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## Acid Rain From Source To Receptor

by *Dr. Hans Martin\**

My topic today will cover three parts of the problem of acid rain: emissions, transportation oblique transformation of pollutants and deposition. I will carry the discussion to the point where the pollutants strike the ground but I will not include ecological effects. That's the point where the hysteria starts to take hold and the horror stories begin to appear.

How did this environmental issue come about? About 40 or 50 years ago, we had pollution problems which were associated with short stacks. Often when the materials were emitted they were in dangerous concentrations near ground level and we took steps to clean up the situation. In the case of point sources which had very high concentrations, we raised the stack height, as in the case of a much quoted 381 meter unidentified source near Sudbury, Ontario. At the same time as we were taking these steps to emit the material from large point sources at greater heights, our cities were growing and the accumulation of thousands of small sources led to the development of a situation where whole air masses were being altered chemically.

We have entered the era of long-range transport of air pollutants (LRTAP), through the injections of high concentrations at higher levels in the atmosphere and the modification of whole air masses which pass over urban or industrialized regions. In both cases these materials can travel for hundreds of miles and still be identified.

The foregoing gives a brief distinction between what might be called a local pollution problem and a large scale pollution problem. In the first instance the sources are small individual factories or power plants, whereas on the large scale the sources include whole cities and industrial areas which produce moving and identifiable air masses. In the first case the effected area may extend 100 kilometers while in the latter hundreds to thousands of kilometers are involved. The primary local pollutants are SO<sub>2</sub> and nitrogen oxides in high concentrations. As they move away, the concentrations drop and through transformation, secondary pollutants are formed, which are the precursors to acid rain. The effects of high local concentrations are often immediate, particularly with regard to health implications. On the other hand in the case of longrange transport we are dealing with vast areas which are affected by concentrations which are sometimes below detection limits, but which are being deposited continuously day and night, year in and year out. The local problems have been

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controlled quite effectively. However, such action has not been undertaken with regard to long-range transport and hence, that situation is deteriorating.

Acid rain is only a part of a larger problem which concerns the transport of all materials over long distances and their deposition in wet and dry forms. The problem is three-fold: first, large single point or numerous regional sources; second, a meteorology which transports this material over the continent; and third, sensitive sites where it may be deposited and may lead to undesirable chemical and biological changes and thus ecological damage. I will discuss the first two parts of this problem: the sources; and the transport of these materials through the atmosphere.

In eastern North America we certainly do have the sources, over 30 million metric tons of sulphur dioxide are emitted each year. This figure can be compared to world emissions of perhaps 130 million metric tons. We focus frequently on sulphur as the polluting material because we understand it well. We have learned much from the European experience. In addition, we consider sulphur oxides to be among the primary products or components causing the damage.

Consider the distribution of these materials. It is important to know where they originate, as will be described in due course. Fig. 1 gives the annual emissions for sulphur dioxide for eastern North America. All of the contributions add up to 30 million metric tons. These data are simplifications of detailed data sets maintained in national archives of emission sources for both the United States and Canada. Typically, for eastern North America the data bank provides information about perhaps 10 to 20 thousand identified sulphur dioxide point sources which includes the amount of material coming out of each one per year, their locations and the exit temperatures of the emissions. This information serves as the cornerstone for the following studies.

In Canada we have current emissions inventories for sulphur dioxide, nitrogen oxides, mercury, organics and fine particulates. In addition we are working on historical data for the year 1955 in order to see how this situation has changed. At the same time we are developing inventories for the year 2000, which will provide a prognosis for our future direction. Therefore, we have a fairly good overall idea of what is being emitted and where. These data are perhaps the best available information. Once a pollutant enters the atmosphere, however, our confidence in our understanding begins to erode.

The second component of the process leading to deposition is meteorology. The average flow of surface winds in North America in the summer is from the "main" emission regions in the eastern United States northward and eastward over the Atlantic. While the various pollutants are in the atmosphere, they can undergo extremely complex and numerous chemical changes. About a year ago one of our colleagues from Norway was discussing what he called a "first approach," simple model for describing what happens in the atmosphere. His simple model contained

40 simultaneous chemical equations.

