United States
Environmental Protection
Agency

Great Lakes National
Program Office
111 West Jackson Blvd.
Chicago, Illinois 60604

EPA-905/4-87-001
GLNPO No. 87-03
March 1987

EPA

Design of a Great Lakes Atmospheric Inputs and Sources (GLAIS) Network

EPA-905/4-87-001
GLNPO No. 87-03
March 1987

DESIGN OF A GREAT LAKES ATMOSPHERIC INPUTS AND SOURCES (GLAIS) NETWORK

Thomas J. Murphy
DePaul University

Edward Klappenbach
Project Officer
Grant No. R005818-01

U.S. Environmental Protection Agency
Great Lakes National Program Office
230 South Dearborn Street
Chicago, Illinois 60604

DISCLAIMER

This report has been reviewed by the Great Lakes National Program Office, U.S. Environmental Protection Agency, and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the U.S. Environmental Protection Agency nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

FOREWORD

The Great Lakes National Program Office (GLNPO) of the United States Environmental Protection Agency was established in Region V, Chicago to focus attention on the significant and complex natural resource represented by the Great Lakes.

GLNPO implements a multi-media environmental management program drawing on a wide range of expertise represented by Universities, private firms, State, Federal and Canadian Governmental Agencies and the International Joint Commission. The goal of the GLNPO program is to develop programs, practices and technology necessary for a better understanding of the Great Lakes system and to eliminate or reduce to the maximum extent practicable the discharge of pollutants into the Great Lakes system. The Office also coordinates U.S. actions in fulfillment of the Agreement between Canada and the United States of American on Great Lakes Water Quality of 1978.

TABLE OF CONTENTS

APPENDIX A - List of Possible Equipment for a GLAIS Site.

APPENDIX B - Summary of Siting Information from GLAD Sites.

1. Site Location Data
2. Siting Criteria Data
3. Map showing distances (km) separating GLAD and GLPN sites

List of Figures | Page

I. CONCLUSIONS

1. Since any useful GLAIS program will of necessity, be complex and expensive, the objectives of any such project must be well thought out in advance, realistic and firmly based on good science and available technology.

2. The accuracy, precision and validity of the measurements will have to documented with a thorough quality assurance program to make their determination worth while.

3. There are at least five distinct mechanisms for inputs of materials from the atmosphere to bodies of water. They are: rain; snow; small particles (<~2 um); large particles (>~2 um); and vapor exchange. In general, very different methods and techniques are necessary to determine the inputs by each of these mechansims. Thus any program to determine total atmospheric inputs will, of necessity, be complex.

4. A program to measure the atmospheric inputs of toxic materials to the Great Lakes will be significantly different from most other atmospheric inputs programs now in operation. The reasons are that the other networks are focused primarily on the measurement of materials related to acid deposition, and they are measuring the inputs to land and vegatative surfaces. The GLAIS network will require different methods and techniques to collect the information on toxics, and to determine the inputs to large water surfaces.

5. In order to obtain information on the sources to the atmosphere of the materials going into the Lakes from the atmosphere, it will require a different set of measurement techniques than are used to obtain atmospheric inputs from the atmosphere.

6. Because of the expense in setting-up and operating (including analyses) a monitoring site for a variety of parameters involved with different mechanisms and/or also obtaining information on sources, a GLAIS network may consist of a relatively few sites.

7. A useful GLAIS network should be expected to evolve with time as the ability to determine specific components is acquired and improved. However, the procedures should be as unchanged as possible to permit measurements made over several annual cycles to be compared.

8. Because of the uncertainties in our capability to determine inputs occurring by any of the five above mechansims, any project to make a reasonable attempt to measure atmospheric inputs by one or more of the operating mechanisms will need some research component.

9. To be successful, expertise in a number of disciplines will be needed to develope a good network plan. These disciplines include: aerodynamics; chemistry; climatology; cloud physics; geochemistry; hydrodynamics; meteorology; and micrometeorology.

10. If the GLAIS network is to be designed and operated in a productive and efficient manner, it has to be kept abreast of the state of the art. It therefore must maintain contact with other networks in U. S., Canada and abroad.

II. INTRODUCTION

This report is a document to aid in the design of a network aimed at determining the identity, amounts, and sources to the atmosphere, of the materials coming into the Great Lakes from the atmosphere. It is meant to discuss what is known and what needs to be known about sampling atmospheric inputs to lakes. It does not strive to be thorough and comprehensive as this is not possible for such a broad subject in a short report. It is based on a variety of articles and reports, and relies heavily on suggestions and discussions with a variety of investigators in different disciplines. It assumes that experts in each of the necessary disciplines will be consulted or will participate in the final network design. Finally, it reflects the strengths, weaknesses, biases and misspellings of the author. Discussion among others is needed to fill it in and to round it off.

Familiarity with the literature in the area of atmospheric inputs or the reading of this report should lead one to conclude that our present abilities to determine atmospheric inputs to large bodies of water are limited. Some of the information desired is not able to be obtained at this time with a useful degree of precision and accuracy. If it is decided to implement a program to determine atmospheric inputs into the Great Lakes and/or their sources to the atmosphere, it must be a realistic one. The specific objectives of such a program will have to be carefully chosen so that they have a reasonable chance of being achieved.

The principal decision which needs to be made is the level of confidence desired in the results. As is discussed in this report, some measurement are able to be made with greater precision and accuracy than are others. For instance, estimates of rain inputs to the Great Lakes might be ±50%, while estimates of vapor exchange will probably be ±300%, and overall atmospheric inputs could be ±100%. If such estimates turn out to be valid, it has to be decided what level of confidence is needed for each type of measurement in order to justify the costs involved in obtaining it. It has to be recognized that there may be some parameters that would be most desirable to determine, but there is no reliable method of doing so at the present time.

The determination of atmospheric inputs is still very much in the development stage. This is particularly true for the toxic materials proposed to be measured in a GLAIS network. This will be a pioneering effort, with all of the hardships that suggests. There are not well proven techniques for making many of the the measurements that are needed. Thus, any atmospheric inputs plan that is implemented will necessarily have some developmental and/or research component associated with it.

In order to be useful, there must be a committment to operate a GLAIS network for several years. The principal reason for this is the great variability in the climatology. There are cold winters and warm winters, wet summers and dry summers, early springs and late springs, etc. Several year's data are needed to get a reliable idea of what the means and the annual variability are in the atmospheric inputs of the materials sampled for. Longer term sampling will be necessary to be able to discern trends in the data.

There are presently a number of other operational networks world-wide, involved with determining inputs from the atmosphere. Also there have been a large number of studies related to the determination of atmospheric inputs. The experiences and expertise gained inthose studies and by the other networks should be beneficial in planning and operating a GLAIS network. But this help has to be sought in the design and development stage.

Two important difference have to be kept in mind, however. The first is that the focus of a Great Lakes atmospheric inputs program is on the inputs to a water surface. Most of the studies and programs of the other networks to date, have dealt with inputs to land and vegetative surfaces. The meteorology over the Lakes is often different from the surrounding land areas, the lakes affect the regional climatology, the chemistry of the surface is different, and the ease and details of locating sampling equipment is different.

Secondly, intercomparisons with other networks will have to be done carefully as they all do things differently. Some of them only determine wet inputs, some determine wet and dry and some, NADP for instance, determine only soluble inputs. A goal of a GLAIS network, will be to determine total inputs. For some materials, the inputs of soluble or reactive forms may also be desired.

III. SAMPLING

General

Most of the problems in determining atmospheric inputs are related to the wind. The cause of the problems is that the particles are suspended in the air, they are a second phase. The movement of particles in the air is determined by several factors: being more dense than air, gravity attracts them to the earth's surface; air molecules collide with them, changing their velocity; and being suspended in the air, they tend to move with the air currents, the winds. The relative importance of these forces on particles is dependent principally on the particle size.

For large particles, like rain drops and particles larger than about 2 micrometers (um), gravity is the dominant factor. However as is apparent on windy days, the direction of fall of even very large particles like rain drops can be affected by the wind. This usually does not greatly affect the collection of rain, and the collection of rain is the least complicated of the different atmospheric inputs.

With small particles, those less than 1 um in diameter, the effect of gravity is small with respect to aerodynamic forces. Turbulence is the factor that most affects their deposition. Their velocity at any one time is almost as likely to be away from the earth as towards it. This greatly complicates their collection.

Snow flakes, while often as massive as rain drops, have a large surface area. This permits the aerodynamic forces to play a more important role, making snow collection much more difficult than rain and more like that of particles.

A program to determine atmospheric inputs to the Great Lakes would involve the collection of particles, droplets, flakes and possibly molecules in the vapor phase from the air. The quantitative sampling of most materials in the air, especially those associated with particles, is difficult. Some of the specific methods which have been used, and some of the difficulties associated with them will be discussed below.

The large variability of atmospheric inputs complicates the interpretation of the results from all precipitation networks. Most studies have found that the concentration and input statistics follow log normal distributions. High concentration or loading events are infrequent, but expected. It is commonly found that 10-15% of the annual loadings of most materials are contributed by 1% or fewer of the events. Thus the quality assurance program for a GLAIS network has to be structured so that these infrequent, large events are not summarily rejected.

QUALITY ASSURANCE

Sievering et al. (undated) and other discussants indicated that a thorough and complete quality assurance program will be REQUIRED for any GLAIS network. Such a program will have to be integrated into the GLAIS design. It will involve: determining the limit of detection, the quantification level, and the precision for all the analytical methods; the use of lab standards and reference standards to determine the accuracy of each of the measurements; determining blank levels for the sampling equipment and sampling system, for the sample containers and shipping system, and for the laboratory reagents, equipment and analytical system; defining the validity of the measurements by using standard operating proceedures, carefully documenting them, and identifying deviations from them (Gertler et al. 1985). With a good quality assurance program, each measurement made should yield a value with a known precision, accuracy and validity.

PRECIPITATION GAUGING

Knowledge of the amount of precipitation (rain + snow + sleet + hail) which falls at a collection site is useful for checking the relative collection efficiency of the samplers and for estimating and comparing the annual rainfall with other years at that station. In order to obtain the needed results, careful consideration must be given to the collection of precipitation and the interpretation of the results obtained.

