ARCTIC HAZE: NATURAL OR POLLUTION?

A Progress Report and Renewal Proposal to the
Office of Naval Research
Arctic Program

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The overall chemical composition of the Arctic aerosol was better defined; sulfur and carbon dominate. Different Mn/V ratios on different sides of the Arctic were found; both were inconsistent with eastern North American source. ^{222}Rn data for Barrow were produced. They indicated a source to the north. Arguments were developed for a Eurasian source and against a North American source of Arctic aerosol. Chemical and physical aspects of long-range transport of polluted air masses to the Arctic were revealed by numerical simulation

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The cloud-active fraction of the aerosol of Iceland was seen to parallel pollution-derived SO_4 and V. Heating by Arctic aerosol in spring was calculated. Deposition of aerosol to the Arctic Ocean was estimated. High Br in North American Arctic in spring was noted--neither source nor effects is yet known. Trace-element composition of desert soils was found to be nearly constant within the aerosol-size range and from desert to desert, but highly variable for larger particles. Particle-size distributions of Fairbanks aerosol were estimated by inverting optical data. Cloudiness in the Arctic was found to be generally unchanged since 1920. Haze was reported in 114 of 3274 Ptarmigan flights, with greatest frequency in March-May.

Statement of Submission

The following progress report and renewal proposal, "Arctic Haze: Natural or Pollution?", is hereby submitted to the Office of Naval Research, Arctic Program, for consideration as an extension of Contract N00014-76-C-0435, of the same title. This document is a joint product of the University of Rhode Island (URI) and the University of Alaska (UA), although it is being submitted by URI. As in the past, URI will be the prime contractor and will subcontract UA's portion. The approval sheets from UA will follow in 2-3 weeks.

This proposal is not being submitted to any other agency for financial support, although certain costs are to be shared with other grants, from NSF.

ABSTRACT

A three-year program of continued research into chemical, physical, optical and meteorological aspects of Arctic haze is planned. Primary areas of interest for 1980-83 include further delineating the spatial and temporal distributions of the Arctic aerosol, deducing its source areas from its chemical composition, understanding the major meteorological aspects of its transport from midlatitudes to the Arctic, modeling the large-scale chemical and physical changes of the polluted air masses during this transport, correlating chemical and optical measurements, investigating historical and climatological aspects of Arctic haze, and better determining the radiative, nucleational and depositional aspects of the Arctic aerosol. Routine sampling of the surface aerosol at Barrow, Poker Flat, Iceland, Narragansett (RI), and Paris will continue. A year-long sampling program in western Ireland will be conducted. A station will be established at Nord, Greenland. In addition, samples from the Norwegian stations of Spitsbergen and Bear Island will continue to be received. Daily aerosol samples will be taken at Barrow during winter. Samples will be analyzed for trace elements by neutron activation and atomic absorption, and for $SO_4^=$ and ^{210}Pb as needed. Routine optical measurements will be taken at Fairbanks. Field campaigns at various Arctic sites will provide supplementary data. Routine optical data from a variety of Arctic Air-Sampling Network sites will come after 10 sun photometers are distributed. Particle-size distributions of the Arctic aerosol will be inferred by inverting optical measurements. Vertical and horizontal profiles of the aerosol from Fairbanks to the Arctic Ocean will come from an aircraft-sampling program during winter 1980-81. Particle dynamics of the Arctic aerosol will be studied. The project on cloud-active aerosol in the Arctic will be concluded. Most of the study of climatology of Arctic haze will be completed. Our chemical and optical data on the Arctic aerosol will be systematized and made available to other researchers. Study of meteorological aspects of Arctic haze and its transport will continue, in large part through cooperative programs. Global sources of atmospheric Mn will be surveyed. If time permits, the puzzling behavior of Br in the North American Arctic will be examined further. A series of cooperative programs with other institutions will be continued, most at no cost. We will continue to support and help coordinate the Arctic Air-Sampling Network. If possible, a chemical-element balance approach to determining sources of the Arctic aerosol will be begun. The Third Symposium on Arctic Air Chemistry will be held in May 1983.

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I. Progress Report: August 1978 - July 1980

   A. Highlights of activities and results

      1. Activities

         • Aerosol sampling at Barrow, Fairbanks and Rhode Island
           continued. The New York station was stopped in October
           1979. An Iceland station was started in August 1979; a
           Paris station started in October 1979. An Ireland station
           is slated to begin in 1980. A second sampler and expanded
           control system was installed at Barrow. The Fairbanks
           sampling apparatus was moved from Ester Dome to Poker Flat.

         • Analysis of Barrow samples by atomic absorption is to begin
           during fall 1980.

         • Daily filter samples were taken at Barrow, December 1979 -
           May 1980. All winter-spring samples were gray.

         • Tests at Narwahl Island north of Prudhoe Bay showed the
           same aerosol as at Barrow; this confirms regional nature
           of the Barrow samples.

         • Cooperative projects on organics in the Arctic aerosol,
           darkness of filter samples, Rn in the Arctic, sampling in
           Greenland, and halocarbon gases in the Arctic atmosphere
           all helped in understanding sources, transport and charac-
           teristics of the Arctic aerosol.

         • All approaches to USSR failed; they will be continued.

         • Field trips for study of cloud-active aerosol in the Arctic
           (Borys, CSU) were successful and are nearly completed.

         • Work on trace elements in desert soils (Schütz, Mainz) is
           nearly complete; the first publication will be in fall 1980.

         • Optical measurements of aerosol at Fairbanks and Barrow
           continued. New data were taken from the Norwegian Arctic.

         • Eddy-diffusion transport model for Arctic aerosol was
           proposed.

         • Development of improved computer program for calculating
           direct radiative effects of Arctic aerosol was begun.

         • Techniques for evaluating the flux of material through
           particle spectrum of Arctic aerosol were evaluated.

         • Instruments were constructed to measure the particle-
           size distribution of Arctic aerosol for $r < 1\mu m$.

         • Study of climatology of Arctic aerosol (Raatz, UA) began.

         • KAR spent 3 months at NILU during 1979.

         • Second Symposium on Arctic Air Chemistry at URI, May 1980,
           was attended by 46 from 6 countries.

         • T. Conway, R. Borys will participate in YMER-80 expedition,
           summer 1980.

         • KAR held three briefings at U.S. State Department, 1978-80.

2. <u>Results</u>

- Overall chemical composition of the Arctic aerosol was better defined; sulfur and carbon dominate.

- Different Mn/V ratios on different sides of the Arctic appeared; this may show the influence of an unrecognized source. The Mn/V ratio on both sides of the Arctic is inconsistent with eastern North American source.

- Complexity of seasonal variations increased; variations by location, element and year began to appear.

- First ^{222}Rn data for Barrow was produced. Concentration was highest in northern Alaska; seemed to indicate a northward origin.

- Many arguments were developed for a Eurasian source and against a North American or an eastern Asian source of Arctic aerosol. Debate on sources continued.

- Broad chemical and physical aspects of long-range transport of polluted air masses to the Arctic were revealed by simple numerical simulation of atmospheric aging. Great stability and inertness of Arctic atmosphere during winter was indicated.

- Cloud-active fraction of the aerosol of Iceland was seen to parallel pollution-derived $SO_4^=$ and V.

- Small but significant heating by Arctic aerosol in spring was calculated.

- Deposition of aerosol to the Arctic Ocean was estimated. It seems to be greatest in winter and affected by pollution, but less so than is the aerosol itself.

- Problem of high Br in North American Arctic in spring was identified—neither source nor effects is yet known.

- Trace-element composition of desert soils was found to be nearly constant within the aerosol-size range and from desert to desert, but highly variable for larger particles. This supports the use of enrichment factors, but augurs against finding unique trace-element signatures of individual deserts.

- First estimates of particle-size distribution of Fairbanks aerosol were obtained by inverting optical data.

- Cloudiness in the Arctic was found to be generally unchanged since 1920, except for a slight increase in Alaska in the early 1940's. Pollution aerosol in Arctic does not affect the overall cloudiness.

- Haze was reported in 114 of 3274 Ptarmigan flights, with greatest frequency in March-May.

B. Activities

1. Routine sampling of Arctic surface aerosol (URI,UA)

During FY79-80 we continued to take routine, continuous aerosol samples at a number of Arctic and Arctic-related sites. Chemical analysis of these samples and interpretation of the resulting data make up one of the cornerstones of our Arctic Haze project, for we believe that the Arctic aerosol and its environmental effects will be properly understood only after several years of data have been accumulated. With the current expansion of the Arctic Air-Sampling Network, we anticipate continuing this type of sampling for some years more.

We would like to note that "routine" is perhaps a misnomer. Sampling of Arctic aerosol is not really routine--it requires specialized equipment, careful attention by the operator, and frequent communication with the home base. Our analysis uses sophisticated techniques, and is only routine in the sense that these techniques have been proven over the last 5-10 years. Lastly, our interpretation of the data is distinctly nonroutine. We must consider both natural and pollution sources, from continents and oceans, and often try to distinguish one continent from another. Superimposed on all this is large-scale meteorology, which is necessary to account for the large seasonal variations found for the Arctic aerosol.

We call our sampling program "routine" to distinguish it from the more common approach of an intensive field campaign which lasts for 2-4 weeks. We believe that, because of the large seasonal changes in the Arctic aerosol, continuous, less intense sampling offers the most efficient way to describe the aerosol and deduce its sources. The timing of the seasonal, monthly, or weekly changes is an extremely important property of the Arctic aerosol, which can be related directly to meteorological changes, hence used to deduce the controlling meteorological features.

In Alaska we continue to sample at Barrow and Fairbanks. Both sites will enter their fifth year of continuous aerosol sampling in September 1980. At Barrow, the sampler remains at the GMCC Clean-Air Observatory to the northeast of NARL. During July and August 1979 an improved, expanded relay box for the smple-control system was installed by R.J. McCaffrey of URI, designed to accept multiple high- and low-volume samplers simultaneously, and operate them in nearly any combination of clean-sector, dirty-sector, and continuous sampling. At the same time, a second URI high-volume aerosol sampler, indentical to the first, was installed, to allow us to take samples for other researchers, or different types of samples for us simultaneously. This sampling unit was purchased with funds from ONR and GMCC.

Altogether, we now have a "mini sampling center" at the GMCC Observatory. The second sampler has already been used by Dr. Joan Daisey of New York University for collecting aerosol samples; Dr. Hal Rosen of Lawrence Berkeley Laboratory has used the sample controller for his low-volume aerosol sampling of carbonaceous aerosol since December 1979. Our second high-volume sampler will be used by Miss Noëlle F. Lewis for her M.S. study of trace elements as a function of particle size in the Barrow aerosol (see Section II.B.16 below). The high-volume cascade impactors which will collect samples for her fit directly onto the filter holders already in these shelters.

During July 1979, the Fairbanks high-volume sampler was moved from Ester Dome, where it had been since September 1976, to the Poker Flat Research Range, 30 km north of Fairbanks and in the next valley. By this move, we hoped to decrease contamination from the Fairbanks area, decrease the concentration of local soil dust (Ester Dome was dry and dusty), transfer operation of the unit to the personnel at the Poker Flat site (formerly someone from the Geophysical Institute had to travel to Ester Dome each time a filter had to be changed, which became difficult in winter), and to associate our air-quality measurements with longer-term ecological studies planned for the Poker Flat area (deposition measurements by Dr. James Galloway of the University of Virginia began there in November 1979). Dr. Neal Brown, director of the Poker Flat Research Range, supervises the sampling for us, and is an enthusiastic collaborator. Recently, sampling of the aerosol on Millipore filters for determination of absorptive properties by diffuse reflectance (UA) was begun

Because of the importance of the northeast United States as a potential source of Arctic aerosol, we have had a program of continuous aero sampling there since August 1976. In October 1979 we terminated operat s in New York City, because we had built up three years of samples from roof of the NYC Medical Center in midtown Manhattan, and because we ha too many other sites of higher priority to deal with. Sampling at Narr. gansett, R.I., a site which represents the northeast U.S. quite well, continues, however. This site has been operating since October 1977.

Mr. Randolph D. Borys of our program installed a high-volume sampling site at Irafoss, Iceland, in August 1979. This site is a hydroelectric facility 30 km east of Reykjavik, and should represent regional Icelandic conditions very well. We are interested in Iceland because it is roughly the halfway mark along the proposed North Atlantic pathway for aerosol from North America to the Arctic. The importance of this pathway has recently been debated; we are happy to have an opportunity to check it directly. The sampler at Irafoss is operated by the Icelandic Meteorological Office, which through their representative Dr. Flosi Sigurdsson has been very kind to us. They are interested in undertaking whatever is necessary to characterize the Icelandic aerosol properly, which probably means a 5-10-year project. After our sampler was installed, they moved their own monitoring equipment to there from Reykjavik (daily samples of SO_2 and $SO_4^=$ as well as pH, Na, and $SO_4^=$ in precipitation). They are very interested in following our results. In addition, Dr. Rei Rasmussen of Oregon Graduate Center will conduct a feasibility study of halocarbons at Irafoss during summer 1980, and hopes to have a longer study there, starting in 1981. So the Iceland program is picking up momentum.

