	Computations along 1460 trajectories	Theoretical formulae: Precipitation gamma-distributed	Theoretical formulae: Precipitation exponentially distributed	Computations with mean precipitation
12	81.2	80.8	74.9	71.6
24	70.4	69.5	61.3	53.1
48	55.4	56.0	46.7	31.9
72	47.2	49.3	39.2	21.2
96	40.1	43.3	34.8	15.3

Table 2: Mean pollutant mass (in % of the emission) arriving at Haapasaari (Finland) from sources which are 12...96 h up-wind (Wet deposition has been considered as the only removal process here).

	Uniform distribution	Normal distribution	Gamma distribution
Range of K	$[\bar{K}-\Delta, \bar{K}+\Delta]$	$(-\infty, \infty)$	$[0, \infty)$
Mean	$\bar{K}$	$\bar{K}$	$\bar{K} = \alpha\beta$
Variance	$\sigma = \frac{1}{\sqrt{3}}\Delta$	σ	$\sigma = \sqrt{\alpha}\beta$
Frequency distribution function $f(K)$	$\frac{1}{2\cdot\Delta}$	$\frac{1}{\sqrt{2\pi}}e^{-\frac{1}{2}\left(\frac{K-\bar{K}}{\sigma}\right)^2}$	$\frac{1}{\beta^\alpha\Gamma(\alpha)}K^{\alpha-1}e^{-\frac{K}{\beta}}$
$E(K\cdot e^{-Kt})$	$\mathcal{A}$	$\bar{K}\cdot e^{-\bar{K}t}\cdot e^{\frac{\sigma^2t^2}{2}}e^{-\frac{\sigma^2t}{\bar{K}}}$	$\bar{K}\cdot\left(\frac{1}{1+\frac{\bar{K}t}{\alpha}}\right)^{\alpha+1}$
$\phi = \bar{K}\cdot e^{-\bar{K}t}/E(K\cdot e^{-Kt})$	$\mathcal{B}$	$e^{-\frac{\sigma^2t^2}{2}}e^{\frac{\sigma^2t}{\bar{K}}}$	$e^{-\bar{K}t}\cdot\left(1+\frac{\bar{K}t}{\alpha}\right)^{\alpha+1}$

$$\mathcal{A} = \bar{K}e^{-\bar{K}t}\cdot\frac{e^{\Delta t}-e^{-\Delta t}}{2\Delta t} - \frac{1}{2\Delta t}e^{-\bar{K}t}\left(\left(\Delta+\frac{1}{t}\right)e^{-\Delta t} + \left(\Delta-\frac{1}{t}\right)e^{\Delta t}\right)$$

$$\mathcal{B} = \left\{\frac{e^{\Delta t}-e^{-\Delta t}}{2\Delta t} - \frac{1}{2\Delta t}\left(\left(\frac{\Delta}{\bar{K}}+\frac{1}{\bar{K}t}\right)e^{-\Delta t} + \left(\frac{\Delta}{\bar{K}}-\frac{1}{\bar{K}t}\right)e^{\Delta t}\right)\right\}^{-1}$$

Table 3: Factor ϕ of the over- or underestimation of the mean deposition in the case of the application of a mean removal coefficient (with perfect correlation along each trajectory, cf. Section 4.2).

Travel time (h)	Computations along 1460 trajectories	Computations with mean precipitation
12	1.19	2.46
24	0.87	1.82
36	0.72	1.40
48	0.64	1.10
60	0.57	0.89
72	0.53	0.73
84	0.50	0.62
96	0.44	0.53

Table 4: Mean pollutant mass (in % of the emission / h of travel time) deposited near Haapasaari (Finland) from sources which are 12...96 h up-wind (Wet deposition has been considered as the only removal process here).

Parameter	Type of Averaging								
	Temporal (T)			Temporal + partially spatial (T/pS)			Temporal + spatial (TS)		
	c	d_{dry}	d_{wet}	c	d_{dry}	d_{wet}	c	d_{dry}	d_{wet}
Precipitation intensity	0.80	0.79	1.74	0.77	0.76	1.67	0.77	0.76	1.29
Dry deposition velocity	0.97	1.06	0.98	1.13	1.23	1.09	1.13	3.17	1.09
Mixing height	0.84	0.85	0.94	0.83	0.84	0.93	0.83	0.83	0.93
All (P, v_d, h_{mix})	0.72	0.79	1.56	0.76	0.83	1.65	0.76	2.05	1.30
Wind speed	0.97	0.97	0.97	1.15	1.17	1.23	0.93	0.93	1.00

Table 5: The effect of applying averaged input data: Ratio of model results for averaged input data and standard model results (for concentration c, dry deposition d_{dry}, and wet deposition d_{wet}), for more details see Section 5.

Mean travel distance			Straight travel distance		
c	d_{dry}	d_{wet}	c	d_{dry}	d_{wet}
0.90	0.89	0.86	1.20	1.22	1.24

Table 6: The effect of applying a modified travel distance of pollutants: Ratio of model results for mean or straight travel distance and standard model results (for concentration c, dry deposition d_{dry}, and wet deposition d_{wet}), for more details see Section 5.