Rain Gauging

The measurment of rainfall is among the best developed sampling techniques. "Standard" eight inch, cylindrical, manual-reading raingauges are frequently used in deposition networks to determine the amount of rain that falls. These are about the most accurate gauges available for rain measurements, but are biased low by 10% percent or more in high winds (Larson and Peck 1974, Fig 1). Recording raingauges, as the Belfort unit used by the GLAD network, have an additional low bias due to updrafts around the opening caused by the sloping side near the top of the gauge (Jones 1969). The principal problems remaining in accurately determining rainfall amounts are in selecting representative sites (sites which are not anomalously affected by hills, trees, buildings, etc.) for the gauges and in properly shielding the gauges from the winds.

A lot of work has been done on the remote sensing of rain (and snow) using RADAR. At this time, the method suffers from the problem that there are two independent variables, the rainfall rate and the RADAR reflectivity of the rainfall. Thus, either the rainfall rate or the size of the droplets (or snow flakes) needs to be known to determine the deposition rate and amount. RADAR is most useful in determining the precipitation amounts to localized areas which contain one or more well located and shielded gauges that can be used to calibrate the RADAR. The accuracy of the results of course, are still limited by the accuracy of the gauges.

An extensive one-year project to determine precipitation inputs to Lake Ontario was carried out in conjunction with IFYGL (Wilson 1977). Two weather RADARs were used along with an extensive rain gauge network. Precipitation into the Lake was estimated using the RADAR data and asssuming that systematic

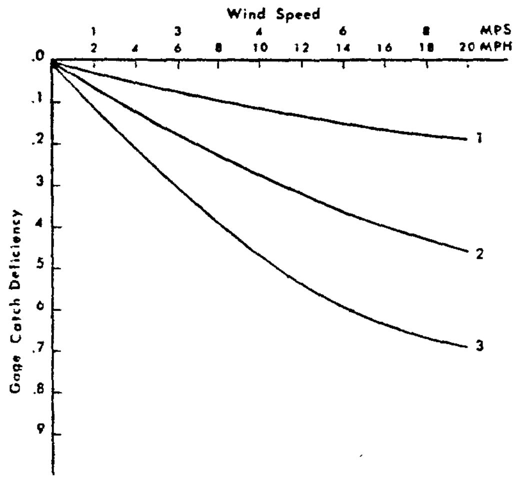


Fig. 1 Gage catch deficiencies versus wind speed. Line 1 is for rain (shield makes little or no difference in deficiencies), line 2 is for snow with a shielded gage, and line 3 is for snow with an unshielded gage

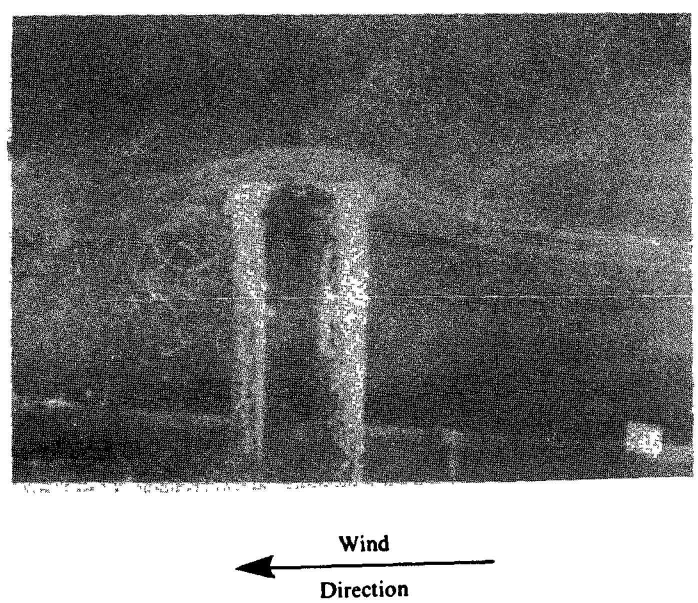


Fig. 2. Airflow pattern over an open cylinder (Goodison et al. 1981)

differences did not exist between overlake and overland gauge/RADAR ratios. The investigators felt that cold season precipitation over the Lake was underestimated because the lake-effect storms had low echo tops which were not seen by the RADAR.

Snow Gauging

Because of the problems with blowing and drifting snow, it is much more difficult to quantitatively sample snow than rain. If it is windy when the snow is falling, the air deflected by the sampler can lead to significant under or over sampling of snow by collectors designed for rain collection (Goodison et al. 1981; Fig. 2). For instance, a standard rain gauge can have a collection efficiency for snow of only ~30% (Fig. 3) in 9 m/s winds.

A number of shielded gauges have been designed which minimize the effects of wind on the collection of snow (Kurtyka 1953). The most widely used of these is the Nipher gauge (Goodison et al. 1981; Fig. 4). It is a cylinder surrounded by (inserted in) a parabolic shaped shield. The principal of the design is that all of the approaching wind that touches the nipher collector is deflected away from the interception cylinder, leaving the air flow over the cylinder completely unobstructed (ideally this air does not know the collector is there). Studies indicate that the Nipher gauge slightly over collects snow at low wind speeds, and the collection efficiency begins to fall off at wind speeds greater than 5 m/s (Fig. 3). At all speeds, however, it is a much more efficient snow sampler than rain gauges, even gauges with Alter, or other type wind shields, and it is a very good collector at low to moderate wind speeds. A Nipher gauge should be used for all snow measurements in a GLAIS network.

Finally, all snow gauging devices can "bridge" over under certain infrequent types of snow conditions. In addition, snow can fill-up the space between the outer shield and the inner cylinder in the Nipher-type collectors, leading to collection errors.

PRECIPITATION SAMPLING

Rain Sampling

Several wet-only rain samplers are commercially available which use a precipitation sensor to uncover a collection container during precipitation events. These collectors are thought to do a very good job of rain sampling. The collection areas of these samplers ranges from about 0.032 m^2 (Sangamo) to 0.21 m^2 (MIC). These samplers may be used to collect event samples, or they may be left out for a time period of a week, month, etc. and accumulate precipitation from one or more events. Some have been adapted for organic

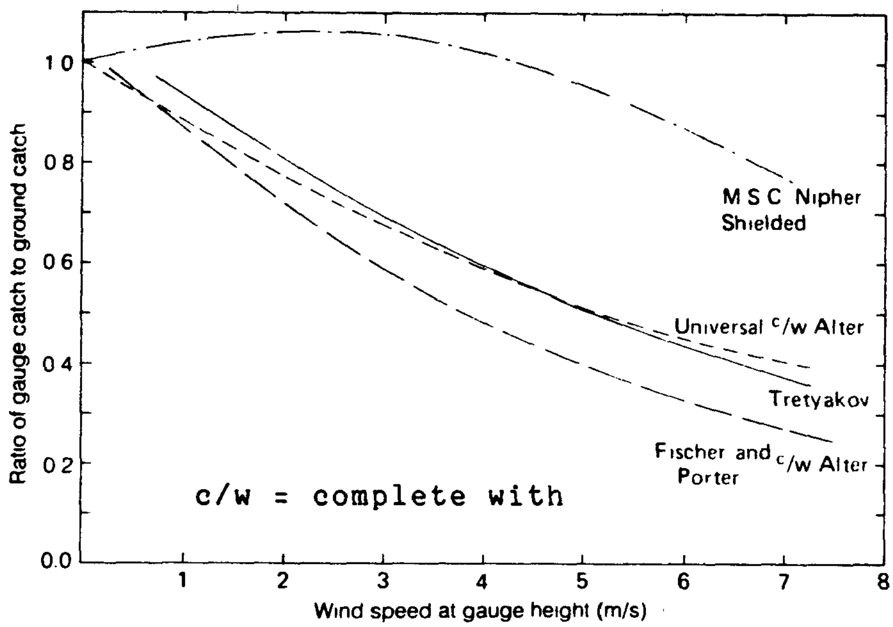


Fig. 3 Relationship between gauge catch and ground catch as a function of wind speed for different types of gauges (Goodison, 1978b).

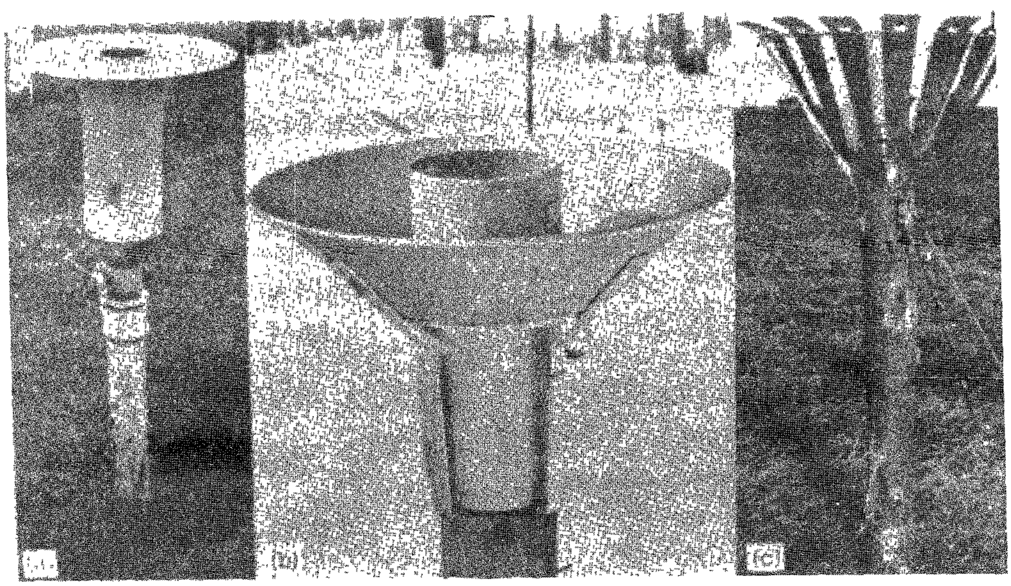


Figure 4. Types of gauges used to measure the water equivalent of snowfall in different countries. (a) MSC Nipher Shielded Guage (Canada). (b) Swedish SMHI Precipitation Gauge. (c) USSR Tretyakov Precipitation Gauge.

sampling by incorporating an extraction set-up in the apparatus for real-time sample processing. Due to aerodynamic effects, they are probably slightly less effecient rain samplers than the standard eight-inch rain gauge.