We are about to begin aerosol sampling in Ireland, in an effort to investigate transport of aerosol across the North Atlantic from North America to Europe. We have developed a cooperative program with Dr. Edward C. Monahan of the Department of Oceanography of University College Galway (and an ONR contractor), under which aerosol samples will be collected at Gort na gCapall (on Inishmore, one of the Aran Islands just outside Galway Bay). Sampling was scheduled to begin in April 1980, but as of July 1980 we have not yet received any filters. This program is a relatively expensive one, much more so than originally anticipated, and any decision to continue it beyond the contracted one year will have to be based on positive results.

In order to better characterize the European aerosol, which is suspected to be the source of much of the Arctic winter aerosol, we have arranged a no-cost, cooperative project with the Service des Pollutions of the Laboratoire Central, Préfecture de Police, Paris, through Mr. G. Thibaut and the director, Prof. H. Viellard. As of 1 October 1979, they are taking continuous samples from the Tour Saint-Jacques in the center of Paris. This site has an elevation of 50 m above local ground, and should be very representative. Samples are taken twice weekly, with their equipment, on 47-mm Whatman No. 40 filters. We have received the first thirty filters. This project should prove to be a valuable one, for there is very little information available about trace elements in the European aerosol, and still less about seasonal variations there.

We are continuing our cooperative arrangement with Dr. B. Ottar of the Norwegian Institute for Air Research (NILU), to receive portions of his high-volume filter samples taken at the NILU Spitsbergen and Bear Island sites. These samples are taken three times weekly; we normally receive one-quarter or one-half the filter. Samples arrive at URI in large groups, usually with a considerable time delay after sampling. We have analyzed samples from both locations so far; the results for Bear Island formed much of the basis for KAR's visit to NILU during summer-fall 1979.

2. Chemical analysis of the routine samples (URI)

Nondestructive neutron activation remains the heart of our analytical program. Whatever else is done to a sample, it is analyzed by neutron activation. Mr. Thomas J. Conway is our analyst; in addition, Mr. Randolph D. Borys of Colorado State University analyzes the samples from his field trips.

We also can analyze samples for $SO_4^=$ and ^{210}Pb, by methods developed by Dr. Richard J. McCaffrey. These techniques were discussed in the last proposal. During this contract period, Dr. McCaffrey calibrated the ^{210}Pb technique, and verified the extremely high concentrations of ^{210}Pb that Environmental Measurements Laboratory had measured at Barrow during 1975-76. At present, $SO_4^=$ and ^{210}Pb are measured as needed, not routinely on all samples.

Mr. Conway is currently learning the atomic absorption technique used by Mr. Patrick Harder of URI, with an eye toward analyzing Barrow samples for Pb, Cd, Mn, Cu, Zn and Ag. If the results prove sufficiently good, the analysis can be extended to other elements as well.

A major problem is that we have more samples coming in than we can analyze. This rather pleasant state of affairs reflects the cooperative spirit of several different organizations in taking routine samples for us in the field. We would like to do a complete neutron-activation procedure on all samples, as well as subsequent atomic absorption, but this is not possible. Even restricting our neutron activation to short irradiations, we are still farther behind than we would like. What is at stake here is more than just completeness; we now see that several of the elements determinable by long irradiation or by atomic absorption may help us resolve some of the puzzling elemental abundances showing up at Barrow, such as enriched Mn relative to V throughout much of the year, and in

particular during fall. We are looking for outside support for a second analyst, but so far without success.

3. Canadian aerosol sampler (URI)

During summer 1978 we constructed a high-volume aerosol sampler for the Canadian Atmospheric Environment Service, a division of Environment Canada. The system was identical to our Barrow sampler, and was intended for an AES-URI cooperative project at Alert, NWT. By the time the system was actually installed, however, Canadian plans had changed and it was placed at Mould Bay, NWT, as part of a much expanded and exclusively Canadian Arctic program. AES has now constructed similar systems for use in their 5-point Arctic Network. At present, two sites are operating, Mould Bay (since April 1979) and Igloolik (since November 1979). A third site, Alert, is scheduled to begin in July 1980. We now have a highly standardized Canadian-American Arctic Network: samplers, filter paper, and analytical techniques are all the same. Benefits of this approach began to show up at the URI Arctic conference in May, 1980, where their results from Mould Bay could be compared to ours from Barrow without hesitation.

4. Daily aerosol samples at Barrow (URI)

During winter 1979-80 we initiated a new program of daily high-volume aerosol samples at Barrow, in addition to the usual 4-day samples. This project was made possible by the summer 1979 installation of the second sampling system at Barrow, mentioned above. This project had several purposes: (1) Better correlate results with other data from the GMCC Observatory, such as CO_2, O_3, light scattering, and particle counts, all of which are much shorter-period than our 4-d samples, and usually cited as hourly means; (2) Compare daily aerosol concentrations with air-mass trajectories to Barrow, which are being generated four times daily by Dr. John Miller of NOAA/ARL in Silver Spring, Maryland. It is much better to compare daily samples than 4-day samples with these trajectories; (3) Better compare changes in aerosol with changes in synoptic systems and air masses near Barrow; (4) Investigate the relative amplitudes of the various temporal cycles at Barrow, and compare with corresponding data in the Norwegian Arctic. The relative importance of the various periodicities can be used to deduce information about the controlling mechanism of aerosol transport, such as long waves vs. short waves vs. eddy diffusion. Through cooperative studies with the Norwegian Institute for Air Research, we have seen a very pronounced quasi-weekly cycle at Bear Island, thanks to their daily samples of SO_2 and $SO_4^=$. This cycle is associated with the passage of synoptic systems, most notably the Barents Low, through the area. But at Barrow the weather seems to be more persistent, and we wonder if this shows up in the cycles of aerosol, too. Our data from Barrow show two major cycles, annual and quasi-monthly (identified with changes in the pattern of planetary waves, or index cycle). Daily samples are needed to properly observe a weekly cycle.

No results are available yet for these new samples, other than the visual observation that they were all noticeably gray.

5.   The Narwahl Island experiment (URI)

Ever since we began to sample surface aerosol at Barrow and found abnormally concentrated pollution aerosol in the winter, there has been nagging doubt that, in spite of all our efforts to exclude local (North Slope) sources, our samples represented local aerosol rather than anything Arctic-wide. Through a variety of chemical arguments we convinced ouselves that we were observing truly regional properties. But others, not familiar with the power of these arguments, were less convinced. A variety of exotic mechanisms for effects of Barrow itself, even when the wind was coming from the opposite direction, were proposed (regional re-circulation, deposition and later resuspension from snow, etc.), none of which was defensible but each of which could raise doubts. To settle the matter once and for all, we devoted our spring 1979 field work in the Arctic to a careful check of the regional representativity of our measure-ments at Barrow.

The program was run by Mr. Thomas J. Conway, and was a great success. Simultaneous daily aerosol samples were taken at Barrow and Narwahl Island, 30 km north of Prudhoe Bay, under clean-air conditions (N/NE winds) at each. During these periods, aerosol observed at Narwahl came from long trajectories over the Arctic Ocean, and reached our sampler before it encountered any other human activity on the North Slope. In addition, a few comparison samples were taken at Deadhorse Airport in Prudhoe Bay, in the center of the biggest aerosol source on the North Slope.

The results showed conclusively that Barrow air from the clean sector truly represents the Alaskan Arctic as a whole. Concentrations of the pollutants V and $SO_4^=$ were nearly the same at Barrow and Narwahl. Equally interestingly, filters from both locations were equally gray (indicating the same concentrations of sooty carbon), and could not be told apart by eye.

Results from this experiment will be written up in detail in the fall of 1980. In the meantime, they have convinced us that our approach to sampling aerosol of the Alaskan Arctic is basically sound. (Canadian results from Mould Bay, a 12-man meteorological outpost 1300 km ENE of Barrow, have confirmed almost all the results from Barrow.)

This section would not be complete without a word of caution. Human activity is increasing rapidly around Barrow, and it is becoming harder and harder to sample regionally representative air. The clean-air sector is being continually eroded, although there are still 130° left. We can see the time coming when Barrow will no longer be an acceptable site, although we do not know when this will be. In the meantime, we do what we can as fast as we can.

6.   Cooperative programs (URI,UA)

The scope of our Arctic studies has been broadened greatly through cooperative activities—mostly unfunded—with other interested institutions.   The current status of each of these is reviewed in this section.

Our major cooperation has of course been with the NOAA GMCC program. At their Barrow observatory they have provided a sampling platform, all the electricity we used (annual rate of 30,000 kWh when both pumps are

running), observers to change our samples and maintain the equipment, as well as abundant chemical and meteorological data from their own measurements. The result of GMCC's generosity is that Barrow is now the most studied site for Arctic air chemistry.

A similar relationship has developed for Poker Flat Research Range, outside Fairbanks, but on a smaller scale. They have provided the electricity and change the filters, in return for our data as it becomes available.

Dr. Joan M. Daisey of the New York University Medical Center, Tuxedo, New York, became interested in organic matter in the Arctic aerosol in early 1979. Since then, she has analyzed filters which we exposed for her at Barrow and Narwahl Island, from spring and summer 1979. Her results have shown that organic matter is probably the second most abundant constituent of the Arctic winter aerosol (excluding sea salt, or course), and that its sources are different from winter to summer. Dr. Daisey has turned a portion of her organic extracts over to Dr. Charles Weschler of Bell Laboratories, Holmdel, N.J., who has further analyzed them for a variety of substances, among the most interesting of which is a variety of silicones, at concentrations many times higher than in New York City. The reasons for this are not yet clear; contamination from the sampler cannot be absolutely excluded but seems unlikely. We will look into the problem of silicones further, because they are potentially of great environmental interest.

In June 1979 we undertook a cooperative ^{222}Rn experiment with Environmental Measurements Laboratory (a facility of the Department of Energy) in New York City. The concentration of ^{222}Rn in the Arctic is of great interest to us, because it is the precursor to ^{210}Pb, which both we and EML have found to be extremely high at Barrow during winter. From the ^{210}Pb/^{222}Rn ratio one can, in principle, derive some idea of the residence time of the Arctic aerosol. We have hypothesized that Arctic residence times must be very long, in order to account for the high ^{210}Pb, $SO_4^=$, V and other constituents of the Arctic aerosol in winter; we wanted to check this idea experimentally. EML provided two small integrating Rn detectors which had been used previously at their Chester, NJ field site. Because neither of these devices was intended for use in the Arctic, where concentrations of Rn may be very low, they were first exposed for a month, then for two weeks when it became apparent that the concentrations of Rn were sufficiently high. Useful data were obtained for November 1979 through January 1980, although the cold of Barrow affected both instruments. Now that we are beginning to get a feeling for the actual concentrations of Rn at Barrow (which are surprisingly high, particularly during early winter), EML has become more interested, and will lend us their ERM-3 (Environmental Radon Monitor), a prototype instrument of great sensitivity. Our proposed work with this instrument is described in Section II.B.15, below. The original contacts with EML involved Dr. Alfred J. Breslin and Mr. Andreas George. Mr. Breslin has since retired; we are now dealing with Mr. George, Acting Director of the Aerosol Studies Division.

We have a continuing cooperative arrangement with Dr. Edward M. Patterson of the School of Geophysical Sciences, Georgia Institute of Technology, Atlanta, for determining the absorptive properties (the darkness) of aerosol on Arctic filters. Dr. Patterson and students receive filters from us, some of which are especially taken and some of which are routine, and use the Kubelka-Munk technique to determine broad-band absorption of

visible radiation via the diffuse transmittance of light through a filter. This information not only can be put directly into calculations of radiative effects of Arctic aerosol, but also provides an estimate of the concentration of sooty carbon in the aerosol. Dr. Patterson originally worked with a suite of 16 samples from 1976-78, but now will broaden his efforts to the total set of approximately 100 samples. A joint article on these results is being prepared.

Together with the Danish Air Pollution Laboratory and the Danish Meteorological Institute, we have analyzed a series of winter and summer aerosol samples from northern and southern Greenland (Thule and Prins Christianssund, respectively). These results were reported in part by Rahn and McCaffrey (1979a). They show northern Greenland to have higher aerosol conentrations than southern Greenland, particularly during winter. Thule generally had less aerosol than did Barrow or Spitsbergen, however. Part of this project was a PIXE-NAA comparison (PIXE was used by the Danish group). For elements in common, results compared to 20% or better. This program was arranged by Dr. Hans Buch of DMI and Dr. Neils Z. Heidam of DAPL.

Of course, cooperation continued between URI and the Norwegian Institute for Air Research. Samples from the Norwegian Arctic were analyzed at URI, results and their significance were discussed, and joint papers were prepared. KAR spent three months there in 1979 (Section 14 below).