We can look at the atmospheric processes in the laboratory where we try to duplicate the real world. This controlled examination of the reactions, the products and the reaction rates is not entirely satisfactory. It is not *in situ*. It is good guidance as to what might be happening in the atmosphere, but it has shortcomings. These controlled studies do not take into consideration all of the active components and synergistic processes: We have to do measurements using instrumented aircraft to follow these pollutants as they traverse the atmosphere. The changes in constituents and the reaction rates can be examined with our mobile laboratories; it is then possible to obtain a fairly good idea of what is actually happening.

This past summer, Canada and the United States undertook a major study of this type in which we focused on persistent elevated "pollution" episodes (PEPE's). For six weeks with the use of four or five aircraft we identified pollution episodes in the northeastern United States and southeastern Canada and then, followed these episodes as they moved eastward and northward over the Atlantic. Satellite imagery has shown hazy areas covering hundreds of square miles developing over days and then moving slowly from eastern North America, visible across the Atlantic for thousands of kilometers. These hazy air masses are in fact polluted air masses and they show us clearly where these materials originate, enter the atmosphere and then move identifiably far into the Atlantic. In the United Kingdom scientists are about to undertake a similar experiment jointly with the United States in which they will fly an instrumented Hercules. The purpose of the study, operated by the Central Electricity Generating Board, is to show that the Board does not cause acid rain or at worst that it makes a negligible contribution to acid rain. This position, however, is not now accepted in Scandinavia and I think the Board will face some surprising results.

Another way of examining or predicting what is happening in the atmosphere is through the use of models. The use of models perhaps sounds like a very esoteric exercise, but it is far less elusive than you might imagine. Consider the following thought process. If my wife stops working tomorrow, we stop running two cars and cut down to one, we go to Colorado for our holidays instead of Spain, then, we will start accumulating some money and we can consider buying a house in some price range, say less than \$150,000. Let's assume we can get a mortgage for 14 percent or less and we can purchase it with a down payment of about \$35,000. If I have a salary increase of 12 percent a year, in five years we will be in fairly good shape financially. That's a model. It includes rational judgments, real data and, in fact, it is an experience expressed in mathematical terms.

The process of developing atmospheric models also includes judgments and data. Twenty thousand point sources with their emissions per year, their temperatures and their heights are inserted into a computer along with their locations and, to those figures are added the regional source estimates, such as the production of sulphur compounds from

home heating in a particular suburb. Then we add three levels of wind speed measured at 300 meteorological stations every six hours for one year.

Judgments are made on the best estimates of the transformation rates of these materials in the atmosphere, of precipitation probability terms, and of dry deposition estimates. All of these data in a very large computer produce a model which can provide certain estimates. Often source-receptor relationships are obtained; with our data set, our mathematics and our chemistry we receive for each region on a predetermined grid, an estimate of the contribution to the deposition in that region by every other region in our grid. This analysis is a very powerful tool.

The machinery and the data sets are available. In fact, under the Memorandum of Intent which was mentioned earlier there are five models which are producing source-receptor relationships in a form called transformation matrices. It is possible to insert into the system ground validation information to see if what the model predicts is, in fact, happening. Clearly preliminary validations show that the models are heading in the right direction. This development is rather encouraging. The intercomparison "between" models also gives encouraging results; the outputs appear to "converge" on a point of agreement. We will see great progress in this area of study in the next 12 months. Model results are of particular interest to the legal community for, they provide estimates of the transboundary flux, which is the movement of materials across a boundary or from one region or jurisdiction into another region within the same country.

We turn now to the processes of atmospheric deposition. Here we reach a "ground truthing" point to validate or test our speculation on what we think has been happening during the residence time of pollutants in the atmosphere. At this point there is a more complete understanding since actual precipitation samples have been collected and analyzed. In Canada there are 55 stations across the country and each station has a specialized automatic collector.