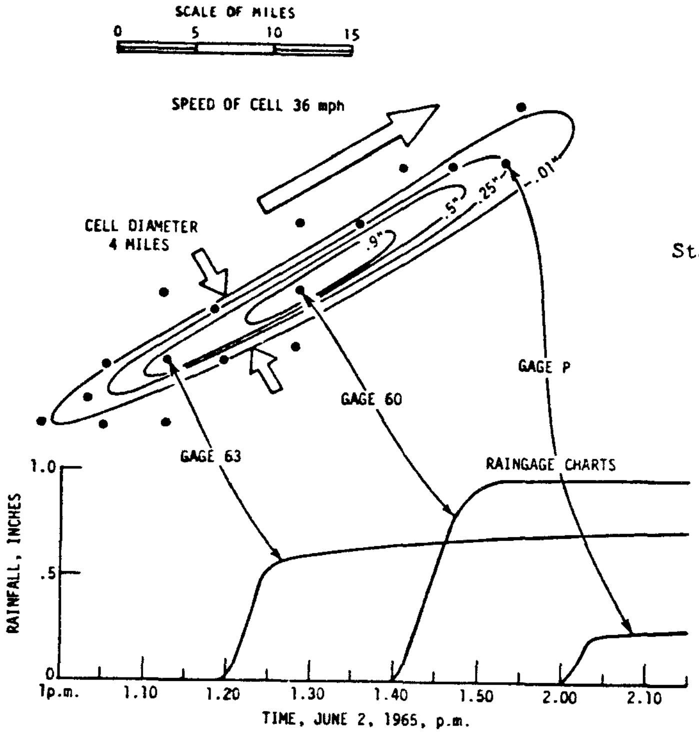
Ships with regular schedules on the Lakes, or which spend a lot of time on the Lakes, could be very useful platforms for collecting rain samples over the Lakes. A properly positioned sampler, operated when the ship is in the open waters, should give samples equivalent to those obtained by shore samplers (concentration information only). However, it might be difficult to determine the direction of the storm because of the motion of the ship.

When event rain samples are collected, brief or low-intensity rain events result in small sample sizes. These small samples cause problems for analyses which require more sample than is available. There are two possible solutions to this problem. First, a number of studies have shown that most materials show much higher concentrations in the first few mm of rain than their average concentration in an entire rain event. This is why it is very necessary to collect this early rain in each event. But it also means that while the volume is low, the amount of each material present in the sample may be sufficient for quantification. All that needs to be done in such cases is to add sufficient distilled water to bring the sample volume up to the minimum volume required for analysis. This volume of water added should be recorded and the appropriate corrections made when the results are interpreted.

Secondly, samples may be accumulated, and analyzed as a composite. This will result in the loss of the information with respect to the individual samples, but will result in the total contribution of these precipitation events being part of the data base. If the precipitation data are going to be analyzed on a directional basis, then these samples should also be composited and analyzed on the basis of the sector from which the air mass originated.

Wet Loadings

The spatial variability of rain in the Great Lakes basin is quite high. This is due mostly to the fact that 40-50% of the total precipitation comes from convective raincells (thunderstorms), often associated with squall lines and squall zones. Thunderstorms are usually composed of a number of individual storms cells, whose characteristics differ widely. However, the most frequent ones average 16 km long and 6 km wide, and last about 50 minutes (Stall and Huff 1971, Fig 5; Changnon 1981, Fig 6). Thus, rain gauges only a few km apart will typically collect quite different amounts of precipitation from each thunderstorm. While the amounts will tend to average out over the long term, annual variations for gauges 20 or so km apart are on the order of a factor of two. Due to this large variability, the amount of precipitation



Stall and Huff (1971)

Fig. 5

STANLEY A. CHANGNON, JR.   AUGUST 1981

JOURNAL OF THE ATMOSPHERIC SCIENCES

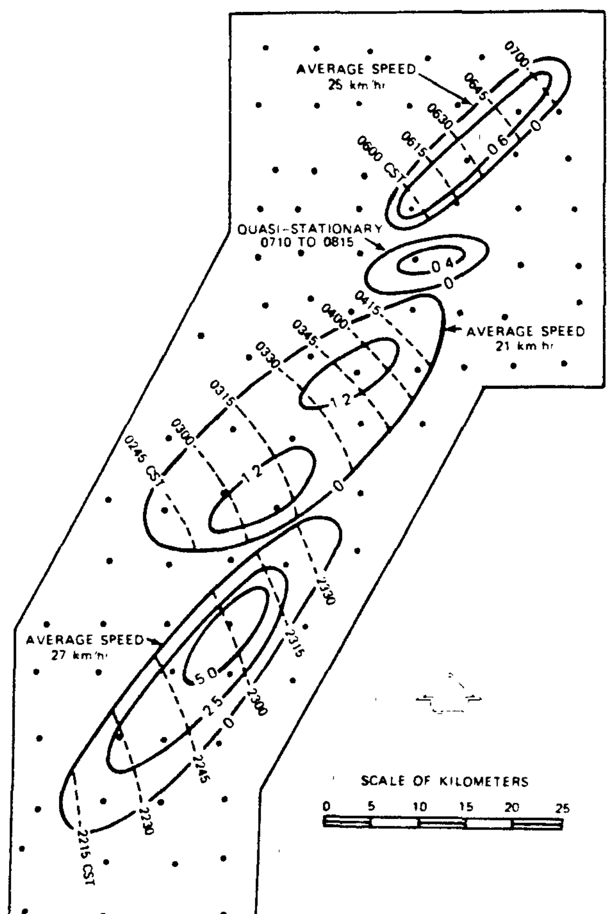


Fig. 6 Raincells during the rain period of 2–3 August 1965. Isochrones depict motion of the leading edge of the raincells.

collected at any one site is a poor indication of the regional amount of precipitation, or of the precipitation inputs for that event. (However, it has been recently found that separate thunderstorm systems can coalesce at night under some conditions, forming storms (Mesoscale Convective Complexes, MCCs) which cover large areas (~$2X10^5$ km^2), have high rainfall intensities, and long durations (Kerr 1985). To the extent that these MCCs occur, they would decrease the variability of rainfall between sites).

Thus, most atmospheric deposition projects use the results from rain sampling to calculate the weighted average concentration in precipitation in an area. Loading are then determined by multiplying this average concentration by the annual average rainfall amount, or by the annual amount as determined by a number of rain gauges distributed over the area of interest. Thus, the collection efficiency of samplers is less important then their having a reproducable efficiency. The amount of rain determined by the rain gauges are not needed for deposition calculations and, in fact, probably should not be used for them. The rain gauges are useful, however, to make a record of the onset, duration and intensity of precipitation events, and to serve as a monitor on the sampling efficiency of the collector.

Snow Sampling-Collectors

The commercially available wet-only rain samplers, described above, have been widely used for snow sampling. There are no reports of their snow sampling efficiency, but as described for snow gauges, they would be expected to do very poorly under windy conditions. While the principles and design of Nipher and Alter-type wind shields is reasonably well understood for cylindrical collectors, it is not clear how a shield might be designed for a rectangular collector.

Also, snow blown onto the precipitation sensor of wet-only collectors when it is not snowing, can cause the sampler to open. Snow can then be blown out of the collector. To prevent this from happening, the collector should have a height/diameter (H/D) ratio of 4 or greater. The collectors used in the commercially available wet-only samplers have a ratio of only about 1.

To alleviate these problems, the CAPMoN network (daily) in Canada uses a modified Sangamo collector with a 35 cm diameter opening and with a H/D ratio of ~2.5:1. Also, the APIOS-C network in Canada uses a standard Sangamo wet-only collector but uses a longer bucket in it during the winter (H/D = 4).

The precision of snow sample collection then is poor. However, since the loading calculations are based on concentration data, these collectors may be satisfactory. Also, if wind speed information is available from the site, the collection efficiency may be able to be estimated for each event and a correction applied.

Recently, snow collectors with heated collection surfaces have been designed. In these collectors, the snow which falls is melted and stored or extracted on site as a water sample. These heated snow collectors have two potentially serious problems. First, since snow flakes typically have only a small mass, it takes a while for the individual droplets of melted snow to coalesce sufficiently to run off the sloping collection surface. This is more of a problem on surfaces like Teflon which is not wetted by water. During this time they are on a warm surface (probably 15-35^{o} above ambient temperature) and can readily evaporate. The samples thus may show higher concentrations of non-volatile materials, loss of volatile materials, and lower amounts of water. A possible solution to this problem is to heat the collector to melt the snow only after the event is over and the roof is closed.

A second problem is the large heated area of the collector. It will cause convection currents above the collector, possibly decreasing the collection efficiency. Ironically, these induced currents may cause the most problem during period of low wind speeds, just the time when these collectors would otherwise have a reasonable collection efficiency.

Finally, all snow collection devices can "bridge" over under certain infrequent types of snow conditions. In addition, snow can fill-up the space between the outer shield and the inner cylinder in the Nipher-type collectors, leading to collection errors.

Snow Collection-Cores

An alternative method of measuring atmospheric inputs associated with snow, is to collect snow cores (see Barrie and Vet 1984, for instance). This method involves the collection of cores of snow after the snowfall, which are representative of the snow which fell. Alternatively, cores can be collected infrequently and the results will then serve as a measure of the bulk inputs (wet + dry + vapor inputs - evaporation) over that period of time. This method has most frequently been applied in areas where little thawing occurs during the winter and cores are collected every several weeks or at the end of the snow season. The sample then represents the net atmospheric inputs over that time period.

Since the first few percent of melt water carries away a major fraction of the soluble materials in the snow, the successful application of this method requires that no melting occurs between the snowfall event and the sample collection. However, an area could be prepared ahead of time to retain any melt water and serve as a collector. A sample obtained at the end of the snow season would then represent the net deposition during that season (or samples could be obtained at intervals and their inputs summed).

Since snow sublimes during the winter the snow pack at the end of the season is less than what fell. This leads to a concentration of the non-volatile components. Thus, the inputs that are calculated using this method will have to be based on loadings and NOT on concentrations.

If this method were applied to the frozen surface of the Great Lakes in protected areas where the ice forms early and remains throughout the winter, or areas adjacent to the frozen surface of the Lakes, a sample collected before breakup of the ice would be a direct measure of the net atmospheric inputs from the ice cover. This method gives information ONLY on the inputs to the snow surface, to the ice covered portion of the Lakes (an average of 19% of the lake surface of Lake Michigan during an average winter). Other methods would still be needed to determine the inputs to the open water portions of the Lakes. Inputs of snow to the open waters of the Lake would be expected to be higher than to the ice-covered portions since the open waters would also serve as a sink for blowing snow.