A cooperative project is presently being worked out with Dr. R.A. Rasmussen of Oregon Graduate Center, Beaverton. Dr. Rasmussen measures atmospheric halocarbons with great precision, and is extremely anxious to begin a systematic program in the Arctic. We plan two feasibility studies: (1) we will take flask samples for him on the YMER this summer; (2) the Icelandic Meteorological Office will take a series of samples at Irafoss this summer and fall. The results of these studies will be used to plan more detailed studies, probably at Irafoss and Barrow. Among other things, the halocarbons should provide tracers for North American vs. Eurasian air masses. In combination with the aerosol tracers we are developing, this should provide a powerful tool for determining the area of origin for the Arctic aerosol.

General Motors Research Laboratories, through Dr. George Wolff, has offered to see whether it can determine concentrations of elemental carbon on our library of back filters from Barrow. If they are successful, an enormous amount of valuable data could result. We have not yet learned the results of their tests, however.

7.   Subcontracts(URI)

The University of Rhode Island let four subcontracts during the FY1979-80 Arctic Haze contract period: the usual one to the University of Alaska to support Dr. Shaw ($35,703 for FY80), one to Colorado State University in FY80 for expenses of Mr. Randolph D. Borys ($8150; for purely administrative reasons), one to University College Galway (Dr. E.C. Monahan) to support aerosol sampling on the Aran Islands (FY80, $3000) and one for $3000 to Dr. Elmar Reiter of Colorado State University to perform a series of meteorological calculations concerning the relation between planetary waves in the atmosphere and transport of aerosol to Barrow. Dr. Reiter's work has been completed; his report is discussed in Section I.C.3.

During the coming contract period we will continue to subcontract to University of Alaska. Because of extreme difficulties in setting up the subcontract with Colorado State University, however, we may resume our earlier practice of handling Mr. Borys' expenses from URI. As noted above, renewal of the Galway subcontract depends upon the first results, which are not yet in. Dr. Reiter's subcontract was a one-time venture, and will not be renewed.

8. Approaches to the USSR (URI)

No study of the Arctic is complete without participation by the USSR. Ever since the Arctic Air-Sampling Network was established in 1977, we have been continually attempting to interest them in joining. Dr. B. Ottar has contacted representatives of the USSR through his EMEP/ ECE/UN activities, to no avail. K. Rahn has tried through his contacts, with the same results. The USSR State Committee on Hydrometeorology and Protection of the Natural Environment, Moscow, was contacted twice about attending the May 1980 Arctic Symposium at URI, but never answered.

The importance of involving the USSR in our Arctic work increases with time, as our estimates of the role of the USSR in Arctic air chemistry increase (see Section I.C.3 below). We are presently trying to arrange some sort of cooperative program through Working Group VIII of the US-USSR Joint Committee on Environmental Protection. The subject of a US-USSR Arctic experiment was raised informally last fall to members of the USSR delegation visiting the US, and was received with cautious optimism by them. No formal answer has been given yet, however.

In October 1980, KAR will present the case for a joint study to a meeting of US and USSR delegations of working Group VIII in Miami. We hope that concrete plans can be laid soon after that.

9. Ph.D. thesis of R.D. Borys, Colorado State University

The Ph.D. thesis of Mr. Randolph D. Borys of the Department of Atmospheric Science of Colorado State University, on cloud-active aerosol in the Arctic, supported one-half by the Arctic Haze Contract, continues on schedule. The planned summer and winter field studies at five Arctic and one midlatitude sites have been completed, except for the summer study at Spitsbergen. This latter visit was scheduled for late summer 1979, but had to be cancelled when costs of reaching the research site at Ny Ålesund rose excessively after the local ship stopped running for the season. Instead, summer data for the Spitsbergen area will be collected by Mr. Borys from the YMER in August and September 1980. A summary of Mr. Borys' field work to date is shown in Table 1.

Samples were collected during approximately 10 consecutive 24-h periods at the sites. Types of samples included high-volume aerosol filters (for determining trace elements and sulfate), cloud-condensation nucleus (CCN) concentrations, condensation-nucleus (CN) concentrations, filters for ice-nucleus concentrations, and filter samples for determination of particle-size distributions by a scanning electron microscope (SEM) and for determination of composition of aerosol particles by an SEM/XRF microprobe.

Table 1. Summary of field studies of cloud-active aerosol

in the Arctic and environs.

Site	Dates.	Number of 24-h samples
Barrow, AK	27 July - 7 August 1979 23 March - 7 April 1980	10 10
Poker Flat, AK	17 - 25 July 1979 4 - 13 April 1980	9 10
Pangnirtung, NWT, Canada	21 June - 3 July 1979 26 January - 3 February 1980	9 7
Irafoss, Iceland	14 - 25 August 1979 16 - 23 February 1980	9 7
Ny Ålesund, Spitsbergen	28 February - 11 March 1980	11
Narragansett, RI	12 - 19 June 1979 6 - 15 January 1980	10 7

Mr. Borys' neutron-activation and sulfate analyses are completed (at URI). Summer CCN and CN counts are in rough tabular form. The winter CCN data require photographic processing before analysis. No ice-nucleus concentrations have been determined yet.

10.   Trace elements in desert soils (URI, Mainz)

The work of Dr. Lothar Schütz on trace elements in size-fractionated desert soils, started during a 9-month research visit to URI in 1977, continues to be processed. This work is being carried on at his new institute, Institut für Meteorologie der Universität, Mainz, where Dr. Schütz moved from the Max-Planck-Institut für Chemie, Mainz, in late 1979. All concentration data have been completed and are stored in the university computer. They have been printed out in various tables, and over 200 plots have been drawn by computer. In March 1980 KAR met with Dr. Schütz to go over these data; a summary of the results appears in Section I.C.9 below.

The essence of the results will be presented by Dr. Schütz at the annual meeting of the German aerosol society GAF in fall 1980. After that, a large report will be prepared, to be followed by one or more journal articles. These data have already been of use to us in under-standing the V/Al ratio of aerosols, and will shortly be used to help interpret Mn/Al patterns emerging in the Arctic. This is only the tip of the iceberg; they should see many more applications.

11.   Optical properties and radiative effects of Arctic haze (UA)

At UA, Dr. Shaw has continued to accumulate sun-photometer measurements of the wavelength dependence of extinction by aerosol in and around the Arctic. From 1978-80, data have been taken at Fairbanks, Barrow, and at Kongsøya in the Norwegian Arctic. At the same time, the scattering phase function of the Arctic aerosol has been evaluated from measurements of the intensity and wavelength distribution of skylight at selected scattering angles (from the sun).

The column-integrated particle-size distribution of the Arctic aerosol is recoverable from this optical data, by means of a constrained linear inversion program which has been developed by GES over the past two years and tested extensively on simulated data (Shaw, 1979; Section I.E.3 here). At present, actual radiation data from 4 or 5 test periods are being in-verted; preliminary results are given in Section I.C.10 below. Currently, the major uncertainty in the inversion procedure is the magnitude of the single-scattering albedo of the aerosol. In an effort to remedy this, GES and KAR have recently received a two-year NSF grant "Radiative effects of Arctic haze", one of whose major goals is to determine the single-scattering albedo of the Arctic aerosol by a variety of independent means. The con-tribution of this ONR Contract to measuring this parameter is determining the absorptive properties of the aerosol by measuring the spectral reflec-tivity of exposed Millipore filters, then applying Kubelka-Munk theory.

The measured and derived optical parameters of the Arctic aerosol (monochromatic optical thickness, phase function, particle-size distribu-tion, single-scattering albedo, etc.) are used to predict heating and cooling rates in the Arctic attributable to the aerosol. Calculations are performed with an efficient computer program for radiative transfer in the Geophysical Institute. It accounts for all orders of scattering,

which is very important for the low solar elevations in the Arctic.
Initial results from an early version of the program were reported at
the aerosol conference of the New York Academy of Sciences in January
1979 (Shaw and Stamnes, 1980; Section I.E.5 here); it predicted an at-
mospheric heating of about 1°C per day in spring near the surface of
the Arctic. This value is high enough to be climatically significant,
and has encouraged us to continue. A new program is now being developed,
with two major improvements: (a) it includes radiation of all wavelengths,
weighted by the wavelength distribution of incoming solar radiation,
instead of the single wavelength of the original program; and (b) it
includes absorption by atmospheric water vapor. This new program,
combined with updated radiative data from the Arctic, should provide a
much improved estimate of the actual direct radiative effects of Arctic
haze.

Other radiation-related activities of UA for 1978-80:

- An aerosol camera photometer has been developed.

- New calibration scales and techniques have been developed to improve
  the accuracy of our field measurements.

- A new, smaller sun photometer has been developed, examples of which
  will be distributed over the Arctic Air-Sampling Network (see Section
  II.B.4 below).

- A photometer was lent to the Norwegian Institute for Air Research.

- A photometer was lent to Dr. B. Holmgren of Uppsala University for
  winter use at Tafala Glacier in northern Sweden.

- A newly developed stellar photometer, whose function is to measure
  Arctic haze in the dark of winter, was tested in 1978-79, but gave
  disappointing results because of instrumental drift and difficulties
  in calibration. Plans are being made to install it in an insulated
  cubicle at Poker Flat Research Range outside Fairbanks.

- We are working out a collaborative program with Dr. W. Sellers of
  the University of Arizona. He will insert some of our derived
  radiation parameters for the Arctic into his climatic evaluation
  program, in order to see whether Arctic haze might be expected to
  have any global climatic effects.

12. Particle dynamics and single-particle chemistry (UA)

In conjunction with his work on determining the particle-
size distribution of the Arctic aerosol (Sections I.B.11 and I.C.10), G.
Shaw of UA is presently studying the mass flux of Arctic aerosol through
the size distribution. These results will aid in interpreting the size
distributions that are eventually found, in terms of types of sources,
types of sinks, age, etc.. Nucleation, coagulation, impaction and diffu-
sion transfer material from the smallest sizes ($r \sim 10^{-3} \mu m$) to the largest
sizes ($r > 1 \mu m$), where they can be removed by sedimentation and impaction.
From the size distribution of the Aitken particles ($r < 0.1 \mu m$), one may
be able to deduce the gas-to-particle conversion rate, hence the flux of
material through the size spectrum, and possibly a lower limit for the
age of the particles as well. The particle-size distribution in the
radius range 0.001 - 1 μm is being determined with a Nuclepore diffusion
chamber and with an accurate, very sensitive air-expansion particle

counter which has been constructed at the Geophysical Institute from
ONR funds. The particle-size distribution is deduced from a computer
program for nonlinear Chahine-type inversion. These instruments and the
computer program were developed in 1979-80 with substantial effort.
Preliminary tests indicate that size distributions inferred from this in-
version procedure agree well with those obtained directly from counting
on electron-microscope images of filters; Dr. Shaw anticipates a fruitful
next year of application.

Electrical-mobility measurements can also provide information on
the size distribution of Aitken particles. Presently available commercial
instruments are unsuitable for Arctic conditions. A newly developed mobility
analyzer of the Institut für Meteorologie (Mainz), which apparently can
detect and size particles automatically even at the low concentrations
expected for the Arctic, will be investigated by W.E. Raatz of UA during
the YMER expedition of July-August 1980. He hopes to persuade the Mainz
group to undertake a collaborative program in the Arctic.

Dr. Shaw is also investigating the possibility of observing a self-
preserving size distribution in the Arctic. Theory predicts that, given
enough time and steady-state of sources and sinks of aerosol, a unique,
so-called self-preserving particle-size distribution of aerosol should be
established which is independent of the original size distribution. The
predicted self-preserving size distribution is never observed in midlati-
tudes, presumably because aging times are too short and/or conditions are
not sufficiently steady-state. But the Arctic, because of its size,
homogeneity, and remoteness, may allow the long-sought self-preserving
size distribution to be established. If so, this would be an exciting
development for both Arctic aerosols and aerosols in general.

Dr. Shaw has also been determining concentrations of sulfate-contain-
ing particles with the Bigg thin-film method, which is slowly becoming a
standard technique at the Geophysical Institute. X-ray spectrometry on
larger particles collected at the Poker Flat Research Range has found
lead-rich, vanadium-rich, and other pollution-derived particles.

13.   Ph.D. thesis of W.E. Raatz, University of Alaska

Dr. Shaw has recently acquired a Ph.D. student, Mr. Wolfgang
E. Raatz of West Germany. Mr. Raatz has a Master's degree in climatology
from Prof. Flohn's group in Bonn, and will do a Ph.D. thesis on the clima-
tology of Arctic haze. His thesis will have three parts: (1) a small his-
torical study of cloudiness in the Arctic since 1920 (completed and submit-
ted as an article to Atmospheric Environment); (2) a larger historical
study of haze and visibility from the Ptarmigan data (begun); and (3) a
contemporary study relating aerosol concentrations at Barrow to synoptic
configurations, cloudiness, precipitation, etc. For his raw chemical
data, Mr. Raatz is drawing on various stations of the Arctic Air-Sampling
Network, Barrow in particular. He is reducing the data on several chemi-
cal elements from 1976-79 at Barrow, Fairbanks, and ultimately other sites
as well, to uniform 5-day means, then running correlations, cross-correla-
tions, and other statistical tests on these data.

Mr. Raatz's graduate stipend comes partially from the Arctic Haze
Contract and partially from the University of Alaska.