All samples are sent once a month to a central laboratory for an analysis of approximately 15 components. One of them is the acidity in terms of pH. From that information, maps similar to the one shown in Fig. 2 are developed. Areas with pH less than about 5.5, a value which usually is assumed to be clean normal rain, are acidic. Acidity increases as the numbers become lower. At 4.5, for example, precipitation is 10 times as acidic as normal rain. Continuously, throughout the year along the St. Lawrence Valley the pH is about 4.2, that is 30 or 40 times more acidic than clean rain. The Canadian data sets are linked with those emerging from the networks in the United States to produce a continental picture.

There is a weakness in the assessment of deposition. The weakness concerns our inability, for technical and engineering reasons, to determine what is falling when it isn't raining. In fact, it is estimated that about 40 percent of all the materials striking the ground occurs in dry form, either

as gases or particulates. There are no reliable, operational instruments available to establish networks to estimate this dry deposition.

However, at some selected experimental sites estimates are being made of both wet and dry deposition. At these sites, rain collectors, similar to those in the national network, are operated on a daily basis. Thus, it is possible to obtain some information on the chemistry of specific storm events. In addition, at these sites a special set of filter papers is mounted 10 meters above the ground. Each day a new filter is exposed to the atmosphere. The amount of material collected on the filter each day is an estimate of the potential for deposition at that site based on the concentration of material in the air. This estimate is not precisely the dry deposition, but it gives an indication of potential.

In February 1979, the sulphate level (a precursor to "sulphuric acid in rain") at our experimental site near Kenora was at a low, near background level until February 19th. On the 19th, the level rose very rapidly to many times the background concentration. From analysis of air mass trajectories it was determined that the air arriving at the Kenora site had passed over industrialized areas of the mid-western United States. Air during the days prior to this episode had its origin in northwestern Canada. Two days later the same episode was observed at a duplicate experimental site at Chalk River to the east.

Similar studies have been undertaken at the experimental site in central Nova Scotia where high concentrations of pollutants in the air have been associated with air mass trajectories from the mid-western industrialized regions in this continent. At this point in the discussion of the acidic precipitation problem it is necessary to examine the impacts of pollutants once they have been delivered to the aquatic and terrestrial receptors. These ecological effects will be discussed by later speakers.