For snow collections then, an event snow collector with a deep bucket and an Alter-type shield should be used at most all of the GLAIS sites. In addition, the snow core method could be used in the north to obtain some information on bulk deposition to snow-covered surfaces.

DRY DEPOSITION

Dry deposition is defined as atmospheric inputs that occur when it is not raining (precipitating). It is thought to be an important input mechanism for many materials to bodies of water, but there are now no well qualified methods of determining these inputs (Slinn 1980, 1983). In addition, most investigations into determining dry deposition have been concerned with inputs to land and vegetative surfaces.

There are two reasons why these results may not be applicable to inputs to water surfaces. The first is the nature of water and its surface. The surface is a liquid and it is uniform. Since it is liquid, it will "wet" many materials and thus irreversibly capture them; the water at the surface will be continually renewed with new water from within the epilimnion; and its surface properties are quite different from those of solid surfaces.

Secondly, because of the high heat capacity of water, and the mixing which occurs in the epilimnion, the surface temperature will be quite constant on a diel basis, regardless of short-term energy gains and/or losses. Thus the temperature of the Lake surfaces, and its variability, are very different from the adjacent land surfaces during many months of the year. These differences will result in different dry deposition rates to the land and water surfaces.

With respect to determining dry deposition inputs, the results of several studies indicate that the dry deposition of small particles (<1 um) is not a significant contributor of atmospheric inputs (Talbot and Andren 1983; Davidson and Friedlander 1978). This is the case even for those materials, such as lead, that are present in the atmosphere chiefly on the small particles. Wet deposition is the principal atmospheric removal process for these materials, and what dry deposition occurs of these materials, seems to be due chiefly to the small percentage of these materials associated with large particles.

At least initially then, direct determination of small particle inputs need not be made. An estimate of their inputs could, however, be included in the atmospheric inputs total. Inputs of vapor are usually included as a component of dry deposition, but in this discussion, vapor inputs will be considered separately.

Most of the materials to be determined in a GLAIS network will probably be associated principally with small particles. It is interesting to note then that the only dry deposition sampling for them suggested here is that fraction associated with large particles!

The principal mechanisms by which large and small particles deposit from the atmosphere are different. The collection methods for the large and small particles are also usually different, but as mentioned above, all of the methods suffer from significant limitations and uncertainties. The better developed methods are briefly discussed below. Deposition collectors could be useful only for large particles (Wesely et al. 1985) micrometeorological methods could be useful only for small particles; and the deposition velocity method could be used to estimate both large and small particles. It is suggested that the Canadian-type deposition velocity method, as discussed below, be used initially in a GLAIS network to estimate dry deposition. It is also recommended that the NOAA/EPA/DOE method and the deposition bucket methods also be evaluated at the Master Station for consideration of their wider use.

Deposition Velocity

A commonly applied method of estimating dry deposition inputs is to assume that each different material in the atmosphere tends to "settle out" or "deposit" at a fixed rate, its deposition velocity, V_d. The deposition rate (mg/(m^2·s)) to a particular surface then is the concentration (mg/m^3) of that material in the atmosphere times its V_d (m/s).

An advantage of the deposition velocity method is that if air monitoring samples are being collected for the determination of sources, then those data would be available for the calculation of inputs based on deposition-velocity type calculations. However, information on the particle-size distribution for each material will be needed. Much information of this type is available for a variety of materials from other studies to serve as a guide.

To estimate atmospheric inputs by the V_d method, the air concentration and the V_d of each material of interest must be known. The limitation of the method is accurately estimating the value of the V_d. The difference is that the value of the V_d is a function of a number of variables. These include: the particle size distribution over which the material of interest is is present, the wind velocity, the nature of the particles, the atmospheric stability, and other factors. Thus while it is a single number, it is quite complex to calculate or measure, and to use (Williams 1982; Slinn 1982).

Also, a number of studies have demonstrated (Chamberlain 1976; see also Davidson et al. 1985) that the sampling efficiencies of high volume air samplers and cascade impactors decrease as the particle size increases. This results in significantly underestimating the number of particles larger than 10 um in the air, and the inputs associated with them.

There are additional complications in determining V_d's over the Great Lakes. These include: the stability of the atmosphere is often very different over the Lakes than over the adjacent land; the complications near shore of upwellings and downwellings in the Lakes on the local stability; changes in wind speed and direction as air moves from the land over one of the Lakes, or vice versa; the complications of lake and land breezes along the shores of the Lakes on many days of the year; and the effect of the sea state on particle deposition.

In general, the V_d for each material or compound of interest in the atmosphere will be different and it will vary for each material as the meteorology and the atmospheric compositions change. V_d's have been estimated in a variety of ways, most commonly using models. The better models attempt to take into account the variables mentioned above, but the uncertainties for most materials may still be as large as an order of magnitude (Sievering 1984).

There are some recent developments that should increase the accuracy and reliability of this method (Wesely et al. 1985; Rahn et al. (1984, 1985) report that in the northeastern U. S., air masses from the same region tend to have similar particle size distributions and particle-size associations of different materials. Air masses from different regions tend to be different. Thus it might be possible to make the effort determine the particle-size association of each material, for each different type of air mass for each sampling site. This information could then be used for all calculations on air masses from those regions.

Secondly, materials whose inputs to the Lakes are known to be predominantly atmospheric, such as lead, some of the radionuclides, and some organics, may be used to calibrate the deposition models. Such an effort has already attempted by Andren (unpublished) for Lake Michigan using lead. The calculations for Lake Michigan were based on information on lead deposition obtained from intensive studies on a small lake in northern Wis. that has no surface water inputs.

Two Canadian networks, CAPMoN and APIOS-D, estimate dry deposition inputs of sulfuric and nitric acids, and SO_2 using a V_d method. Daily samples are collected on a filter pack consisting of a particulate filter, a nylon filter to absorb nitric acid, and a carbonate-impregnated glass fiber filter to capture acidic sulfur compounds. The day's meteorology, along with a deposition model, are used to estimate the dry deposition of these materials.

Over-the-lake meteorological information (air (~2m) and water surface temperature, humidity, wind speed and direction, water current speed and direction, and wave heights) for all of the Great Lakes is available during the ice-free season from NOAA bouys in the Lakes. These data can be available in real time, and data from past years are available on tape or on microfiche. These data should be useful for determining what goes on in mid-lake during the time of year the bouys are present.

Deposition Collectors

Probably the most commonly used but also the most widely criticized depostion collection method is the use of dry deposition collectors. These are usually surfaces or containers that are exposed between precipitation events for a measured period of time, and the material that they accumulate is determined. It is widely believed that the shape of the collector (plate; low aspect bucket; high aspect bucket; etc.) affects the amount of material collected. Two recent reports address these questions (Dolske and Gatz 1985; Dasch 1985). The findings seem to be that teflon surfaces undercollect; that the height of the bucket is not an important variable (the sides are not acting as the principal collecting surface, or the collection of small particles is not important); that a bucket with water collects more material than does a dry bucket (one would want to use a wet collector for lake input studies); and finally that the errors involved in the use of bucket collectors is less than a factor of three, and probably less than a factor of two.

The reports on the collection of sulfate (Dolske and Gatz 1985; Feely et al. 1985) indicate that the deposition rate found by bucket collectors were higher than that found by other deposition measurement techniques, perhaps by a factor of 1.8 or so. It is to be expected that the other methods underestimate the deposition. Thus the buckets may over sample, but the error involved is not large. If it is known that the buckets may slightly over collect, this could be factored into the estimates produced using them. Since dry deposition is not going to be an important input route for many materials, being able to put an upper limit on it will be very useful.

The problem this method shares with all the other dry deposition collection methods is an unknown collection efficiency. However, if water is kept in the collector, the method is calibrated using ^{210}Pb, lake studies or other methods, and determination of the the small particle fraction is not attempted, the accuracy of this method may be as good or better than other dry deposition estimates.

An advantage of the method is that wet/dry samplers will be in use at all of the collection sites, but only the wet side will be in use. Certainly this method should be evaluated at the Master Station, and the results compared with those found using other methods.

Micrometeorological Methods

For small particles (<1 um) for which gravitational settling is not important, and which tend to move with the eddies in the air, a number of other methods have been tried to determine their transfer rates to surfaces. These include: eddy correlation, eddy accumulation, modified Bowen ratio, gradients, and varience (see Hicks et al. 1980 for a description and discussion).

Despite the optimism in the 1980 report, these methods continue to have several drawbacks. 1) They are still in the research stage or are not yet developed as field-tested operational equipment able to be used in a routine manner. 2) The fast-response sensors needed by some of the methods are not available for any materials on particles. 3) The calibration of these techniques is difficult under high wind and sea conditions with an unstable atmosphere, when the air to water transfer rates may be high.

A joint NOAA/EPA/DOE effort is developing a routine monitoring V_d method for dry deposition inputs of small particles. This method uses filter packs similar to those used in the Canadian networks, but determines V_d using micrometeorological methods. Thus only the deposition of small particles can be estimated, and the air sampler has a large particle "denuder" on the front to prevent these particles from being collected. The meteorological parameters determined are temperature, solar radiation, wind speed and the standard deviation of the wind direction (a measure of the turbulance and therefore of atmospheric stability), and humidity.

A weekly air sample is collected and the meteorological parameters are determined throughout the sampling period. An average V_d is calculated for the week from the meteorological data. This is multiplied by the average concentration of the materials of interest from the filter sample, to yield an estimate of the dry deposition inputs for that week.

Because of the large uncertainty associated with estimating the dry inputs of small particles, a direct measure of their contribution would be very useful to any GLAIS program. Thus it is recommended that this method be evaluated at the Master Station for possible inclusion into a GLAIS network. If air samples are already being collected for source determination, only the micrometeorological measuring system need be added.

VAPOR EXCHANGE

Compounds present in the atmosphere as gases, such as oxygen, dissolve to some extent in the Lakes. In the environment then, there is continuous exchange or transport of these compounds across the air/water interface. Each compound will dissolve or evaporate until the equilibrium condition for that compound is satisfied. At equilibrium the fugacity (escaping tendency) of the compound is the same in the air and water phases and, while the flux through the water surface may be quite large, no net transfer occurs. The equilibrium condition is determined by the relative tendency of the material to evaporate (vapor pressure) and dissolve (solubility). The distribution coefficient (Henry's Constant) is the ratio of the vapor pressure of the compound to its solubility.