14. **K. Rahn's research visit to NILU, 1979**

During July-October 1979, K. Rahn spent 3 months at the Norwegian Institute for Air Research, Lillestrøm, as a research visitor. This was seen as the first in a series of exchange visits within the Arctic Air-Sampling Network. It was enormously profitable. The results included the development of the hypothesis of return-flow circulation of air to the Norwegian Arctic during winter, a study of transport across the North Atlantic from EMEP data (none found for SO_2, only $0.4\mu g\ m^{-3}$ for $SO_4^=$), a NILU report on the relative importance of western Europe, eastern Europe, and the western USSR as sources of Arctic aerosol, and the first draft of a letter on SO_2 at Bear Island for Nature (submitted May 1980). These ideas are discussed further in Section I.C below. In addition, KAR was given access to the huge library of EMEP data, which is a valuable resource for evaluating potential European sources of Arctic aerosol and SO_2. We hope to have further exchanges in the near future.

15. **Progress of the Arctic Air-Sampling Network, 1977-80**

The Arctic Air-Sampling Network continues to thrive. Because ONR contributes to the Network via its support of sampling stations at Barrow, Fairbanks, Rhode Island, Iceland and Ireland, comments on the general state of the Network may be of interest.

From its original 9 sampling stations in 1977, the Network has grown to a present roster of 16 sites. Programs of individual countries or laboratories are also much better funded than they were three years ago, with the most progress made in Canada and Denmark. The variety of research carried out at a given site is generally greater than it was three years ago, as well.

The Network has fallen into a pattern of small, technical meetings annually and larger, more diversified symposia each three years. Symposia were at NILU in 1977 and at URI in 1980; technical meetings were at the Danish Air Pollution Laboratory in 1978 and at URI in 1979. The next technical meeting will be held at Downsview, Ontario (Atmospheric Environment Service) in May 1981; the third Symposium on Arctic Air Chemistry will be held in spring 1983, at an undetermined location.

Communication within the network is provided by the Arctic Newsletter, edited by KAR. Distribution is just over 100 copies. At the May 1980 symposium KAR agreed to continue the Newsletter at least until 1983.

16. **Conferences attended**

URI Group

- GAMETAG Aerosol Measurements Workshop, Georgia Institute of Technology, Atlanta, 7-8 September 1978 (KAR). Participated in discussions and presented informal talk on Arctic haze.

- Americal Chemical Society Meeting, Miami Beach, 11-12 September 1978 (KAR).

- GAF (Gesellschaft für Aerosolforschung) Annual Meeting, Vienna, 26-28 September 1978 (KAR).

Papers presented (GAF 78):

(1) Aerosol-snow fractionation of trace elements
    (Rahn-McCaffrey).
(2) Elemental composition of Aitken particles (Schütz-
    Ketseridis-Rahn).

- NSF Workshop on Atmospheric Chemistry, National Center
  for Atmospheric Research, Boulder, CO, 16-18 October
  1978 (KAR). Member, Trace Element Panel.

- U.S. Department of State, Bureau of Oceans and Inter-
  national Environmental and Scientific Affairs, Washington,
  DC, 20 November 1978 (KAR). First briefing on the Arctic
  aerosol.

- Planning meeting for YMER-80, Department of Physical
  Geography, University of Stockholm, 8-9 December 1978
  (KAR). Talk on Arctic air chemistry and why it should
  be part of YMER-80.

- Arctic Air-Sampling Network Technical Meeting, URI,
  5-6 January 1979 (KAR, RJM, TJC).

- New York Academy of Sciences Conference on Aerosols:
  Anthropogenic and Natural—Sources and Transport, New
  York City, 9-12 Januay 1979 (KAR,RJM). Chaired session
  on long-range transport of aerosol (KAR). Presented Rahn-
  McCaffrey paper on winter Arctic aerosol and Shaw-Stamnes
  paper on radiative effects of Arctic aerosol (KAR).

- NOAA/GMCC Annual Meeting; GMCC Aerosol Chemistry Workshop,
  Boulder, Colorado, 6-9 March 1979 (KAR). Brief talk on
  what species in the Arctic aerosol should be monitored
  by GMCC.

- Environmental Measurements Laboratory, New York City,
  13 June 1979 (KAR, RJM). Met with H. Feely. Arranged
  for cooperative ^{222}Rn program with A. Breslin and A.
  George.

- U.S. Department of State, Bureau of Oceans and International
  Environmental and Scientific Affairs, Washington, DC, 16
  July 1979 (KAR). Second briefing on the Arctic aerosol.

- WMO Symposium on Long-Range Transport of Pollutants and
  its Relation to General Circulation, Including Strato-
  spheric/Tropospheric Exchange Processes, Sofia, Bulgaria,
  1-5 October 1979 (KAR). Presented paper on Arctic air
  chemistry, with reasons why USSR should be a part.

- Symposium on High Latitude Climate Systems of the American
  Meteorological Society, Boston, Mass., 6-8 November 1979
  (KAR,RJM, TJC). Presented brief talk on radiative effects
  of the Arctic aerosol (KAR).

- U.S. Department of State, Bureau of Oceans and International
  Environmental and Scientific Affairs, Washington, DC,
  9 February 1980 (KAR,TJC). Third briefing on the Arctic
  aerosol.

- Aerosol Receptor Model Workshop, Durham, NC, 25-28 February 1980 (KAR). Participated in working groups; presented a short talk on similarities between receptor modeling of aerosol in urban and remote areas.

- Second Symposium on Arctic Air Chemistry, URI, 6-8 May 1980 (KAR, RJM, TJC). Many different activities.

  UA Group

- WMO Technical Conference on Regional and Global Scale Observation of Atmospheric Pollution relative to Climate, Boulder, Colorado, 20-24 August 1979 (GES). Presented paper "The meaning of atmospheric turbidity".

- International Union of Geodesy and Geophysics and International Association of Meteorology and Atmospheric Physics General Assembly, Canberra, Australia, 2-15 December 1979 (GES). Presented paper "Considerations on the polar aerosol". After the meeting, GES spent 4 days at the laboratory of CSIRO Division of Cloud Physics, with K. Bigg, G. Ayers, J. Gras, and A. Ono (Nagoya Univeristy). Dr. Bigg lent UA a G.E. nucleus counter for use under this contract.

- Second Symposium on Arctic Air Chemistry, URI, 6-8 May 1980 (GES,WER). GES presented the paper "Eddy-diffusion transport of Arctic haze: A preliminary model"; WER presented the paper "Trends in cloudiness in the Arctic since 1920".

17. <u>Second Symposium on Arctic Air Chemistry, URI, 6-8 May 1980</u>

The Arctic Air Chemistry Group of URI held this symposium, with KAR as chairman. It was co-sponsored by ONR, NSF, NOAA/GMCC, and URI. Total attendance was 46, from 6 countries; 26 papers and several shorter communications were given. An issue of <u>Atmospheric Environment</u> will be devoted to papers from this conference. The final program is included here for reference.

The results of this conference will do much to stimulate progress in Arctic air chemistry.

18. <u>YMER-80 expedition (URI, UA, CSU)</u>

We are presently participating in the Swedish YMER-80 scientific expedition to the Greenland-Spitsbergen-Franz Joseph Land Arctic. This project constitutes our Arctic field work for 1980. From URI, T.J. Conway is on the entire first leg (24 June - 7 August 1980); R.D. Borys of CSU will be present for parts a and b of the second leg (9 August - 7 September 1980). Samples taken include high-volume filters for trace-element analysis, high-volume filters for organic analysis, low-volume filters for microscopy, and sun-photometry measurements (TJC). During the second leg, Mr. Borys will do all of the above, plus his own cloud-active nucleus counting and flask sampling for analysis of halocarbons by Dr. R.A. Rasmussen of Oregon Graduate Center. We are antʼ ʻi-pating a very worthwhile experiment.

SYMPOSIUM PROGRAM

Second Symposium on Arctic Air Chemistry
6 - 8 May 1980

Tuesday, 6 May 1980

0800   Bus leaves Holiday Inn for Bay Campus

0810-0830   REGISTRATION

0830-1200   MORNING SESSION   Studies on the Alaskan Arctic
            Chairman: Leonard A. Barrie, Atmospheric Environment Service, Canada.

            Welcome: Kenneth A. Rahn (0830-0835)

            Introduction to Tuesday's Session: L.A. Barrie (0835-0840)

            "Progress in Arctic air chemistry, 1977-80"
                K.A. Rahn (0840-0900)

            "Particulate organic matter in the Arctic aerosol"
                J.M. Daisey, R.J. McCaffrey and R.A. Gallagher (0900-0930)

            "Identification of selected organics in Arctic aerosols"
                C.J. Weschler (0930-1000)

            COFFEE BREAK (1000-1030)

            "Aerosol light scattering and condensation nuclei measurements
            at Barrow, Alaska"
                B.A. Bodhaine and J.M. Harris (1030-1100)

            "Coefficients of absorption of visible radiation by the Barrow
            aerosol and their seasonal variations"
                E. Patterson and K.A. Rahn (1100-1130)

            "Determination of soot in the Arctic haze near Barrow,
            Alaska"
                H. Rosen (1130-1200)

1200-1330   LUNCH

1330-1715   AFTERNOON SESSION Studies on the Alaskan Arctic, continued

            "A 5-year climatology of 5-day air-mass trajectories to Barrow,
            Alaska"
                J. Miller (1330-1400)

            "A source-oriented and meteorological interpretation of the NOAA/ARL
            5-day air-mass trajectories to Barrow, Alaska"
                T.J. Conway, K.A. Rahn and J. Miller (1400-1430)

"Planetary-wave behavior and Arctic air pollution"
    E.R. Reiter (1430-1500)

COFFEE BREAK (1500-1530)

"Monte Carlo techniques for studying the long-range atmospheric
transport of pollution aerosol"
    R. Husar (1530-1600)

"Synoptic meteorology as a factor in the summer decrease
of carbon dioxide at Barrow, Alaska"
    B.C. Halter and J.T. Peterson (1600-1630)

Short presentations (1630-1700)

Summary and general discussion (1700-1715)

1715	Bus leaves for Holiday Inn
1730-1925	Supper
1925	Bus leaves hotel for Bay Campus
1930-2100	Social hour

Wednesday, 7 May 1980

0800   Bus leaves Holiday Inn for Bay Campus

0830-1200   MORNING SESSION   Studies in the Canadian, European, and
                                   Greenland Arctic
           Chairman: Hans Flyger, Danish Air Pollution Laboratory

           Introduction to Wednesday morning's session: H. Flyger (0830-0835)

           "The influence of mid-latitudinal pollution sources on haze
           in the Canadian Arctic"
               L.A. Barrie, R. Hoff, and S. Daggupaty (0835-0900)

           "The transfer of airborne pollutants to the Arctic region"
               B. Ottar (0900-0930)

           "Trajectories of polluted air masses from Eurasia to Bear
           Island"
               T.J. Conway and E. Joranger (0930-1000)

           COFFEE BREAK (1000-1030)

           "On the origin of the Arctic aerosol: A statistical approach"
               N.Z. Heidam (1030-1100)

           "Airborne concentrations of trace elements in South-Central
           Greenland"
               C.I. Davidson, L. Chu, T.C. Grimm, M.A. Nasta and
               M.P. Qamoos (1100-1130)

Short presentations and summary (1130-1200)

1200-1330   LUNCH

1330-1715   AFTERNOON SESSION   Larger-scale chemical and meteorological
                                Studies
             Chairman: Hans Flyger, Danish Air Pollution Laboratory

             Introduction to Wednesday afternoon session: H. Flyger (1330-1335)

             "Dispersion of aerosols in the Arctic"
                  F. Fanaki (1335-1400)

             "An eddy-diffusion transport model for Arctic haze"
                  G.E. Shaw (1400-1430)

             "Remote sensing of the vertical concentration of aerosols and
             ozone in the Arctic atmosphere"
                  T.J. Pepin and R. Lane (1430-1500)

             COFFEE BREAK (1500-1530)

             "The role of ^{210}Pb and ^{222}Rn in understanding sources and
             transport of the Arctic aerosol"
                  R.J. McCaffrey (1530-1600)

             "The role of COS and CS_2 in the global sulfur budget: Implication
             on the Arctic aerosol haze"
                  M.K.W. Ko and N.D. Sze (1600-1630)

             Short presentations (1630-1700)

             Summary and general discussion (1700-1715)

1715   Bus leaves for Holiday Inn

1830   Bus leaves for conference dinner

1900   Conference dinner

2130   Bus to Holiday Inn

                         Thursday, 8 May 1980

0800   Bus leaves Holiday Inn for Bay Campus

0830-1230   MORNING SESSION   Environmental effects of the Arctic aerosol
             Chairman: Kenneth A. Rahn, University of Rhode Island

             Introduction to Thursday morning's session: K.A. Rahn (0830-0835)

             "Measurements of cloud-active aerosol in the Arctic"
                  R.D. Borys (0835-0900)

"Trends in cloudiness in the Arctic since 1920"
     W.E. Raatz (0900-0930)

"Potential direct radiative effects of the Arctic aerosol"
     G.E. Shaw and K.A. Rahn (0930-1000)

COFFEE BREAK (1000-1030)

"Potential use of the diffuse-direct radiation method to deter-
mine the vertically integrated absorptive properties of the
Arctic aerosol"
     J.J. DeLuisi (1030-1100)

"Atmospheric, riverine and oceanic transport of trace elements
to the Arctic Ocean"
     K.A. Rahn (1100-1130)

Short presentations, general discussion and summary (1130-1200)

Overall summary of Symposium
     N.Z. Heidam (1200-1230)

1230-1330   LUNCH

1330-1730   AFTERNOON SESSION   Second General Meeting of the Arctic
                                Air-Sampling Network
            Chairman: B. Ottar, Norwegian Institute for Air Research

            Meeting (1330-1530)

            COFFEE BREAK (1500-1530)

            Meeting (1530-1730)

1730   Bus leaves for Holiday Inn

From UA, W.E. Raatz will be based on Kongsøya with a meteorological team from the Universities of Uppsala and Stockholm. His efforts include: (a) multi-wavelength sun-photometer measurements of atmospheric turbidity; (b) aerosol sampling with Millipore and Nuclepore filters for determining the absorptive properties of the aerosol; (c) sampling aerosols with a micro-impactor, for later determination of chemical composition and particle-size distribution; and (d) periodically sampling cloud droplets with a kite-borne and balloon-borne replicator, to identify and classify cloud-active nuclei (in cooperation with T. Ohtake and K. Jayaweera of UA). This field program will take place from 1 July through 7 August 1980.

