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## SPATIAL VARIABILITY OF RAIN WATER IMPURITIES IN MESOSCALE EVENTS

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#### ABSTRACT

Information on spatial variability of rain water impurities is important to assessing the accuracy of atmospheric deposition estimates. The time period of deposition that is of interest ranges from a single event to monthly or annual, depending on the application. Of the few measurements of rain water impurity variability that exist, most are for monthly deposition over space scales measured in hundreds of kilometers.

This paper reports variability of soluble Li, Na, Mg, K, Ca, and Zn over 14 single events, measured by 80 collectors over 2000 km<sup>2</sup> near St. Louis during summers, 1972-1974. Variability is expressed in terms of the distance at which correlations drop to 0.5 (the correlation distance). We observed correlation distances ranging from 0.9 to 3 km, for event deposition, in sharp contrast to correlation distances of 100 km or more measured previously for monthly concentration and deposition. Taking account of these correlations results in decreases in the confidence intervals about the network mean. For the impurities measured, these improvements ranged from 19 to 46%, relative to those obtained when correlations were ignored.

## INTRODUCTION

Information on total rain volume over an area is useful to a variety of scientists and engineers. The specific time period of the rain accumulation that is of interest varies with the application of the information. For example, engineers responsible for urban storm water runoff are concerned with time periods measured in hours, while those interested in crop yields may need weekly or monthly values.

Similarly, users of information on total deposition of the impurities in rain will vary in their requirements for the time period of the deposition measurements. Atmospheric scientists measuring the deposition of tracer materials released into a thunderstorm or charting deposition patterns of different impurities to learn more about precipitation scavenging processes need measurements on the time scale of individual storm duration. However, those concerned with long-term trends in deposition, or with nutrient budgets in a watershed, may be satisfied to know seasonal or annual inputs. . Similarly, if rainfall varied, but rainfall constituent concentrations were uniform over an area, we could estimate the deposition of constituents from a single rain water collector (to provide a sample for analysis) and multiple raingages. However, again, concentrations vary spatially within rains, so multiple collectors are required to estimate the mean or total deposition in an area.

Intuition tells us that the greater the variability of rainfall or rain impurity deposition over an area, the greater the number of raingages or collectors that are required to reach a given accuracy in estimating the area mean. Thus, several earlier studies have been directed toward measuring variability of rainfall or deposition over areas of varying size and over time periods of varying length. Some of these have emphasized the sampling requirements for various purposes, while others have been concerned with evaluating the accuracy achievable for a given network size and instrument spacing.

The purposes of this paper are 1) to show the spatial correlation structure of rainfall and rain water impurities in single convective events, and 2) to show how taking account of spatial correlation improves the accuracy of deposition measurements.

## MODEL DEVELOPMENT

Our first step was to develop estimates of the mean rainfall, or of the mean impurity deposition or concentration over some area, using observed values at a number of discrete points within the area. The usual estimator for this value is simply the sum of the observed values divided by the number of observations.

It has been shown previously [1] that the variance of any linear unbiased estimator (including the sample mean) is the product of the point variance (i.e., the usual variance of the network observations) and the variance reduction factor (VRF). The VRF depends only on the correlation function (i.e., the variation of the correlation coefficient with distance between samplers), and the geometry of the network (i.e., sampler density, number of samplers, and the shape and size of the network).

The VRF that might be chosen by an analyst who does not take account of the correlation between observations would be the reciprocal of the number of observations. For our network of 80 collectors, this VRF is 1/80, or 0.0125. The square root of the variance reduced in this way is, of course, the standard error of the mean.

To take account of correlation between sampling points, we adopted a model of the correlation function that is a smooth non-negative function which decays monotonically to zero in any direction from the origin. This expresses our belief that the correlation between samplers should be near one when they are very close, and decrease to zero as the distance between the collectors increases. For the 1972-1974 METROMEX precipitation chemistry network, we computed VRF's for the simple exponental correlation function

$$\rho(\mathbf{r}) = e^{-\kappa}$$

where r is the distance between collectors. Results are shown in Figure 1 for the VRF of the sample mean as a function of the observed parameter k. The figure also shows (solid curve) the relative improvement over the standard error of the mean that results from taking account of spatial correlation.