Net transport from the air to water, or water to air will occur until equilibrium is attained. So, if the water is undersaturated with respect to the air, net transfer to the water will occur. This would constitute an atmospheric input. Conversely, if the air is undersaturated with respect to the water, net transfer to the atmosphere will occur. This would be a loss mechanism for the lake.

To determine which direction net transport is occurring, the fugacities of the material in the air and water phases, and the Henry's constant need to be known. The amount of material transported across the air/water interface is then calculated from the difference in fugacity between the two phases and a mass transfer coefficient for that compound under the prevailing conditions. The Henry's constants are known for some of the compounds of interest, but others need to be determined.

The fugacity determinations to date are usually complicated by the association of the compounds of interest with small particles in the air and water phases. For some compounds, this is only a small effect, for others, it may be significant. The use of denuders on air samplers allows the fugacities in the vapor phase to be determined, but suitable methods are not available for water samples. However, since the effect is probably small (<25%) for most compounds, a correction may be estimated and applied. The use of model or surrogate compound could be very useful in making these estimates.

A more significant problem is determining reasonable values for the mass transfer coefficient under all conditions over the Lakes. The problem in determining these values is that they are very dependent on the wind speed, temperature, sea state, the presence of a surface microlayer and other factors. Again, a lot of modeling work, wind tunnel studies and ambient measurements have been done which have improved the state of the art greatly in recent years, including: Liss 1982; Mackay and Yuen 1983; Tucker et al. 1983.

MACRO METHODS

The term macro here means methods that obtain information on atmopheric inputs indirectly by a method involving measurements on the Lakes or atmosphere. The information is usually obtained from measurements made at different times and/or different locations in the Lakes, with the input term being related to the differences found. In general, these methods measure a parameter that integrates the inputs over an annual or seasonal period. For some of the methods, one or a few GOOD measurements per year is all that is required (of course, great care and a lot of planning is necessary to obtain a few GOOD measurements).

The appeal of these methods often is that they serve as a direct measure of atmospheric inputs and they are independent of the effects of the winds, precipitation, etc. In an atmospheric inputs program, besides being useful in their own right, they could serve the most useful function of being an independent method of verifying the results obtained using other methods.

Surrogate Compounds

A tracer-compound method might also be useful to determine the inputs of a variety of organic compounds. The method requires identifing a compound whose only source to the Lakes is the atmosphere. Other compounds whose atmospheric distribution and deposition mechanisms are similar to the tracer compound, then would be expected to deposit in the same manner. Thus, their atmospheric inputs relative to the tracer compound, corrected for differences between the compounds, would be assumed to be proportional to their ratio to the tracer compound in the air. For compounds that partition between the air and water, this method could be useful in separating the net inputs from the total inputs.

For organic materials, care would be needed in choosing the tracer compounds. One of the necessary properties would be that the compound be resistent to degradation to permit accumulations in the water and sediments to be determined. It would be most useful if several compounds could be found with different properties. For instance, one with a low volatility could serve as a tracer for particulate-associated organics; one present in the vapor phase with a high Henry's Constant could help in determining washout by rain; etc.

Accumulation Methods

There are recent reports of methods that use the epilimnion of Lake Michigan as an integrating collector of atmospheric inputs. The idea is that during stratification, the epilimnion is cut-off from all direct inputs other than from the atmosphere. For those materials that do not undergo transformations, the change in the amounts of them per unit area over the period of measurement then is the atmospheric input loading rate (wet + dry + net vapor). Determining the accumulation over a period of time then would yield the net atmospheric inputs.

There are two reports of the application of this type method to the Great Lakes in the literature. Tisue (1984) observed that the profile of trace metals in the water column was uniform before stratification. At the end of the stratified season, the concentrations of the metals were higher near the surface. The amount of the metals needed to increase the concentration above that found at the thermocline, was assumed to have come from the atmosphere since stratification began (corrected for losses).

Chambers et al (1983) measured the concentrations of a number of substances in the epilimnion after stratification began and then at maximum stratification. They also deployed sediment traps toward the top of the thermocline during stratification. From the change in concentration of a material in the water during stratification, and the amount of that material found in the sediment trap, the inputs to the epilimnion for that period of time could be calculated (the contributions due to upwellings, diffusion, zooplankton grazing, and other possible gains and losses were not estimated or corrected for).

It is unfortunate that these methods probably will not work well during the winter when the stratification in the Lakes is weak, dry deposition is probably at its maximum rate, and snow is hard to collect. It should work best during the summer, but this is when the atmosphere is very stable and dry deposition may not be occurring.

An additional method might use "lakes within the Lakes". This would be an application and extension of the studies pioneered by Swain (1978) in his studies on atmospheric inputs to Siskiwit Lake on Isle Royale in Lake Superior. This method is an ingenious approach to solving the problem of designing a collector for atmospheric inputs that mimics a lake surface. It uses a lake!! The idea is that many islands in the Great Lakes contain lakes. Studies on these lakes, similar to the detailed loadings, budget and inventory studies conducted in recent years by Andren and co-workers on Crystal Lake in northern Wisconsin, and by Hites and co-workers on Siskiwit Lake could provide information directly applicable to atmospheric inputs to the Great Lakes.

A number of islands in the Great Lakes, including Isle Royale, Beaver, and Manitoulin contain a number of lakes, and a number of other islands contain one or more lakes. Some of these should have suitable exposure and hydrology to yield useful information on loadings and for the calibration of models. It is possible that some lakes on land, but close enough the Great Lakes to be within their climatological and meteorological regime, could also be useful for such studies.

SOURCE DETERMINATION

The program element to determine the sources to the atmosphere of the materials coming into the Great Lakes could consist of several components. The first should be to determine the direction from which the materials of concern are coming. The second could be to collect emission data in those regions found to be sources, for the different materials from point and non-point sources. A third component could be air monitoring in regions away from the lakes to help identify source regions for the materials of interest.

There are a number of initiatives in the U. S., Canada and Western Europe to determine the sources of acid forming materials in the atmosphere. Many of the methods developed by these programs should be applicable to a program to determine sources of materials to the Great Lakes.

Identification of Source Regions

In order to determine the source regions of materials being transported to the Great Lakes, short term sampling or intermittent sampling will be required. For instance, wet deposition will have to be collected on an event basis so that the materials and concentrations determined in the samples can be related to the appropriate region upwind.

If it is decided to obtain information on the source regions for the dry deposition material, then air samples should be collected such that each sample represents air from only one air mass. Back trajectories could then be determined to identify the region that was upwind of the each of the air samples.

One suggested sampling scheme (Stearns 1985) would be to have three air samplers, one to sample when the wind was out of a particular sector. The sectors are related to the synoptic weather patterns around the Great Lakes and would be: 0^{o} to 180^{o}; 180^{o} to 270^{o}; and 270^{o} to 360^{o}. Since most storms are out of the SW or NW, this scheme separates these two source regions from each other and from the rest of the regions. Which sampler was operated at a particular time, and the beginning and end of each sampling period, would be determined by the wind direction. The time and duration of each sampling event, and the wind speed and direction during each event would be recorded for each sample. At the end of a week, or month, or etc., analyses of the three samples would indicate the average air concentration for air masses from each sector for the sampling time period.

If such a sampling scheme were to be used, care would have to be taken in its design to minimize passive sampling by the samplers not turned on. In addition, the quality assurance program would have to be able to quantify the amount of passive sampling which did occur so that it could be corrected for.

Emission Data

The collection of emission data for different materials in regions adjacent and remote from the Lakes would need to be co-ordinated with other agencies (Rothblatt 1985). These data, in conjunction with the presence and amounts of the different materials found in the GLAIS sampling program, could help identify the sources of these materials to the atmosphere.

Regional Air Monitoring

If it is determined that some source regions seem to be significant contributors to the atmosphere of materials going into the Lakes, then efforts could be made to determine the sources of these materials. This information could be available from EPA Regional Air Quality Programs. It might also involve additional air sampling in the regions thought to be the sources.

IV. SITING

If samples of precipitation and/or air are collected to determine atmospheric inputs, it is important that the collectors be located such that useful samples are obtained. The general requirements are that a site should not be affected by local sources, it should be accessible to 120V electric power, it should have a dedicated and qualified operator, and it should be close to the lake. It is not necessarily easy to find sites in many areas of the Great Lakes where these requirements are all met. Areas remote from sources and urban areas tend to have few people and power lines, and the power is often not reliable, particularly in the winter. Open areas are sometimes hard to find in the forested northern areas, and secure, unobstructed sites are difficult to find in and close to urban areas.

Alternate power sources such as diesel or gasoline powered generators would remove the need for 120V electricity, but they emit organics and need regular attention. Solar cells are not practical in many areas adjacent to the Great Lakes in the winter due to the large number of days with dense cloud cover. They may, however, be useful in the summer on islands or other remote locations.

Climatology

People who live adjacent to the Great Lakes know, and the meteorologists and climatologists who have have studied the Lakes have demonstrated, that the Great Lakes affect the weather in the vicinity of the Lakes (Gatz and Changnon 1976; Eichenlaub 1979; Cole and Lyons 1972). There is also some good evidence that the presence of man in the Great Lakes basin effects the weather over the Lakes. The siting and operation of a GLAIS network must take into account the effects of the Lakes on the mesoscale climatology.