19. State Department briefings (URI,UA)

The Bureau of Oceans and International Environmental and Scientific Affairs of the U.S. Department of State first came into contact with our Arctic haze research through Dr. Shaw, and has become very interested in it. KAR has briefed them three times on the progress of our studies and their implications, 20 November 1978, 16 July 1979 and 9 February 1980. A fourth, large briefing is anticipated for fall 1980, in connection with the recent formation of the U.S. Government's Interagency Arctic Policy Group, headed by L.A. Rose of the Office of Oceans and Polar Affairs of the above bureau.

C. Results, interpretation and ideas

1. Chemical composition of the Arctic aerosol (URI)

The Arctic aerosol seems to be dominated by sulfate, carbonaceous matter and water. (We speak here only of the submicron fraction of the aerosol, which can be transported long distances. The supermicron fraction, comprised mostly of sea salt and soil, is probably local in origin.) At Barrow in March, for example, nonmarine sulfate is 2.2 µg m^{-3}, extractable organics are 0.9 µg m^{-3}(Daisey et al., 1981), and elemental carbon is about 0.3 µg m^{-3} (from transmittance measurements by E. Patterson of Georgia Tech). Other primary pollutants are 0.4 µg m^{-3}, and nitrate is about 0.2 µg m^{-3} (Data for Mould Bay, NWT; Barrie et al., 1981). The amount of water in the aerosol is still uncertain, but is surely large; it may be on the order of 2-3 µg m^{-3}. Thus, sulfate and carbonaceous matter together account for about 85% of the dry submicron aerosol and about 50% of the total (wet plus dry) submicron aerosol. This dominance of sulfate and carbon in the submicron Arctic aerosol is strikingly similar to the composition of the Aitken range (r < 0.1µm) of remote aerosol (Schütz et al., 1978), and suggests a common origin for particles in these two ranges. The composition of the giant range (r > 1µm) is very different, however, both in the Arctic and elsewhere.

As noted earlier, we have confirmed the extremely high concentrations of the continental indicator ^{210}Pb found earlier by Environmental Measurements Laboratory for Barrow in winter. Their concentrations, which reach 30 ƒ Ci m^{-3}, are the highest yet recorded routinely for ^{210}Pb, to the best of our knowledge. They are all the more surprising because most of the air of winter comes to Barrow from the Arctic Ocean, not directly from land. There are several possible reasons for these high concentrations: locally high concentrations of precursor ^{222}Rn, high ^{222}Rn from distant continental areas, or long residence times for Arctic aerosol in winter.

Our preliminary results for ^{222}Rn at Barrow during three months of winter 1979-80, shown in Figure 1 (from a cooperative experiment with EML), indicate that it is more abundant there than in southern Alaska, as are ^{210}Pb and many other species in the aerosol. For ^{222}Rn this is consistent with either a distant northern source or an Alaskan southern source and northward air flow. Repeated simulations suggest, however, that the distant northern source is a much more likely explanation. A more detailed experiment with EML's ERM-3 Rn detector will determine whether high Rn at Barrow is associated with northern or southern air. We anticipate beginning this experiment in late 1980.

As more chemical data on the Arctic aerosol are generated, small but perhaps significant differences in composition are beginning to appear in different parts of the Arctic. It would seem that the Arctic aerosol is not quite the monolith that we once thought it was. The best example of this so far is the Mn/V ratio, which is turning out to be about two times higher in the North American Arctic (Barrow and Mould Bay) than in the Norwegian Arctic (Spitsbergen and Bear Island). An unusually high Mn/V ratio was also found for snow near Barrow by Weiss et al. (1978), but its meaning was overlooked completely until similar ratios were noted in mid-1980 for Barrow and Mould Bay. We feel that this factor of two is significant. It means that there is a different mix of pollution sources for the two sides of the Arctic, because if the Barrow-Mould Bay aerosol were derived from the Spitsbergen-Bear Island aerosol, as it would be if Eurasia alone were the primary source reggion, aging effects should make the Mn/V ratio lower, not higher, on the North American side. At present the explanation for these differences is not known with certainty; long irradiations and atomic absorption analysis of the samples should provide valuable clues, however. The preprint in Section I.E.7 discusses the Mn/V ratio and its meaning in more detail.

2. Seasonal variations of the Arctic aerosol (URI,UA)

The large seasonal variations of the Arctic aerosol seen in earlier years continue to be confirmed, both for later years and at other sites (Mould Bay, for example). As data increase a certain fine structure appears, however, with systematic differences in seasonal variations showing up for different locations, different years and different elements. It is now clear that seasonal raviations are greater on the North American side of the Arctic than on the Eurasian side. A hint of this was seen earlier for V (Rahn and McCaffrey, 1980; Section I.E.4 here); it is now confirmed for more elements at more sites. In general, we interpret this difference to mean that the major sources of Arctic aerosol are on the Euraisan side, i.e., Eurasia itself. An interesting point is that the lesser seasonal variation in the Eurasian Arctic comes from elevated summer minima rather than decreased winter maxima—winter concentrations of aerosol are about the same on both sides of the Arctic. Thus, the picture is emerging that aerosol is injected into the Arctic from the Eurasian side both summer and winter, albeit less frequently and with less intensity during summer. In winter, once the aerosol reaches the Arctic, where precipitation is meager and residence times of aerosol are long, it can travel freely all the way to the other side. But aerosol injected into the Arctic in summer is removed much faster by greater precipitation, hence is confined to a region much closer to its source. As a result, the "pole of minimum concentration" in summer is shifted from the North Geographic Pole to the North American Arctic, into the Barrow-Mould Bay region.
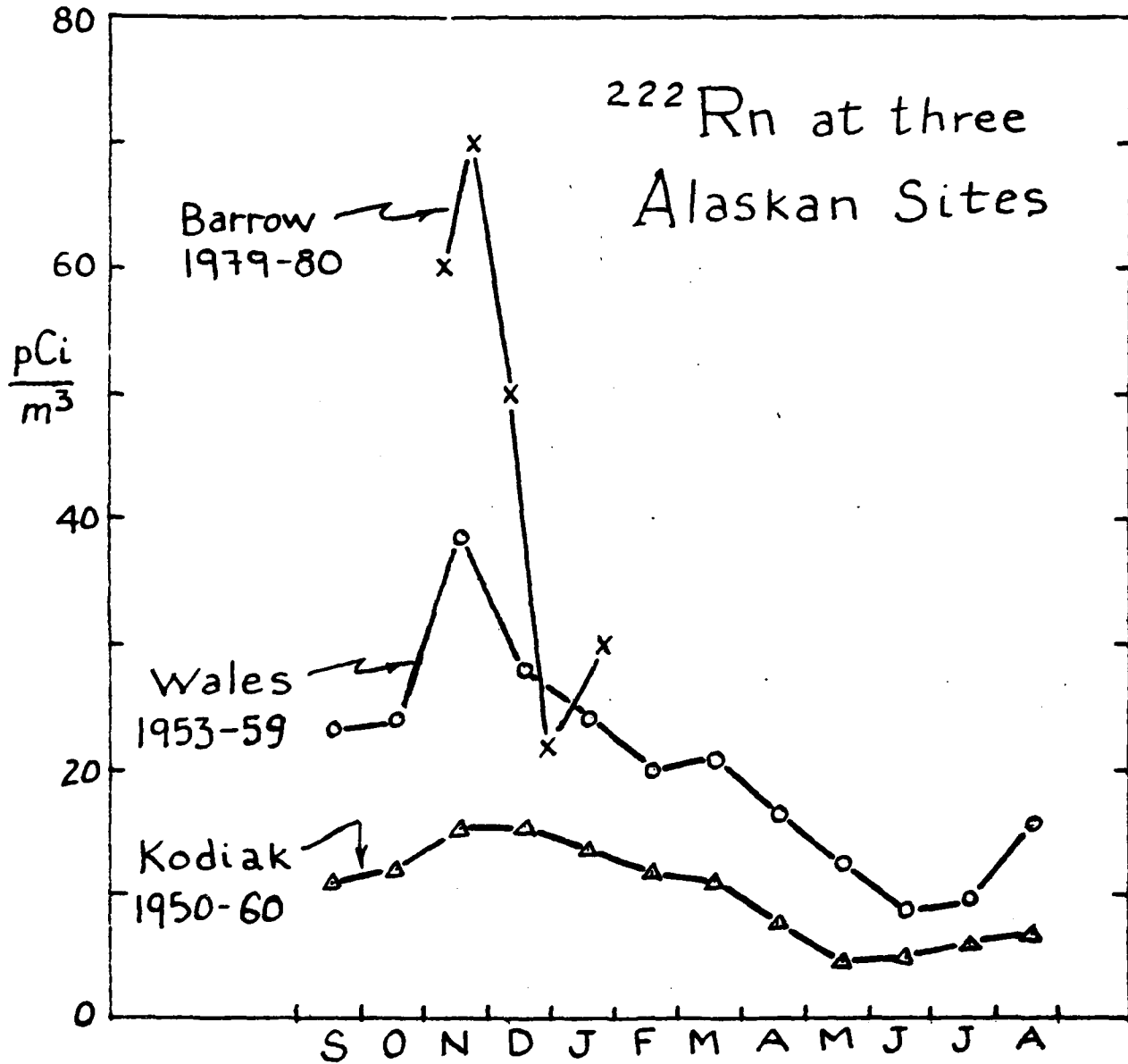
Figure 1. Monthly mean concentrations of the continental gas^{222}Rn at three Alaskan sites. The Wales and Kodiak data are from Lockhart (1962); Barrow data are from this Contract.

Our understanding of year-to-year differences in seasonal variations of Arctic aerosol is less advanced than is our understanding of geographic or elemental variations. Nevertheless, some features are beginning to stand out. The early-November onset of high concentrations of V and $SO_4^=$ at Barrow in 1976 seems now to be unusually strong and early. Later years had smoother increases, with high concentrations appearing only in December. Further, the January dip at Barrow was much less pronounced in 1978 than in 1977. Both these observations are consistent with the winter of 1976-77 being the most abnormal in 100 years. As data become developed for 1978-80 at Barrow, we will be able to place this first winter of sampling into a better perspective. (It has also been pointed out to us by Harry van Loon of NCAR that the first three winters of our sampling at Barrow, 1976-77 through 1978-79, formed a smooth progression from strongly meridional to strongly zonal circulation in the Northern Hemisphere. We will be looking for related trends in air chemistry at Barrow.)

Some very interesting elemental differences in seasonal variations in the Arctic are beginning to appear. The first to be noticed was a 2-4-week later spring maximum for $SO_4^=$ than for V, which is now atributed to late-spring photochemical oxidation of SO_2. (The $SO_4^=$/V ratio shows a distinct relation to the amount of sunlight in the Arctic.) Next, we noticed a November-December maximum for ^{210}Pb, not a spring maximum (Rahn and McCaffrey, 1979b, 1980; Sections I.E.2 and 4 here); this is now seen to be accompanied by a similar maximum in ^{222}Rn (Figure 1). Recently, early-fall rises of Mn have been noted for Barrow and Mould Bay (Barrie et al., 1981), and fall rises of darkness of filter and elemental carbon at Barrow (Rosen and Novakov, 1981; Patterson and Rahn, 1981). We are not yet sure of the significance of any of these fall maxima, but we suspect that they are related to locations of aerosol sources affecting the Arctic, as well as seasonally varying transport paths. As data for other elements are built up for the Arctic aerosol, we hope to construct a better chemical profile of these additional sources.