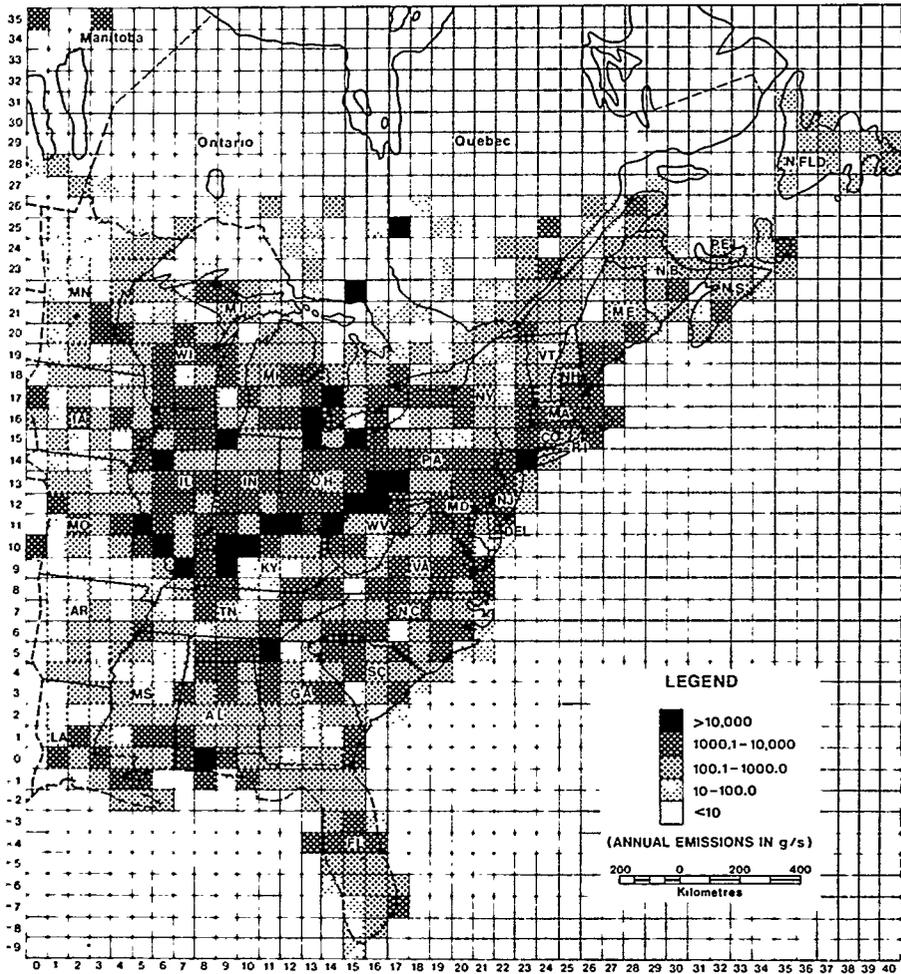
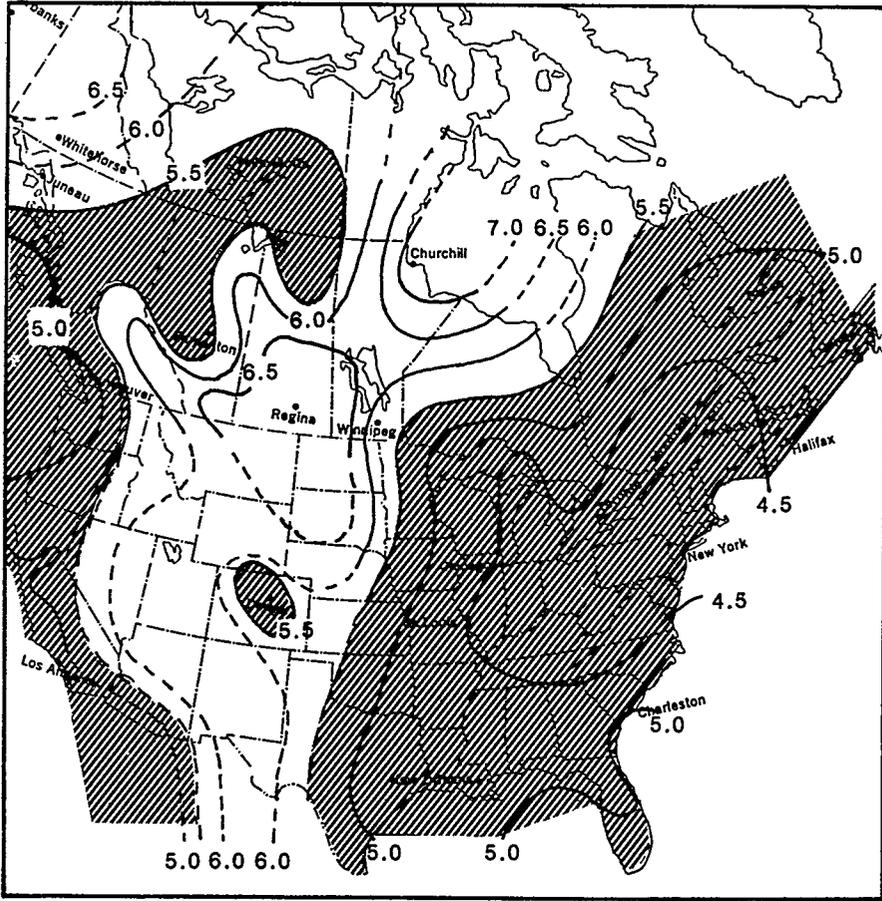


FIGURE 1. MAGNITUDE AND DISTRIBUTION OF SULPHUR DIOXIDE (SO<sub>2</sub>) EMISSIONS IN EASTERN NORTH AMERICA

Source: U.S.A. emissions from SURE II data base.  
Canadian emissions from Environment Canada.



----- Dashed lines indicate where data are sparse and thus only the general pattern is indicated

 Region with precipitation more acid than the normal "clean" rain pH <5.6

Figure 2 Map showing the weighted mean pH of precipitation for the period 1976 - 1979 based on the CANSAP network in Canada and a composite of 10 networks in the USA (Wisniewski and Keitz, 1980)