The effects of the Lakes are due primarily to their large heat capacities and thus their more constant temperatures. For instance, the surface temperatures of the open waters of Lake Superior range from $0^{o}C$ to about $15^{o}C$ over an annual cycle, while air temperatures range from about $-40^{o}C$ to $+35^{o}$ over the adjacent land areas. The result is that the open waters of the Lakes

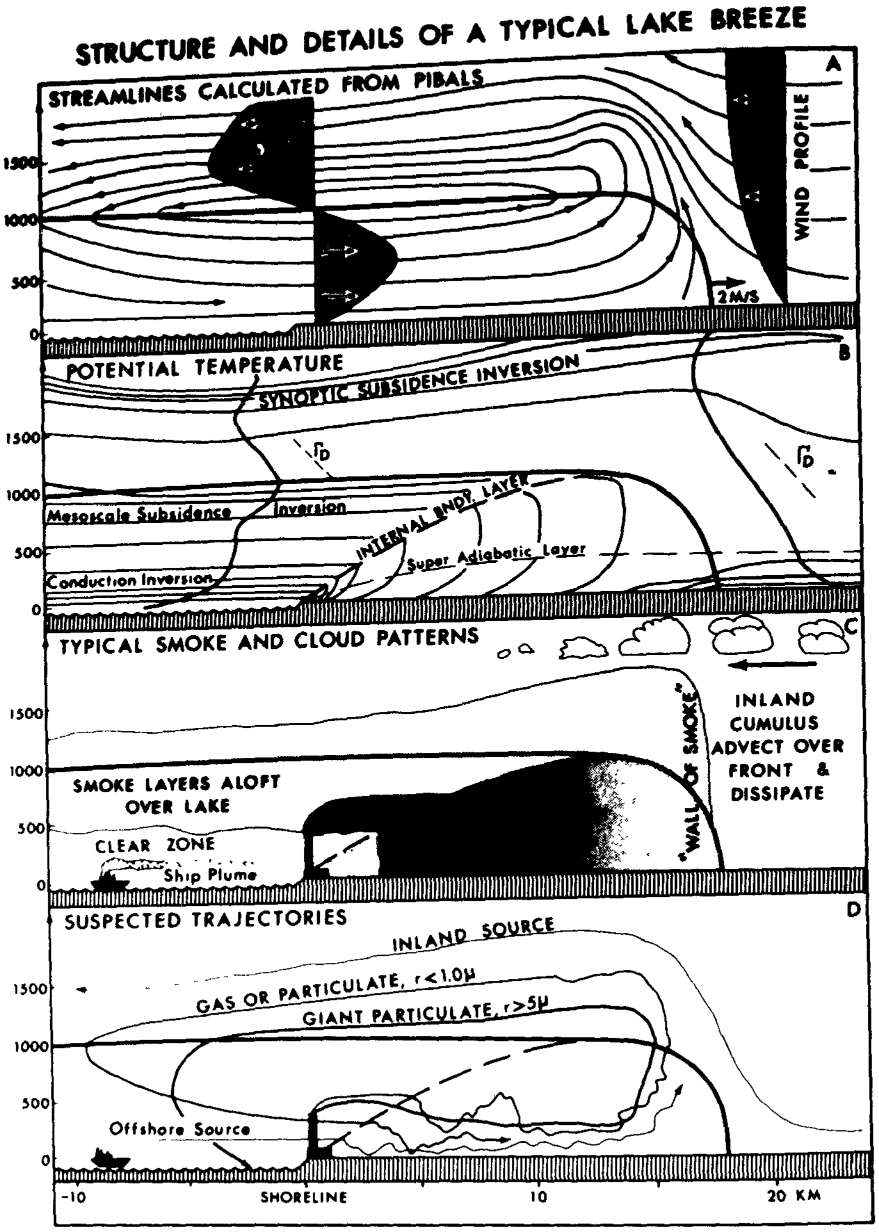


FIG. 8. Schematic representation of the cross-sectional structure of a lake breeze cell and its effect on the diffusion and transport of pollutants. Heights are given in meters. The heavy solid line found in each diagram represents the mesoscale lake breeze front (Lyons 1971b). a) Streamline patterns calculated from pibals illustrate inflow and return flow layers, the convergence zone updraft and subsidence over the lake. b) Thermal structure: light solid lines are constant potential temperature. Over the lake, stability is indicated by an increase in potential temperature with height. Over the land, the internal boundary layer is defined by values of potential temperature that decrease or are constant with height. Temperature soundings are indicated by the solid dark lines. The lake sounding and lines of constant potential show three inversions. Over the land, a deep adiabatic boundary layer is overlain by a synoptic scale subsidence inversion. c) Smoke and cloud patterns associated with typical lake breeze cell. Shown is a ship's plume that would advect shoreward with little vertical diffusion until it crosses the shoreline into the turbulent boundary layer. The plume from a tall stack, initially emitted into stable air fumigates when it is intercepted by the building turbulent boundary layer. The diagram also shows a "wall of smoke" in the convergence zone and concentration of smoke aloft in the bottom of the return flow layer and in the upper portion of the inflow. d) Estimated trajectories of particles with different sizes and sources of origin. The effect of size sorting is discussed in the text.

are warmer than the atmosphere in the fall and winter, and colder in the spring and summer. To the extent that the Lakes affect the air above them, they make the stability of the atmosphere over the Lakes different from adjacent land areas. Also, while the atmosphere is frequently stable over the land at night and unstable during the day, during some seasons there often is no diel change over the Lakes. This has a profound effect on the local meteorology, and is the principal factor which complicates siting collectors to determine inputs of materials from the atmosphere and their sources.

The warmer Lake surface in the fall and winter with respect to the air leads to unstable atmospheric conditions over the Lakes, to high evaporation rates, increased cloud formation and lake-effect snows, and probably to increased dry deposition rates. The colder lake surface in the spring and summer with respect to the air leads to stable atmospheric conditions above the lake, the formation of a stable inversion layer, decreased evaporation, cloud formation and precipitation, the formation of Lake-breeze conditions about 50% of the time, and probably to decreased dry deposition rates as compared to the adjacent land (see following figure). The effects of the lake climatology on wet and dry deposition rates need to be verified and quantified.

Snowfall along the shores of the Lakes is usually more frequent (lake-effect snows), but lower in amount than areas 30 km or more inland. If the shore snowfall reflects the snowfall over the Lakes, then the snow gauging must take place in the near-shore zone. The location of collection sites for snow is probably less critical than for rain, since most of the snow in the Great Lakes is associated with synoptic storm events and the materials being scavenged have typically come a long distance. Thus, local inputs or effects are probably less important.

Urban areas occupy a significant fraction of the Great Lakes shoreline, and their inputs to the Lakes thru the atmosphere were known to be significant (Gatz 1975). Also, Sievering et al. (undated) reported that the samples collected at the GLAD site on Beaver Island was significantly affected by the town of St. James. This GLAD sites was located at the Mi. DNR fire control station on Beaver Island, about 1.2 km east and across the bay from St. James. St. James has a low density, permanent population of about 200, with several times that many additional residents and visitors during July and August. If one of the objectives of a GLAIS network is to quantify the atmospheric inputs to the Great Lakes, then the contributions to these inputs from urban areas can not be ignored.

It will be difficult to sample these inputs representatively. It could involve siting samplers in the plumes from these areas, along the shore or on towers or islands offshore. Comparisons with inputs from unaffected regions should then permit the contribution from the urban areas to be estimated.

Over Water Measurements

Most of the atmospheric inputs sampling that has been done to date has been from the shores of the Lakes. The assumption has been that deposition along the shore is the same (within experimental limits) as the deposition into the Lakes. If shore sampling is to be a major component of an atmospherics inputs program, the errors in shore sampling will have to be quantified, and any necessary changes made in siting, sampling or interpretation.

Almost without exception, those who made recommendations concerning a GLAIS program, recommended an over the Lakes sampling component in order to understand the mesoscale meteorology of the Great Lakes with respect to atmospheric inputs to the Lakes. The most discussed problem was the ratio of the amount of precipitation that occurs over the Lakes compared to that which occurs at the shore. Several studies have been conducted on Lakes Michigan and Ontario to quantify these differences. These studies all concluded that there is more precipitation over the Lake during the winter, but less rain over the Lake during the summer than on shore. This is in accord with what would be expected based on air vs. water temperatures.

One interesting observation related to this question was made with the weather RADAR used in the IFYGL projects over Lake Ontario. On several occasions, stationary snow storms were observed over the Lake (Wilson 1977). On those occasions, the air was being scavenged over the Lake, but no observable or measurable precipitation was occurring on land.

However, in a careful consideration of all the studies on Lake Michigan, Bolsegna (1979) concluded that the differences found in land vs. lake precipiation were smaller than the errors associated with the precipitation gauges and the gauge networks. Thus it does not seem that a program to attempt to quantify differences in the on-shore vs. off-shore precipitation would be fruitful at this time.

However, inquiries into the effects of land- and lake-breeze cells on the samples collected by shore-based air and precipitation samplers; differences in wind speeds and direction over the Lakes compared to adjacent shore sites; differences in the atmospheric stability over the Lakes with respect to adjacent shore areas during the different seasons, and its variation over the Lakes due to winds, sunshine, ice cover, upwellings and downwellings; and the effect of urban areas on over-the-lake precipitation frequency, intensity and amount, need to be carried out. Observations over the Lakes to understand these effects could be made from islands, ships, bouys and towers. The people who made suggestions for a GLAIS design had conflicting views on ships, bouys and towers (Philbert 1985; Wesely et al. 1985).

Islands are probably ideal, IF they are small and in the right place, because they are rigid and permanent (Philbert et al. 1985). Specific considerations would be the size, ease of access, topography, habitation and vegetative cover on the island. Islands possibly of use include Isle Royale the Apostles and Caribou in Lake Superior; Beaver, N. & S. Fox, S. Manitou, St. Martin and Poverty in Lake Michigan; Great Duck, Charity and the North Channel islands in Lake Huron; South, Middle and North Bass, Pelee and Long Point in Lake Erie; and Galloo, Stony and the Thousand Islands in Lake Ontario. Since many of these islands are unoccupied, the equipment sited on the islands would have to be designed for remote operation. It could include recording rain gauges and meteorological instruments, as well as some samplers. Data could be recorded on site or telemetered to shore.

Ships could be useful, but probably only if they had a REGULAR and frequent schedule in the desired area. In Lake Michigan, the Ludington-Manitowoc and the Charlevoix-Beaver Island ferries could be useful. In Lake Superior the ferries to Isle Royale from Copper Harbor and Grand Portage, might be used. Research ships on the Great Lakes are attractive sampling platforms. Their advantages are that they are equipped for sample collections, could hold a variety of instruments and collectors, have technically qualified personnel on board and often spend significant periods of time in the open waters of the Lakes. Ships are perhaps the only sampling platform that will be available in deep-water areas of the Lakes. Finally, there is a lot of experience on the Great Lakes and the Oceans in collecting meteorological, air and precipitation samples from ships. This experience should be useful in designing useful projects within the limitations of ship-board sampling.

If information is needed in relatively shallow water, guyed towers may be the platform of choice. The advantage is that they are stable, can be built relatively tall and can hold a variety of instruments and collectors. A number of these have been, or are being, operated in the Great Lakes, and so there is already a body of experience with them. A problem with this and other sampling platforms in the lakes is that of insects and birds. During the warm months, insects and birds are attracted to any type of structure out in the lakes. Their presence could lead to sample contamination and to possible interference with the operation of some of the equipment.