Perhaps it should be pointed out that, in spite of all the intriguing new data on seasonal variations now appearing, no satisfactory explanation for the basic spring maximum in the Arctic has yet been proposed! We attempted this about a year ago, without real success (Rahn and McCaffrey, 1980; Section I.E.2 here). G. Shaw has recently (Shaw, 1981) proposed that cloudiness, not precipitation, accounts for the seasonal variations. This question should be answered before other seasonal patterns are considered.

A URI paper on seasonal variations in the Barrow aerosol will be prepared for Atmospheric Environment.

3. Sources of the Arctic aerosol (URI)

In 1976 we realized that at least the V of the Arctic aerosol had to be derived from pollution sources far from the Arctic. In 1978, with the advent of sulfate data for the Arctic, we concluded that much of the sulfate, hence much of the Arctic aerosol (of winter) was also pollution-derived. In the last two years, we have seen nothing to change our mind on this point. If anything, the geographic, seasonal, and elemental patterns of the Arctic aerosol point toward a pollution source more strongly

than ever. Only the specific source areas are still in question. [Pollution may even account for much of the summer sulfate, previously thought to be natural, via photochemical oxidation of the long-lived gaseous pollutants COS and CS_2.]

Over the last 2-3 years there have been two important trends in our thinking concerning which pollution sources are the most important for the Arctic. The first is about the relative importance of North America vs. Eurasia. In 1976-77 we looked, perhaps naïvely, to eastern North America as the probable main source of pollution to the Arctic, via the North Atlantic pathway. The weight of evidence since that time has forced us to change our mind, however. As of early 1979 we recognized that there was too much ^{210}Pb at Barrow to have come from North America via the North Atlantic; a Eurasian source for SO_2 and V, coupled with a pathway through the western USSR, fit the Barrow data much better. The $SO_4^=/V$ ratio at Barrow was more compatible with European conditions than with North American, also. Lastly, there were glimmerings that the proportions of Mn and V at Barrow were more like Eurasian than like eastern North American conditions (Rahn and McCaffrey, 1980; Section I.E.4 here). Later, as more data from the Arctic Air-Sampling Network became available, empirical maps of the geographical distributions of $SO_4^=$ and V in winter strongly indicated a Eurasian, not North American origin for the Arctic aerosol (Rahn and McCaffrey, 1979b; Section I.E.2 here). Theoretical reasons for this lack of transport over the North Atlantic were proposed in early 1980 (Rahn, 1980).

The second major progression in our thoughts about sources was eastward within Eurasia. In 1976-78 we considered the main question to be North American vs. Europe. In late 1978 we recognized that a pathway through at least the western USSR seemed to be necessary to account for the high ^{210}Pb at Barrow (Rahn and McCaffrey, 1980; Section I.E.4 here). No pollution sources within eastern Europe or the USSR were considered yet, however. In late 1979, simulations of SO_2 amd $SO_4^=$ along two pathways leading to the Arctic from western Europe, eastern Europe and the western and central USSR showed that the USSR could no longer be neglected as a source (Rahn, 1979). As a result, we now consider the Arctic aerosol to originate from some combination of European and USSR sources. We also recognize that when an air mass passes over the highly industrialized area of the southern Urals in the central USSR, all previous non-USSR sources become unimportant. The greater Mn/V ratios in the North American Arctic than in the Norwegian Arctic may be evidence of the effect of these USSR sources.

Our ideas about the dominance of Eurasian sources for the Arctic aerosol are not universally accepted, however. The most recent dissenter is Dr. E.R. Reiter of Colorado State University, who concluded form his study of planetary waves and V at Barrow (funded by a $3000 subcontract from URI in FY80) that the most likely source was New England. We consider such debate healthy. Copies of Dr. Reiter's lengthy report are available from URI.

4. Transport of aerosol to the Arctic (URI,UA)

Our study of the Arctic aerosol involves understanding long-range transport of aerosol (and trace gases) on the scale of 5,000 to 15,000 km. Although there has been study of this scale of transport for

radionuclides and volcanic aerosol in the stratosphere and upper troposphere, to the best of our knowledge there have been no studies of such long-range transport of aerosol, especially of pollution aerosol, in the lower troposphere. Add to this that the bulk of the transport to the Arctic may occur in the lowest 2-3 km only, and there is clearly no precedent to what we are trying to understand and model. The situation is further aggravated by the fact that much of the transport of the Arctic takes place over highly remote, largely uninhabited areas from which few meteorological observations are available, and where it is very difficult to establish monitoring stations. For these reasons we have taken a very broad, general approach to understanding long-range transport to the Arctic, and have concentrated initially on identifying its major meteorological features rather than becoming involved with unverifiable detail. We now discuss these major features, some of which have been mentioned above.

Neither the North Pacific nor the North Atlantic seems to be a major pathway of aerosol from midlatitudes to the Arctic. This eliminates Japan/Korea/China and the eastern United States as important sources. Transport of aerosol can be seen across the North Atlantic, but the amounts of aerosol are small enough to be easily masked by Eurasian emissions. No SO_2 has been observed to cross the North Atlantic. There seems to be less transport of pollution across the Pacific than across the Atlantic, because of the former's greater dimension, weaker sources, more southerly placement of sources and less direct pathways to the Arctic. Even for the Atlantic, close examination of meteorological maps seems to reveal far fewer cases of transport of air from the northeast United States eastward, then northward over Iceland to the Arctic, than we had expected. Air from the United States usually winds up only about 10° farther north, in Europe rather than in the Arctic. In the vicinity of Iceland, air is usually traveling southward, not northward during winter.

In the broadest sense, we attribute the lack of transport of aerosol to the Arctic via marine pathways to the relatively great precipitation and turbulence over oceans in winter, as well as to the southerly placement of pollution sources on the western sides of both oceans. By contrast, Eurasian sources are located 10-15° farther north; their emissions travel over continents rather than over oceans on their way to the Arctic, hence are exposed to much less precipitation en route. If, as we believe, the removal of aerosols in and around the Arctic is controlled largely by the amount of precipitation, aerosol from Eurasia is removed much less quickly than is aerosol from eastern Asia or eastern North America, hence can reach the Arctic in greater concentration. Once aerosol is in the Arctic, the slight degree of precipitation there during winter assures it of a long lifetime, and allows it to mix over the entire Arctic if circulation permits. Thus, long residence times in and around the Arctic in general, and particularly during winter, are a key element in our understanding of transport.

We are not yet sure just how long residence times of Arctic aerosol are during winter. If they were controlled exclusively by precipitation, they could easily reach 20-50 days or more. Dry deposition may reduce these figures. Transit times, on the other hand (not to be confused with residence times, which are the inverse of the rate of removal), appear to be 10-20 days. We would like to determine experimentally values of residence and transit times by radionuclide measurements; we are presently evaluating whether this is possible.

There is a variety of pathways for transport from Eurasia. From Europe proper, aerosol can flow anticyclonically via the Norwegian Sea, due northward via the Baltic Sea, or cyclonically via the western USSR. From the western USSR it generally flows cyclonically, under the influence of the Icelandic low. From the central USSR it can flow anticyclonically, northward, or cyclonically, depending on the positions of the Icelandic low and the Asiatic high. Much of our work on transport in the near future will be devoted to determining the relative frequencies and efficiencies of these various northward pathways from Eurasia.

It was only in late 1979 that the existence and importance of the return-flow pathway from Eurasia to the Norwegian Arctic was recognized. This came from a study of the Bear Island aerosol while KAR was spending 3 months at the Norwegian Institute for Air Research. To create this pathway, air flows cyclonically around the Barents low, the eastward extension of the Icelandic low, and brings high concentrations of aerosol and SO_2 to Bear Island and Spitsbergen from the northeast (Figure 2). The existence of this pathway was first proposed by Rahn (1979) to account for 10 of 13 cases of high SO_2 at Bear Island during the winter of 1977-78; its existence was confirmed by trajectories calculated by Mr. Thomas J. Conway in early 1980 (Rahn et al., 1980). We consider this an extremely important pathway for understanding air pollution in the Norwegian Arctic. It probably also plays a sizeable role, but not an exclusive one, in determining air quality in the North American Arctic.

There has been much discussion recently about the altitude of transport of aerosol to the Arctic. While Arctic haze occasionally extends to the tropopause (Mitchell, 1956), its maximum altitude is more normally approximately 5 km (Ptarmigan data). This does not necessarily mean, however, that aerosol observed in the boundary layer of the Arctic has actually reached these altitudes during transport. On the basis of numerous reports in the literature, we suspect that air above the Arctic surface inversion (top at 1-1.5 km) is decoupled from that below it, hence may be of different composition, different pathway, and different origin. This would imply that aerosol observed near the surface in the Arctic could have traveled a long distance within the surface layer. Because similar inversion conditions are found over the (snow-covered) Eurasian source regions during winter, transport near the source would be low-level as well. Thus, it seems quite possible to imagine aerosol traveling a large fraction of the way from Eurasia to the Arctic within the surface boundary layer of the atmosphere. Although this idea fits the facts of the Arctic as we see them, it breaks all the "rules" of long-range transport and is consequently not generally accepted. Unfortunatley, we do not see an easy way to test it.

Another topic of long-range transport with which we must deal is the degree of large-scale dilution of polluted air masses by outside air. We have used dilution factors up to 6, but they are only guesses. In actual practice they are "fudge factors", used to adjust simulated Arctic concentrations to the observed concentrations. At present, we are not aware of any way to estimate large-scale dilution independently and accurately.

Our models of long-range transport are expressed as simulated aging of air masses, usually for 10-20 d. Aging diagrams are given in Rahn and
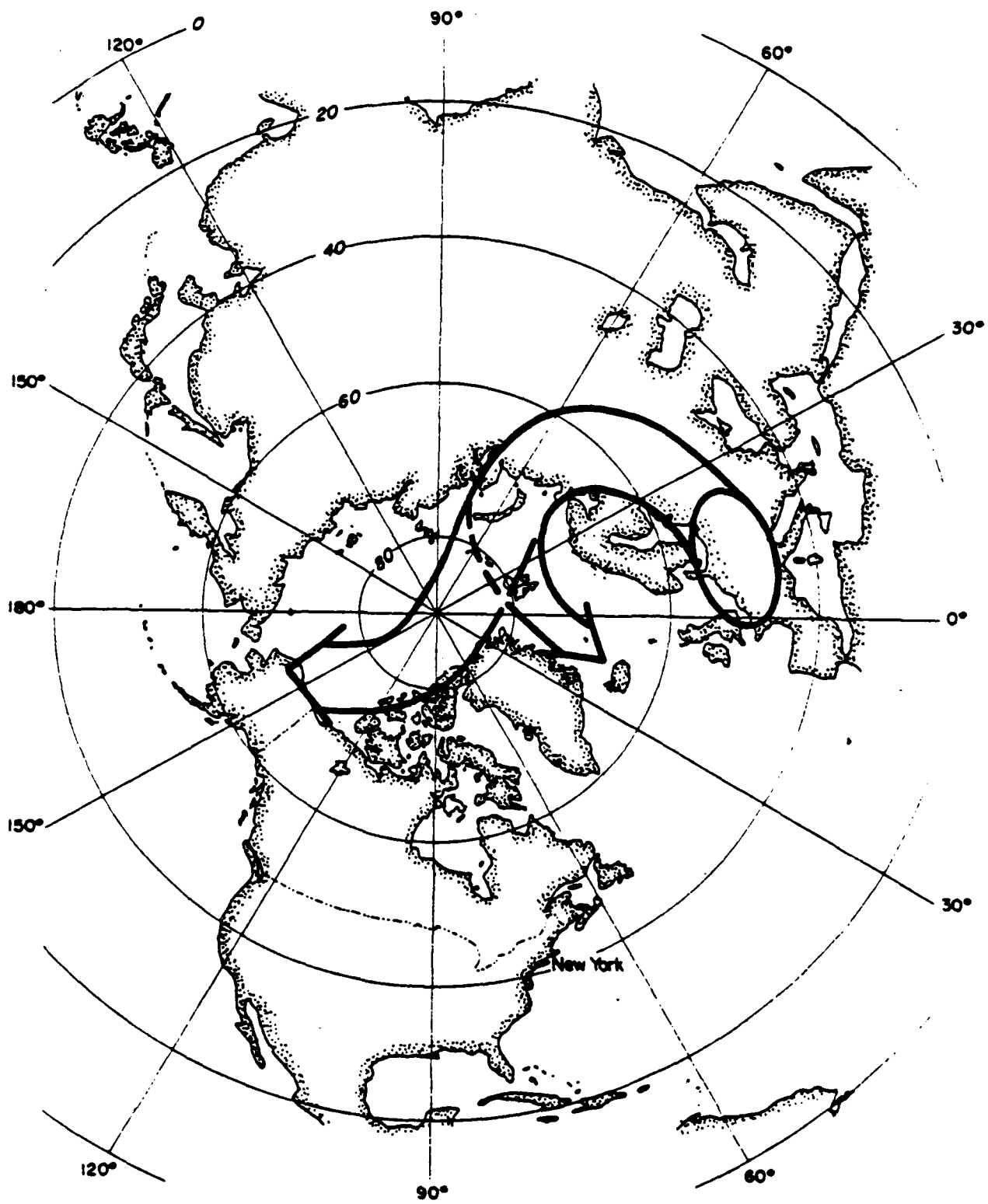
Figure 2. The return-flow pathway from Eurasia to the Arctic.