## RESULTS

Our data base consisted of 14 events from 1972-1974 having measurements of soluble Li, Na, Mg, K, Ca, and Zn. Figure 2 shows the variation of correlation with sampler separation distance for rainfall, where the individual points are mean values for all combinations of samplers in a given separation interval and over all 14 events. Standard error estimates of the correlations are also shown as a function of sampler separation at the bottom of Figure 2. The solid curve is the maximum likelihood fit to the data points, using the exponential function [1] above.

Similar results are given for Ca <u>deposition</u> in Figure 3. The correlations for Ca <u>concentration</u> (not shown) decayed very rapidly to zero, indicating little or no spatial correlation. For rainfall and deposition (Figures 2 and 3) the estimated correlations were usually negative for separations beyond about 20 km, so the family of functions used for fitting was not ideal. However, this probably had a minor effect on the degree of variance reduction achieved since the short distance correlations tend to be much larger than the more distant correlations in terms of absolute value.

A summary of results for rainfall and the soluble portions of six elements is given in Table 1. The results are summarized in terms of the parameter k in the exponential correlation function, the corresponding separation distance,  $r_{0.5}$ , at which the correlation becomes 0.5, the VRF, and the relative confidence interval (i.e., compared to the standard error) on the network mean deposition when spatial correlation is accounted for.

The results show that the dropoff in correlation was most rapid for Li, which reached a correlation of 0.5 at a separation of only 0.87 km, and least rapid for rainfall, for which  $r_{0.5} = 3.0$  km. Soluble Zn had the second most rapid decrease in correlation, while the remaining elements were clustered at  $r_{0.5}$  values only slightly smaller than that of the rainfall. The corresponding relative confidence intervals on the areal mean deposition, taking account of spatial correlation, ranged from 0.54 to 0.81 of those that would be obtained otherwise. Thus, taking account of spatial correlation gave improvements over the usual standard error ranging from 19 to 46%. The effect that accounting for spatial

[1]

Table 1. Summary of results for rainfall and soluble element deposition for 14 events in 1972-1974 METROMEX precipitation chemistry network -

	k km <sup>-</sup> 1	r0.5, km	VRF	/VRF/0.0125
Rainfall	0.234	3.0	0.0036	0.54
Li	0.795	0.87	0.0082	0.81
Na	0.244	2.8	0.0037	0.54
Mg	0.291	2.4	0.0042	0.58
К	0.250	2.8	0.0038	0.55
Ca	0.247	2.8	0.0037	0.54
Zn	0.451	1.5	0.0058	0.68

correlation has on the uncertainty of a measured network mean is illustrated for a typical event in Table 2.

		95% Confidence Interval		
	Units	Sample mean	Not accounting for correlation	Accounting for correlation
Rainfall	Cm	1.55	± 0.23	± 0.13 (8%)
Li	pg cm <sup>-2</sup>	240.	± 28.	± 23. (10%)
Na	ng cm <sup>-2</sup>	274.	± 34.	± 18. (7%)
Mg	ng cm <sup>-2</sup>	56.9	± 21.8	± 12.6 (22%)
K	ng cm <sup>-2</sup>	651.	±253.	±139. (21%)
Ca	ng cm <sup>-2</sup>	4830.	±690.	±370. (8%)
Zn	ng cm <sup>-2</sup>	17.1	± 7.4	± 5.0 (29%)

Table 2. Illustration of the effect of taking account of correlation on the uncertainty (95% confidence interval) of the network mean deposition for a typical event

# DISCUSSION AND CONCLUSIONS

This is a unique set of data on precipitation impurity variability, since it was collected in single events in summer, and over a relatively limited area (2000 km<sup>2</sup>) near a large urban center. It shows a much more rapid dropoff in the spatial correlation function than has been observed in monthly samples, for example. In 14 summer events from 1972 to 1974, the mean correlation function for rainfall dropped from 1.0 to 0.5 in 3.0 km. The correlation function for deposition of soluble impurities dropped even more rapidly. Zinc, which is known to have a number of point sources in the area, reached a correlation of 0.5 in only 1.5 km. The dropoff for Li was even more rapid, but the reasons for this behavior are not clear. Perhaps, as we have suspected for some time, it too has point sources in the area. It is also possible that the behavior of Li may also be related to its use as a tracer in a number of the events analyzed.

This rapid decrease of the correlation function with distance for events stands in contrast to previous results for monthly samples. Karol and Myatch [2] observed that correlations of several elements or ions reached 0.5 at distances ranging from 90 to 450 km in the USSR, and Granat [3] obtained very similar results in Sweden.