NOAA presently operates 8 moored bouys (3 & 6 meter) in the upper four Great Lakes during the ice-free season. Also, they are testing a 40' diameter all-season bouy in Lake Superior during the winter of 1985-86. Many of these bouys have been operating for 4 to 8 years collecting air temperature and humidity, wind speed and direction, water temperature and sea state, and the wave spectrum. This data can be available from real-time telemetry (see Hartmann 1985) and on microfiche and magnetic tape. It could be a useful resource. Problems with the NOAA bouys are that they are designed to follow the waves, and the fact that instruments can not be placed very high above the water (effectively ruling out precipitation sampling). The motion of these bouys complicates the detemination of the wind speed and direction. An instrument package compatible with the present operations of the bouys could possibly be placed on the bouys for a minimal cost (W. Shepard, NOAA).

Locations of Sampling Sites

One of the more important decisions in an atmospheric sampling network is choosing the locations of the sampling sites. A number of the general factors involved were discussed above. A number of different criteria have to be met and each site should meet as many of them as is possible. Some of these criteria are: some sites should be in mid-lake; some sites should be upwind of the Lakes; some sites should be downwind of the Lakes; some sites should be in remote areas; some sites should be downwind of urban areas; all the different "regions" of the Great Lakes basin should be represented; and all of the sites should be well good sites. Easier said than done.

The data collected by the GLAD network should be useful in locating the GLAIS network sites. The data could indicate areas where the GLAD network was redundant and they could indicate that some sites were affected adversly by local sources. The GLAD locations that have been found to be excellent sites (siting criteria and data evaluation) should be given a high priority for inclusion into a GLAIS network. The GLAD sites that are in large open areas, away from major sources, have good access to power, and are adjacent or close to one of the lakes are Hovland, Cornucopia, Green Bay (perhaps too close to the town of Green Bay) and Hammond Bay. A summary of some of the siting information for the GLAD sites is shown in App. B. These data were collected during the summer of 1985.

There are other GLAD sites which do not meet all of the above criteria. But they have open areas adjacent to the present location of the samplers which could possibly be used. These include: Benton Harbor (urban), Mount Clemens (urban), Put-in-Bay (urban??), Bay City (urban), Conneaut and Olcott (POTW). In addition, the site at Empire is open, but is about 2.8 km inland behind a 400' dune; while the GLAD site at Milwaukee is on a building at a

Figure 8. Ecoregions of the Conterminous United States (after Bailey, 1976).

CAMP site, the water filtration plant at the shore has a large open field; Escanaba has a open site available at the end of a point, about one km east of the present site in town. New sites may have to be developed along the eastern end of Lake Superior; southwestern Lake Michigan; southern Lake Huron; western Lake Erie and western Lake Ontario.

Sites in the National Trends Network (NTN) were located based on bioregions as defined by Bailey (Robertson and Wilson 1985). Since climatology is the major differentiation between bioregions, these regions (Fig. 8) should also serve as a rational for locating GLAIS sites, at least on the upwind sides of the lake.

Finally, some idea of the number of sites, or the number of different types of sites will need to be known before specific sites can be chosen.

Locations of Collectors at a Site

A final important concern is how to locate samplers at a site. At the 36 GLAD sites, 18 of the Aerochem samplers were on roofs while the others were on the ground. For accuracy in collection, the ideal location for the opening of precipitation samplers is at ground level with some provision to prevent incorporation of splash and run-off (Kurtyka 1953). In practice of course, this is not practical and the standard rain gauge is 3' tall. But the higher a collector is placed above the ground, the more variable its collection efficiency is due to increased wind speeds. Putting a precipitation sampler on a roof also adds the problems caused by the increase in turbulance due to the building blocking the winds.

Roof sites can have several advantages over ground sites, however, for rain samplers. Often roof sites are less shielded by adjacent structures and vegetation; there are few problems with samplers in the middle of roofs due to bird droppings, and roof samplers are usually much more secure. If an accurate measure of precipitation amounts are not needed, roof locations may often be suitable sites for the samplers (with the gauges on the ground).

Roof sites are an advantage for air samplers as the collection efficiency for all but the larger particles is not affected, and the problems due to the incorporation of resuspended materials into the samples is minimized.

MASTER STATION

If a GLAIS network is planned which contains a variety of different types of samplers, it would probably be very beneficial to have one of the collection sites be a "master site" (Elder 1985). The site should provide a well-equipped location where development and research projects associated with the network could be carried out. A major purpose would be to perform all of the involved and detailed calibration and verification checks on the different sampling systems used in the network that are needed to verify their operation. Also, new types of samplers and collection and measurement methods could be evaluated and checked-out in the field before being used in the network. It would be a good place to site duplicate samplers, and samplers of new and old designs for inter-comparisons. This site would probably have a full-time operator who could also service other sampling sites. and it could serve a variety of purposes Finally, the operator could conduct a thorough and diverse set of quality assurance procedures on samples, the collectors and sampling procedures.

A master station would serve as a good place to co-locate samplers with the GLPN and other Canadian networks, and it might be advantagous to equip, certify and operate the site as a member of the NADP network.

A component of such a site could be a large (~5m^2) manually operated, wet-only precipitation sampler. The large samples collected from such a collector (50 l/cm ppt) would allow the quantification of materials which are normally below the detection limits for GLAIS samples. It would also serve as a source of large volumes of natural precipitation for quality control purposes.

If such a site was planned, finding a good location for it would be critical. The site should be away from the influence of point or diffuse sources; it probably should be downwide from one of the Great Lakes, on or close to the shore; the site should be reasonably flat for several hundred meters or so; it would be very useful to have one or more islands off-shore to obtain up-wind samples and meteorological information; and it should be reasonably accessible in all seasons. Unfortunately, most of the areas around the Great Lakes that could meet the above criteria are in Canada. One area which could meet the criteria would be the north eastern shore of Lake Michigan, somewhere east of the Manitou, Fox or Beaver Islands.

V. MATERIALS TO BE DETERMINED

At this point, it is not necessary that the specific materials and compounds to be determined are specified. It is important, however, to consider the types of materials which have different sampling requirements that may be determined. Sites equipped to measure these materials, should be able to also measure a wide variety of others. The materials to be sampled will probably fall into four categories, depending on the material required for the sampler and on the preservation techniques (if any). The four categories are: organics, metals, mercury, and nutrients.

The Great Lakes Environmental Administrators met in August 1984 and recommended that Polynuclear Aromatic Hydrocarbons, PCBs, Toxaphene, Chlorinated Dibenzodioxins, Chlorinated Dibenzofurans, routine Organochlorine Pesticides, Mercury, Cadmium and Lead be determined (Su 1984). They recommended using 10 sampling sites in the U. S. Some of those who made recommendations concerning GLAIS also suggested materials to be determined (Bidleman 1985; Eadie 1985).

Organics

Organic materials need to be collected and handled in equipment which contains no organic materials. Glass, metals and teflon are satisfactory collector materials. In addition, because of the small concentrations of many organic materials in precipitation, a sampler with a relatively large interception area is required to collect sufficient sample. If the organic collector to be used will process and discard the water on-site (adsorption or extraction), the portions of this unpreserved sample will not be available for other measurements (pH, conductivity, etc.).

Metals

Metals need to be collected and handled in equipment which contains no metals. Polyethylene or teflon are frequently used. Nitric acid is sometimes added to the sample collector to minimize loss of the metals to the collector surfaces. Many metals are present in air and precipiation at quite low levels. Thus, before efforts are made to collect a particular metal, analytical methods must be available with sufficient sensitivity to reliably quantify the metal at the levels expected.

Mercury

Mercury is present in the atmosphere chiefly in the elemental form which has little tendency to partition into water. However some is oxidized to an ionic form which quickly associates with particles and then can be removed from the air by wet or dry deposition. However, since all the oxidized and reduced forms of mercury are readily interconverted, a preservative (oxidant) must be added to precipitation samples to prevent the reduction and loss of the mercury in the collected sample. The concentrations of mercury in precipitation samples are usually high enough, with respect to analytical detection limits, that a collector with an interception area of .02 m^2 should be sufficient. Glass et al. (1985) have used beakers placed within an Aerochem #301 (.09 m^2) to collect samples. If such an arrangement is used, the collection containers should have straight sides to prevent splashing and to be able to precisely define the interception area. A wet-only collector need <u>not</u> be dedicated only to ionic mercury collection, as there should be sufficient space in the covered bucket for several such cylindrical collectors, only one of which needs to be for the ionic forms of mercury.

Nutrients

A preservative (usually sulfuric acid) is often added to precipitation samples for nutrient analyses to prevent loss of ammonia and slow the interconversion of the forms of phosphorus and nitrogen present. If event precipitation samples are collected, these collections may need a collector with the area of .09 m^2 or more to obtain sufficient sample. Otherwise, it could be possible to share space in the mercury sampler.

VI. ACKNOWLEDGMENTS

In writing this paper, I corresponded with, and/or had useful discussions with a number of people. They included Anders Andren, Len Barrie, Terry Bidleman, C. H. Chan, Walter Chan, Julian Chazin, Stanley Changnon, Jean Dasch, Brian Eadie, Cary Eaton, Steven Eisenreich, Floyd Elder, Peter Finkelstein, Donald Gatz, Gary Glass, Lennert Granat, Holly Hartmann, Bruce Hicks, Douglas Jones, Paul Kapinos, Barry Lesht, James Pankow, Francis Philbert, Stephen Rothblatt, Herman Sievering, Doug Sisterson, Gary Stensland, Charles Stearns, William Strachan, Malcolm Still, Tom Tisue, Robert Vet, Eva Voldner, Tony Wagner, Dave Warry, and Marvin Wesely. I thank all of them for the time and help they gave me.

VII. REFERENCES

Barrie, L. A. and R. J. Vet 1984. The Concentration and Deposition of Acidity, Major Ions and Trace Metals in the Snowpack of the Eastern Canadian Shield during the Winter of 1980-81. Atmos. Envir., 18, 1459-69.

Bidleman, T. F. 1985. Personnal communication.

Bolsenga, S. J. 1979. Determining overwater precipitation from overland data: the methodological controversy analyzed. J. Great Lakes Res., 5,301-11.