McCaffrey (1980; Section I.E.4 here), Rahn (1979) and Rahn (1980).
Further results of simulated aging are given in Rahn and McCaffrey
(1979b; Section I.E.2 here).

A novel approach to understanding transport of aerosol the the Arctic
has recently been offered by Shaw (1981). He proposes that eddy diffusion,
rather than organized motion, controls transport in the Arctic on the time
scales of 5-20 days that we are dealing with. The basis for this work is
the idea that organized motion of air masses (hence the trajectory approach
to understanding sources of Arctic aerosol) disappears when the lateral
scale of eddy diffusion from a point source reaches the mean size of
the basic system of air flow in and around the Arctic. If the latter is
taken to be a synoptic system, and a characteristic distance of spread
due to eddy diffusion is given by $x = \sqrt{Dt}$, where D is the coefficient of
horizontal eddy diffusion, x may reach 500-1000 km in the 10-20 days of
travel one usually associates with transport from midlatitudes to the
Arctic. In a simple model, Shaw estimates the concentration of $SO_4^=$ at
Barrow from estimated mean concentrations at 50°N, 10% frequency of north-
ward injection, eddy diffusion alone north of 50°, and various residence
times. The model predicts 6.9 µg m^{-3} $SO_4^=$ at Barrow, about three times
higher than the maximum monthly mean of 2.2 µg m^{-3} for March. Considering
the simplicity of the model, this agreement was considered very good.
An interesting finding of the model was a similarity between the seasonal
variation of Arctic aerosol and the timing of cloudiness in the Arctic,
from which Shaw hypothesized that clouds, not precipitation, may be the
major remover of Arctic aerosol. The eddy-diffusion model is being written
up for submission to Atmospheric Environment.

The question of eddy diffusion vs. organized flow for air flow to the
Arctic is an important one, and is sure to be discussed much more in the
future.

5. Cloud-active aerosol in the Arctic (CSU)

This is the Ph.D. thesis topic of Mr. Randolph D. Borys
of the Department of Atmospheric Science of Colorado State University.
Our Contract supports one-half of Mr. Borys' graduate stipend and most of
his Arctic field work.

Mr. Borys has nearly completed his summer and winter Arctic field work,
as discussed in Section I.B.9 above. Overall, the field programs have pro-
ceeded smoothly and all the data sought were obtained. Spitsbergen and
Barrow were the most polluted sites during winter. At Spitsbergen, an
episode of pollution from Eurasia was easily visible. Iceland was very
clean in summer, except for an episode of pollution from Europe. At least
during the short stay at Pangnirtung, Baffin Island, which is along the
proposed pathway from eastern North America to the Arctic via the west
coast of Greenland, this pathway was not an active one. The aerosol at
Poker Flat (outside Fairbanks) was very different from that at Barrow,
especially during winter. During the winter visit to Poker Flat, it was
35°C (60°F) warmer than Barrow. Thus, Poker Flat and Barrow were in en-
tirely different air masses. This apparently held true for much of the
rest of winter of 1979-80 as well, which was the third warmest in the his-
tory of Fairbanks, and, according to G. Shaw, free of Arctic haze.

The results of Mr. Borys' summer 1979 field study in Iceland are being prepared for submission to Atmospheric Environment; a preprint is included here as Section I.E.6. It forms an interesting case study. Concentrations of cloud-condensation nuclei (CCN) paralleled those of noncrustal vanadium and nonmarine sulfate, two indicators of pollution aerosol. Condensation nuclei (CN) did not parallel CCN, however. This suggests that CCN over Iceland, but not CN, are derived from large-scale air pollution. During the first days of sampling, air came from North America. There may have been weak transport from North America, but this is still uncertain. During the middle of the sampling period, air came from the vicinity of Greenland and was very clean. During the last part of the period, the air came from Europe, approximately 3500 km away, and was the dirtiest of the period: there was visible haze; aerosol increased by factors of 5-10. If CCN can thus be transported 3500 km in summer, they probably can reach the Arctic in winter.

6. Radiative effects of the Arctic aerosol (UA, URI)

During the last two years, GES and KAR have attempted to evaluate potential direct radiative effects of the Arctic aerosol on the Arctic atmosphere, sponsored in part by a one-year grant from NSF. A two-year grant has just been received from NSF for further work.

There seems to be little doubt that the Arctic aerosol heats the local atmosphere rather than cools it, during spring. This is because of the great absorbing character of the winter Arctic aerosol, which in turn stems from its high concentration of black carbon. Concentrations of black carbon in the Barrow aerosol of March are estimated to be 0.3 µg m^{-3} (Rahn and Shaw, 1981), many times higher than expected for such a remote area and even several times higher than would be estimated from the concentrations of other pollutants at Barrow. One thing is certain: it is not a local effect.

The first estimates of radiative effects appeared in Shaw and Stamnes (1980), where the heating rate in the lowest 2 km of the Arctic atmosphere during spring was estimated at roughly 1°C per day. This is a significantly large heating rate. Using updated chemical and radiative properties of the Arctic aerosol, Rahn and Shaw (1981) estimate the heating rate to be 0.2 to 1.0°C per day, with a most probable value near 0.5°C per day. This is still a significant rate, for it is as large as the heating rate from all trace gases in the Arctic combined. It should be noted, though, that heating rates are much less in other seasons, when the sun is down or the aerosol is much less concentrated. We are very interested in the potentially important heating by the Arctic aerosol, and continue to use properties of the Arctic aerosol measured under this Contract as input for radiative calculations (see Section I.B.11 above).

7. Deposition of the Arctic aerosol (URI)

Although studies of deposition of Arctic aerosol to the Arctic Ocean and surrounding tundra are not a part of our Arctic Haze Contract per se, nor are they likely to become so in the foreseeable future, deposition is so much a part of the Arctic environment, and is potentially so important to ecological, oceanic, and radiative/climatic concerns of the Arctic that we must mention our thoughts here.

Deposition of aerosol in the Arctic is presently a very confused topic. Trace elements in ice and snow of the Greenland Ice Sheet have been studied for many years in an effort to obtain a historical view of contamination in the atmosphere, with diverse and often contradictory results. The state of the art is illustrated by the three-part exchange of letters in Nature, 10 April 1980. There seems to be little agreement on which data, if any, from the Ice Sheet are reliable, and how the reliable data are to be interpreted.

The situation is only a little better at Barrow. Here at least, reliable data on the aerosol exist. But there is only one published set of data for snow near Barrow (Weiss et al., 1978), and they give an entirely different, natural picture of the Barrow environment than does the Barrow aerosol (Rahn and McCaffrey, 1979a; Section I.E.1 here). For the last year K. Rahn and R. McCaffrey have had an NSF grant to study possible aerosol-snow fractionation as it might occur in the Arctic. The problem has not yet been resolved, however, and a follow-up grant is planned.

Thanks at least in part to our efforts, the polar-science community now recognizes that neither the relation between aerosol and snow nor the historical interpetation of trace elements in ice cores is as straightforward as was once thought. The French group at Grenoble (C. Boutron, C. Lorius, etc.) has grasped these facts the most quickly, and is spearheading a French-American effort to hold a workshop on the relations between polar aerosol and snow, to be held in Grenoble in May 1981. KAR is organizing the American side. We anticipate extremely difficult and hard-hitting, but valuable, discussions at this workshop.

Achieving a basic understanding of deposition of aerosol in the Arctic is complicated by at least three major factors: (1) The small amount of precipitation--an order of magnitude less than in midlatitudes;; which may make the relation between wet and dry depositon differ from that of midlatitudes; (2) The very low concentrations of aerosol--1-2 orders of magnitude less than in midlatitudes--which makes it difficult to extrapolate depositional behavior from midlatitudes; and (3) The out-of-phase, order-of-magnitude variations of aerosol and precipitation. In winter and spring, when aerosol is the most abundant, it precipitates at least in the Arctic. In summer, when aerosol is the least abundant, it precipitates the most. For these reasons, we have been very reluctant to estimate the amount of deposition into the Arctic. A recent opportunity was provided, however, by the March 1980 London Conference on the Arctic Ocean, at which KAR presented a paper on the Arctic aerosol and its possible effects on the Arctic Ocean (Rahn, 1980). Rough estimates of annual deposition of several trace species to the Arctic Ocean were made here, and compared to annual inputs of these same trace species to the Arctic Ocean from rivers and from the influx of water from the Atlantic and Pacific Oceans. These estimates were refined and extended, and will be submitted for publication in Atmospheric Environment (Rahn, 1981). Briefly, they show that most of the aerosol should be deposited in winter, when the aerosol is most concentrated, even though precipitation is least then. Because this winter aerosol is strongly pollution-derived, the effects of air pollution in the Arctic should be felt in Arctic snow, although to a lesser degree than in the aerosol itself.

The calculations also showed that the atmosphere should be the dominant source of at least Pb to the Arctic Ocean. This conclusion was made possible by very recent measurements of Pb in oceans and rivers, which show the true concentrations to be very small (Settle and Patterson, 1980). In rivers, for example, the concentrations of Pb are some 200 times lower than found by other investigators. One wonders how many other elements are like Pb, and have true concentrations in various waters which are much lower than are presently recognized. Such elements would probably also have the atmosphere as their main source to the Arctic Ocean.

The question of deposition of trace elements to the tundra will soon become very important. Arctic lichens, the prime source of winter food for the caribou herds, derive their water and nutrients from the atmosphere. As pollution in the Arctic atmosphere increases, with attendant increases in acidity and various trace elements which can be toxic at high concentrations, such as Pb, Cd, As, Sb, Se, etc., the lichens will presumably absorb increasing amounts of these species. The effect of this is completely unknown, but the stakes may be high and the problem should be investigated. Another potential concern is the effect of acidity on the nitorgen-fixing blue-green algae of the Arctic. We are presently in contact with several interested individuals, such as Edmund Schofield of The Institute of Ecology in Indianapolis, Wayne C. Hanson of Battelle Pacific Northwest Laboratories, Richland, Washington, and Miss Marianne Krasny, a graduate student at the University of Washington, who wishes to do a Master's thesis on Arctic lichens. We suspect that some sort of project on the tundra will emerge out of all this.

Deposition of pollution aerosol via snow may have radiative or climatic consequences as well, through darkening of the snow by the sooty carbon in the aerosol. There is now evidence that the albedo of Arctic snow is a few percent less than that expected for pure snow, with the most reasonable explanation being the presence of sooty carbon taken up from the aerosol (S.G. Warren and W.J. Wiscombe, NCAR, in two articles submitted to J. Atmos. Sci.). The amount of carbon needed to account for the apparent change in albedo seems to be consistent with amounts predicted by scavenging from Arctic aerosol. Black carbon has never been directly measured in Arctic snow; H. Rosen of Lawrence Berkeley Laboratory is very interested in this problem, and will try to confirm the predicted concentrations by direct measurement in late 1980 or early 1981. This question of carbon in Arctic snow is potentially very important, because this carbon is exposed to the strong solar radiation of Arctic summer, when atmospheric carbon is long gone. Thus, radiative effects of carbon in snow are much longer-lasting than are those of carbon in the aerosol.

8.   Bromine in the Arctic atmosphere (URI)

For at least two years we have been aware of very high concentrations of Br in the Barrow aerosol during spring. These concentrations, typically 50-100 ng m^{-3} during March and April, are far too high to be explained by sea salt, general long-range transport of aerosol (even including gas-to-particle conversion of Br), volcanism, or the stratosphere. They are as high as in many large U.S. cities, like Boston. We don't have any idea where all this Br is coming from.

The problem was a frustrating curiosity until February 1980, when we learned from J. Miller of NOAA/ARL that during this same time of year, mid-February through mid-April, surface ozone observed at the Barrow GMCC

Observatory behaves very erratically, often disappearing for days at a time. NOAA/GMCC presently has no explanation for this; we wonder whether high concentrations of gaseous Br (presumably associated in some way with the high particulate Br) might not be destroying this ozone catalytically via the familiar cycle:

$$O_3 + Br = O_2 + BrO$$

$$BrO + CO = Br + CO_2$$

$$O_3 + CO = O_2 + CO_2$$
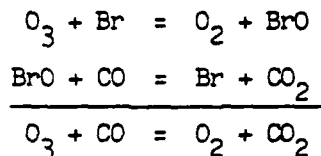
Both daily and monthly mean concentrations of O_3 and Br are negatively correlated at Barrow during spring. If Br and O_3 actually interact, this would be an extremely interesting topic to pursue.

There are several pieces of evidence that suggest a local Arctic source for this excess Br of spring. It is also observed at Mould Bay, both in the aerosol and in the snow (L.A. Barrie, personal communication), but not at Bear Island and in much smaller concentrations at Spitsbergen. At Barrow it is associated with cold air, both monthly and daily. It is not associated with V, i.e., is not from long-range transport.