For rainfall and the soil derived elements, which have rather uniformly distributed sources, taking account of the spatial correlation of deposition gives confidence intervals on the network mean that are about half of those that would otherwise be obtained. For elements such as Zn, which have more localized sources, there is still improvement, but not as much; the improvement is about 30%. It should be noted that these results were obtained from sampling during summer seasons, when convective rainfall is prevalent. Convective rainfall is known to be more variable than that falling from layered clouds. In addition, the influence imposed by the city on both pollutant source distributions and downwind rainfall may also contribute to increased variability in this data set.

At this point in the research, and for the conditions sampled, it appears that between rainfall itself, deposition of impurities, and concentration of impurities, rainfall has the most spatial correlation. <u>Deposition</u> of elements having relatively uniform sources, such as Mg, Ca, K, and Na from the earth's crust, has slightly less spatial correlation on an event basis. Impurity <u>concentrations</u> have very little spatial correlation.

These results indicate that it requires a relatively dense network (on the order of 5 km spacing) of collectors to attain accuracies (95% confidence limits) of  $\pm 10-30\%$ , depending on the element measured, in single convective storms. Such events are worst cases, however. Lesser densities should give comparable accuracies in non-convective precipitation, or over longer sampling periods.

These data can also be used to guide the design of new collection networks. Network design is beyond the scope of this paper, however.

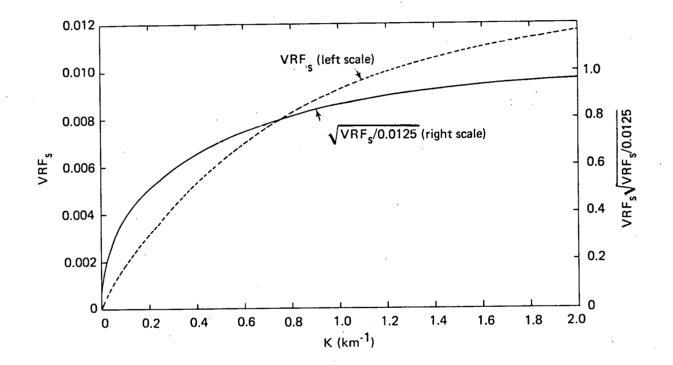
Future research on rain impurity variability should concentrate on time periods between event and monthly, for which some data are now available. Weekly data are now becoming available from the National Atmospheric Deposition Program (NADP). Similar evaluations of NADP weekly data, and their monthly, seasonal, and annual composites, should be done soon to evaluate their usefulness for their intended purposes.

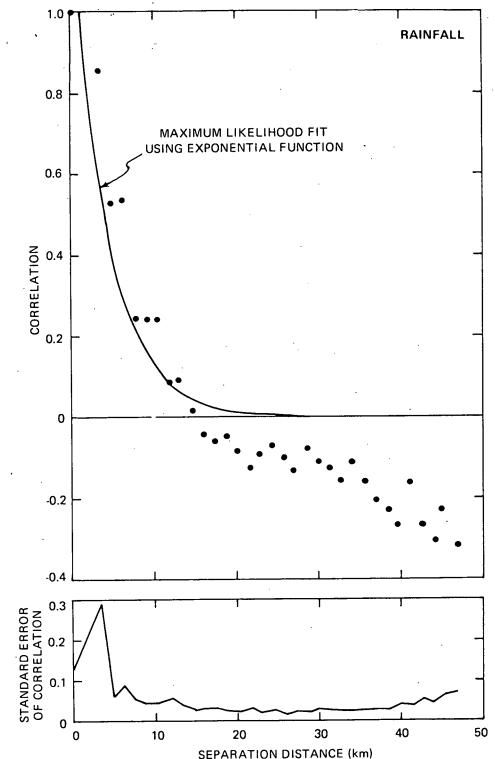
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- 2. I. L. Karol, and L. T. Myatch, "Contribution to the planning of the station network for measuring the chemical composition of precipitation," Tellus, 24, 421-427 (1972).
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# FIGURE LIST

Figure N	0.
1	Variation of VRF and VRF/0.0125 with sampler separation distance, for 14 events in the 1972-1974 METROMEX, precipitation chemistry network.
2	Observed spatial correlation for rainfall.
3	• Observed spatial correlations for Ca deposition.
4	Observed spatial correlations for Ca concentrations.





SEPARATION DISTANCE (km)