Chamberlain, A. C. 1976. Response to "Approaches to Evaluating Dry Deposition of Atmospehric Aerosols Pollutants onto lake Surfaces", by J. W. Winchester, J. Great Lakes Res., 2, Suppl. 1, 38-41.

Changnon, S. A. 1981. Convective Raincells. J. Atm. Sci., 38, 1793-97.

Cole, H. S. and Lyons, W. C. The Impact of the Great Lakes on the Air Quality of Urban Shoreline Areas: some practical applications with regard to air pollution control policy and environmental decision-making. Proc. 15th Conf. on Great Lakes Res., 436-63 (1972).

Davidson, C. I., S. E. Lindberg, J. A. Schmidt, L. G. Cartwright and L. A. Lands 1985. Dry Deposition of Sulfate onto Surrogate Surfaces. J. Geophy. Res., 90, D1, 2123-30.

Davidson, C. I. and S. K. Friedlander. 1978. A Filtration Model for Aerosol Dry Deposition: Application to Dry Deposition from the Atmosphere. J. Geophy. Res., 83, 2343-52.

Dasch, J. M. 1985 Direct Measurement of Dry Deposition to a Polyethylese Bucket and Various Surrogate Surfaces. Env. Sci. & Tech., 19, 721-25.

Dolske, D. A. and D. F. Gatz 1985 A Field Intercomparison of Methods for the Measurement of Particle and Gas Dry Deposition. J. Geophy. Res., 90, D1, 2076-84.

Eadie, B. J. 1985. Personnal communication.

Eadie, B. J., R. L. Chambers, W. S. Gardner and G. L. Bell 1984. Sediment Trap Studies in Lake Michigan: Resuspension and Chemical Fluxes in the Southern Basin. J. Great Lakes Res., 10, 307-21.

Eichenlaub, V. 1979. Weather and Climate of the Great Lakes Region. Univ. of Notre Dame Press.

Elder, F. C. 1985. Personnal communication.

Feely, H. W., D, C. Bogen, S. J. Nagourney and C. C. Torquato. 1985. Rates of Dry Deposition Determined using Wet/Dry Collectors. J. Geophy. Res., 90, D1, 2161-65.

Gatz, D. F. Pollutant Aerosol Deposition into Southern Lake Michigan. Water, Air and Soil Poll., 5, 239-51 (1975).

Gatz, D. F. and S. A. Changnon 1976. Atmospheric Environment of the Lake Michigan Drainage Basin. In: Environmental Status of the Lake Michigan Region. Argonne National Laboratory, ANL/ES-40 Vol. 8

Gertler, A. W., J. G. Watson and C. I. Lin 1985. Methods to Estimate Precision, Accuracy and Validity of Wet and Dry Deposition Measurements. Abstracts, NADP Tech. Comm. Meeting, Fort Collins, CO, Oct. 811.

Glass, G. E; Leonard, E. N.; Chan, W. H.; and Orr, D. B. Airborne Mercury in Precipitation in the Lake Superior Region. J. Great Lakes Res., In Press.

Graustein, W. C. and Turkekian, K. K. ^{210}Pb as a Tracer of the Deposition of Sub-Micron Aerosols. In: Precipitation Scavenging, Dry Deposition and Resuspension, Pruppacher, H. R.; Semonin, R. G.; and Slinn, W. G. N. Eds., Elsevier, 1983, p. 1315-24.

Goodison, B. E., H. L. Ferguson and G. A. Kay 1981 Snow Measurement and Data Analysis. In: Snow Handbook, D. M. Gray and D. H. Male Eds., Pergamon, pp 141-175.

Hicks, B. B. 1985. Results of the First Six Months of a Trial Dry Deposition Network Operation. Abstracts, NADP Tech. Comm. Meeting, Fort Collins, CO, Oct. 811.

Jones, D. M. A. 1969. Effect of Housing Shape on the Catch of Recording Gages. Monthly Weather Rev., 97, 604-6.

Kerr, R. A. 1985. Tracking a Stormy Beast in the Night. Sci., 229, 848-9.

Kurtyka, J. C. 1953. Precipitation Measurement Study. Ill. State Water Survey, Report of Investigation #20, 178 pp.

Larson, L. W. and E. L. Peck 1974. Accuracy of Precipitation Measurements for Hydrologic Modeling. Water Res. Res., 10, 857-63.

Liss, P. S. 1982 Gas Transfer: Experiments and Geochemical Implications. In: Air-Sea Exchange of Gases and Particles, Liss, P. S. and Slinn, W. G. N. Eds., Reidel, pp. 241-298.

Mackay, D. and A. T. K. Yuen 1983 Mass Transfer Coefficient Correlations for Volatilization of Organic Solutes from Water. Envir. Sci. and Tech., 17, 211-17.

Rahn, K. A. and Lowenthal, D. H. Pollution Aerosol in the Northwest: Northeastern-Midwestern Contributions. Science, 228, 275-284 (1985).

Rahn, K. A. and Lowenthal, D. H. Elementary Tracers of Distant Regional Pollution Aerosols. Science, 223, 132-39 (1984).

Robertson, J. K. and J. W. Wilson 1985. Design of the National Trends Network for Minitoring the Chemistry of Atmospheric Precipitation. U. S. Geological Survey Circular 964, 46 pp.

Rothblatt, S. 1985. Personnal communication.

Sievering, H. Small Particle Dry Deposition on Natural Waters: How large the Uncertainity? Atmos. Envir., 18, 2271-2 (1984)

Sievering, H.; Crawley, J.; Ton., N. Undated. Final Report for GLNPO Grant #R005697-01. GLAD Network Data Review and GLAD Bulk Sampler Considerations.

Slinn, W. G. N. Air-to-Sea Transfer of Particles. 1982 In: Air-Sea Exchange of Gases and Particles, Liss, P. S. and Slinn, W. G. N. Eds., Reidel, pp. 299-405.

Slinn, W. G. N. 1983. A Potpourri of Deposition and Resuspension Questions. In: Precipitation Scavenging, Dry Deposition and Resuspension, Pruppacher, H. R.; Semonin, R. G.; and Slinn, W. G. N. Eds., Elsevier, 1983, p. 1361-1416.

Slinn, S. A. and W. G. N. Slinn (1980). Predictions for Particle Deposition on Natural Waters. Atmos. Envir. 14, 1013-1016.

Smith, J. H., D. C. Bomberger, Jr., and D. L. Haynes (1980) Prediction of the Volatilization Rates of High-Volatility Chemicals from Natural Water Bodies. Envir. Sci. Technol., 14, 1332-1337.

Stall, J. B. and F. A. Huff 1971. The Structure of Thunderstorm Rainfall. Meeting Preprt., ASCE Nat. Water Res. Eng. Meet., Phoenix, AZ, Jan 1115. 30 pp.

Stearns, C. R. 1985. Personnal communication.

Su, G. 1984. A Proposal for a Program to Study Atmospheric Loading of Toxic Chemicals to the Great Lakes. Report to Great Lakes Environmental Administrators. August. 6 pp.

Swain, W. R. (1978). Chlorinated Organic Residues in fish, water and precipitation from the vicinity of Isle Royale, Lake Superior. J. of Great Lakes Research, 4, 398-407.

Talbot, R. W. and A. W. Andren. 1983. Relationship Between Pb and ^{210}Pb in Aerosol and Precipitation at a Semiremote Site in Northern Wisconsin. J. Geophy. Res., 88, C11, 6752-60.

Tisue, T. and D. Fingleton 1983. Atmospheric Inputs and the Dynamics of Trace Elements in Lake Michigan. In: M. Simmons and J. Nriagu, Eds., Toxic Contaminants in the Great Lakes, Adv. Envir. Sci. and Tech. J. Wiley and Sons. P. 105-126.

Tucker, W. A., Lyman, W. J. and Preston, A. L. 1983. Estimation of the Dry Deposition Velocity and Scavenging Ratio for Organic Chemicals. In: Precipitation Scavenging, Dry Deposition and Resuspension, Pruppacher, H. R.; Semonin, R. G.; and Slinn, W. G. N. Eds., Elsevier, 1983, p. 1243-57.

Wesely, M. L., B. M. Lesht and D. L. Sisterson. 1985. Personnal communication.

Williams, R. M. 1982. A Model for the Dry Deposition of Particles to Natural Water Surfaces. Atmos. Envir., 16, 1933-38.

Wilson, J. W. 1977. Effect of Lake Ontario on precipitation. Mon. Weather Rev., 105, 207-214.

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA-905/4-87-001	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE Design of a Great Lakes Atmospheric Inputs and Sources (GLAIS) Network		5. REPORT DATE March 1987
		6. PERFORMING ORGANIZATION CODE 5GL
7. AUTHOR(S) Thomas J. Murphy - DePaul University		8. PERFORMING ORGANIZATION REPORT NO. GLNPO No. 87-03
9. PERFORMING ORGANIZATION NAME AND ADDRESS DePaul University 2323 North Seminary Avenue Chicago, Illinois 60614		10. PROGRAM ELEMENT NO.
		11. CONTRACT/GRANT NO. R005818-01
12. SPONSORING AGENCY NAME AND ADDRESS U.S. Environmental Protection Agency Great Lakes National Program Office 230 South Dearborn Street Chicago, Illinois 60604		13. TYPE OF REPORT AND PERIOD COVERED GLAIS 1985
		14. SPONSORING AGENCY CODE Great Lakes National Program Office, USEPA, Region V

15. SUPPLEMENTARY NOTES

Edward Klappenbach
Project Officer

16. ABSTRACT

This report is a document to aid in the design of a network aimed at determining the identity, amounts, and sources of the materials coming into the Great Lakes from the atmosphere. It is meant to discuss what is known and what needs to be known about sampling atmospheric inputs to lakes. It does not strive to be thorough and comprehensive as this is not possible for such a broad subject in a short report. It is based on a variety of articles and reports, and relies heavily on suggestions and discussions with a variety of investigators in different disciplines. It assumes that experts in each of the necessary disciplines will be consulted or will participate in the final network design. Finally, it reflects the strengths, weaknesses, biases and misspellings of the author. Discussion among others is needed to fill it in and to round it off.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Atmospheric Inputs Rain Gauges Snow Gauges Wet/Dry Samplers Air Monitoring Dry Deposition		

18. DISTRIBUTION STATEMENT Document is available to public through the National Technical Information Service (NTIS) Springfield, VA 22161	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES 44
	20. SECURITY CLASS *(This page)*	22. PRICE

EPA Form 2220-1 (9-73)