The only reservoir of Br in the Arctic capable of producing such high concentrations in the atmosphere seems to be the sea. We are presently hypothesizing that Br may be released to the atmosphere during spring by ice flowers, efflorescent crystals of sea salt and water that grow from rapidly refrozen leads during nights when the surface temperature dips below -24°C, the precipitation point of NaCl from seawater. Ice flowers occur primarily during February, March and April (N. Untersteiner, personal communication), i.e., just the months when atmospheric Br is the most concentrated. Our calculations indicate that ice flowers contain great excesses of Br relative to what is needed to produce even these high atmospheric concentrations. The Br could be released photochemically from the needles of the ice flowers, as it appears to be lost from marine aerosol.

During spring 1980 Mr. R.D. Borys grew some ice flowers for us during his field work at Barrow. We analyzed three samples: fresh flowers grown overnight and harvested before they were struck by the sun's rays, flowers after 4-5 hours exposure to the sun, and flowers after 12 hours exposure. We found abundant Br in all three samples, in seawater proportions with Na. As a result, ice flowers can now be regarded as a potential source of Br. There was, however, no measurable loss of Br (within 10%) after 12 hours irradiation. This may not necessarily negate our hypothesis, however, for only a small loss of Br is sufficient to create high Br in the atmosphere.

At present we are not sure of where we stand. We realize that the ice-flower hypothesis is esoteric, and very possibly completely wrong. But it is our only reasonable explanation so far for an Arctic environmental occurence that is so bizarre that it will surely yield big dividends to whomever can look into it. During the next three years we would like to follow up these initial studies if manpower is available.

9.   Trace elements in size-fractionated desert soils (Mainz, URI)

This study, undertaken with Dr. L. Schütz in 1977, was moti-
vated by our finding of Asian desert dust over Alaska in 1976. Its ultimate
goal is to determine whether aerosol from various deserts can be discrimi-
nated chemically through tracer elements. In order to check this, a more
fundamental study of elemental composition vs. particle size was necessary,
for a variety of soils from deserts and other locations. The basic results
which are coming out of this study are the following:

(a) Elemental concentrations are nearly constant with size in agri-
cultural, or fresh soils (Cotton Tower Field; fallow land of the Sudan).
In other, older soils, there is an increase of elemental concentrations
towards smaller particle sizes. The magnitude of this effect increases
with degree of weathering.

(b) Many elements have the same basic pattern, with low concentra-
tions above radius 60 μm, increasing concentrations from 60 to 20 μm, then
a broad plateau of concentration for smaller radii. This means that elemen-
tal compositions, hence enrichment factors, are nearly constant over the
aerosol size range. Thus crustal reference materials used for enrichment-
factor calculations need not be size-dependent, to a first approximation.

(c) The diluent in the large sizes is SiO_2. Hence the concentration
of Si increases with size.

(d) A few elements like Na, K and Ca are present in minerals of all
size ranges, hence are quasi-uniformly distributed with particle size.

(e) A few puzzling elements like Zr, Hf and to a lesser extent the
rare earths, have a maximum in concentration and enrichment factor in the
middle of the size range. We hypothesize that these elements are found in
resistive minerals of small grain size in the original rock, and are basi-
cally unchanged by weathering. They thus accumulate during weathering,
and can reach extremely high concentrations in certain highly weathered
soils.

(f) Soils from different deserts are suprisingly similar in compo-
sition, especially in the aerosol size range. Because of this, it seems
unlikely that tracer elements for aerosol from individual deserts will
be found.

(g) The other side of the ooin is that this homogeneity supports
the use of a single crustal reference material for aerosols from different
sources, at least from different deserts.

(h) This work confirms the use of Mason's (1966) rock as a crustal
reference material (Rahn, 1976).

To supply additional detail on the above points, we include as the
next few pages a set of notes made by KAR of discussions with Dr. Schütz
in Mainz during March 1980. These were meant for internal use only, hence
are a bit rough, but may be of some use here.

Outline of main results of L. Schütz soil analysis.

1. **Evidence of elemental fingerprints for concentration, relative mass distribution and enrichment factors.**

   A. There is a great deal of redundancy in the elemental measurements. The same fingerprint was found for most elements (calcium was an exception.

   B. Cause:
   (1) Seems to be related in a gross way to the mass distribution (at least for relative elemental mass).
   (2) Seems to be caused to a certain extent by physical and chemical weathering.

2. • There is usually an increase of elemental concentration towards smaller particle sizes.

   • The magnitude of this increase (or slope of the plot of relative concentration) seems to be related to the extent of weathering.

   • It is greatest in dune-like (desert-like) soils, (S-12, S-24, S-29, S-33).

   • It is smallest, and nearly flat, in the garden soils (CTF-7, fallow land).

   • There are intermediate cases which seem to correspond to soils where there is presently active weathering and physical removal of particles (Texas, Anthony, Harmada, S-2, S-10, S-39, Rock desert).

   • There are more recent additional soil samples as part of Guillame's thesis, such as from the Sudan, from the Bilma area of Niger (which is supposed to be one of the most active areas of dust storms in the Sahara), river floodlands (Niger, Senegal).

     Mass distributions of new soils appear to fit well into the above picture. Specifically, the floodlands appear to be fresh soils like CTF. Soils from Bilma appear to be intermediate in character, approaching characteristics of dunes. They come from areas near mountains where mountain debris is collected on high plateaus, where there are no rivers. The soils close to Toumboucto and Mali are very aged in character, they come from small dunes with very coarse mass distributions.

   • Lothar believes that river beds are very important sources of desert aerosol. These beds are typically covered by water only once a year. This reminds us of Yaalon's wadis Many people believe that the Erg of Bilma is a big source of desert dust, but Lothar does not believe this. Instead, the aerosol appears to come actively from the mountain areas north and northwest of Bilma and the Tibesti mountains.

   • The increase of concentration of elements with decreasing radius could also be explained by an increasing number of mineral species in the smaller size range. The largest soil particles have relatively few mineral species such as silica, dolomite, calcite, etc.; the smaller particles have a larger variety of species such as clays.

- The size range where the strongest increase in concentration in most elements is found is roughly r≈20-60μm. There is a broad plateau of concentrations for radii less than about 30μm.

- We have expressed our results in three types of plots: (a) concentrations (ppm) vs. radius, (b) relative concentrations vs. radius, where the relative concentration is $m^*(r)$ x ppm (the form of the plot of relative concentration is determined mainly by the mass distribution of the soil), (c) enrichment factor vs. radius.

3. Possible explanation of the enrichment factor plots.
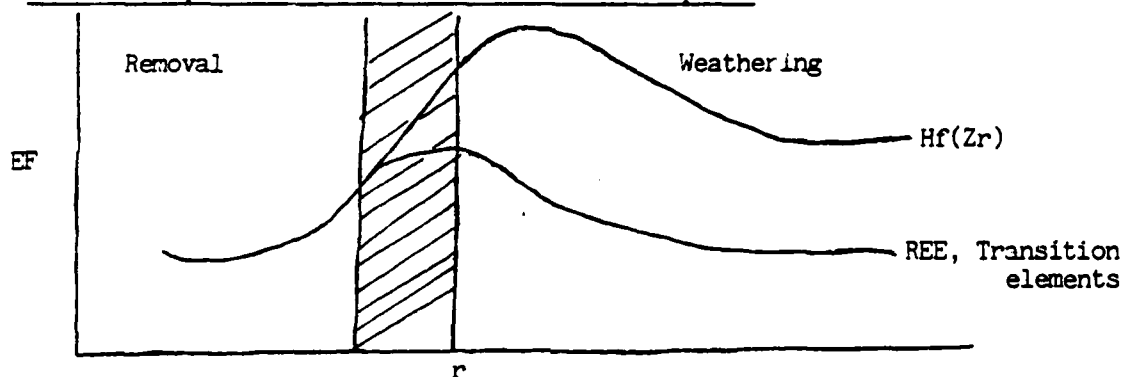
These elements have a mass distribution different from the others in that they have a puzzling peak in the middle of the size spectrum. This peak can be very large in certain highly weathered soils. The elemental mass usually peaks at radii 20-50μm. The enrichment factor also peaks here, presumably because the distribution of Al is different from those of these elements. Lothar proposes that these elements occur in specific minerals which strongly resist weathering. An example would be zircons, which contain zirconium and hafnium. If this is correct, and if the minerals have a relatively small grain size, the lower side of the peaks in this plot represent the upper limits of removal of particles from the soil by water and air, as shown in the plot above. According to this picture the resistive minerals, being of small abundance in the original rock, have small grain sizes. During weathering these grains, because they are more resistant than the rock around them, are released as individual particles with a certain size distribution, with a peak at 20-50μm. Weathering of the bulk rock leaves these tiny grains basically unaffected. They can only be removed from the soils by wind or water.
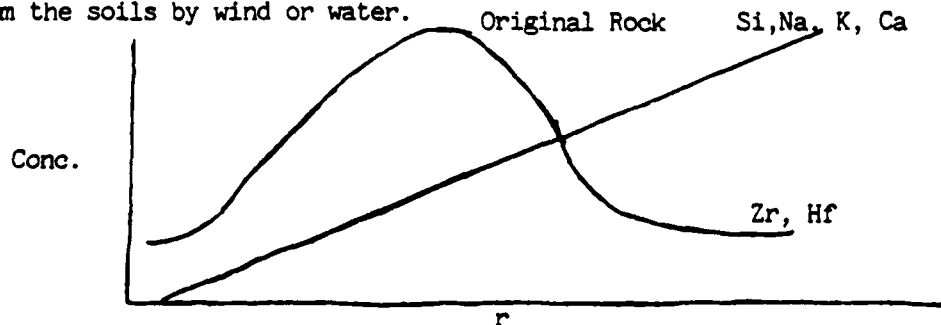
Types of elements. (a) Very resistant, major-Si; (b) Very resistant, minor-Zr,Hf; (c) Weatherable, leachable - Na, K; (d) Weatherable, clay-forming (less soluble)-Al; (e) The more weatherable, not so soluble elements tend to be found in clays.

WEATHERING
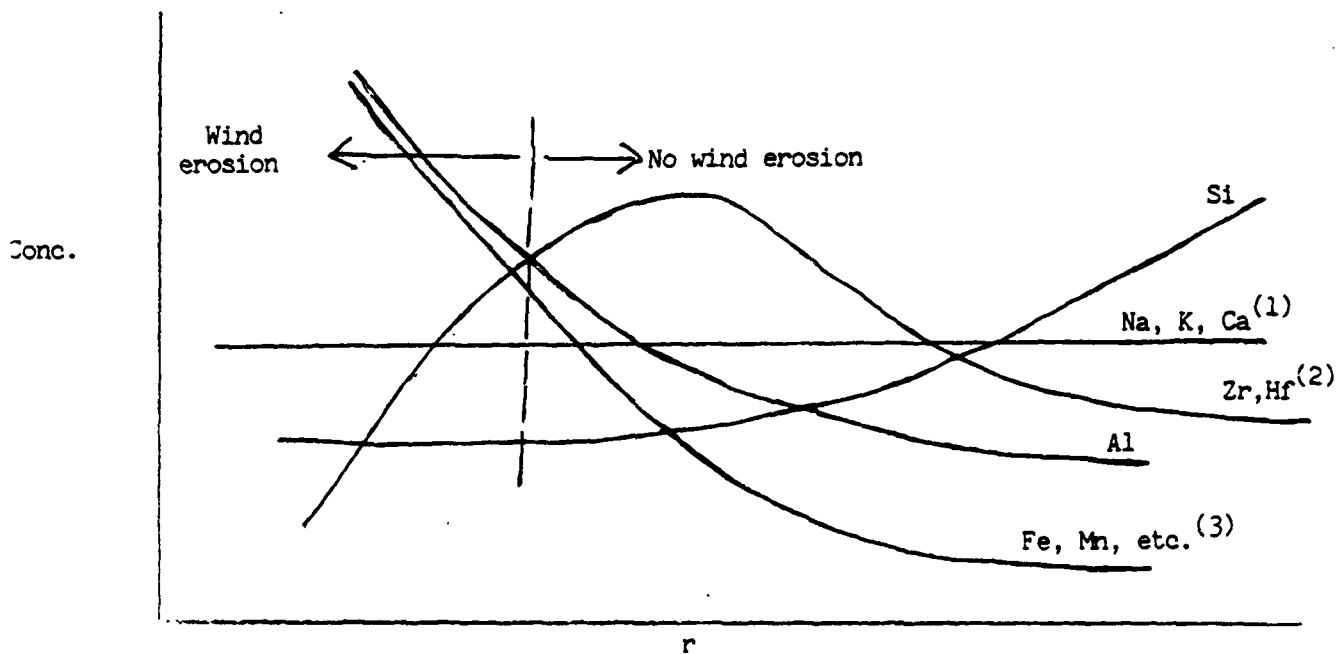
(1) Weatherable, leachable elements.
(2) Unchanged by weathering
(3) More soluble than Al, but strongly participating in clays.

WEATHERING & EROSION
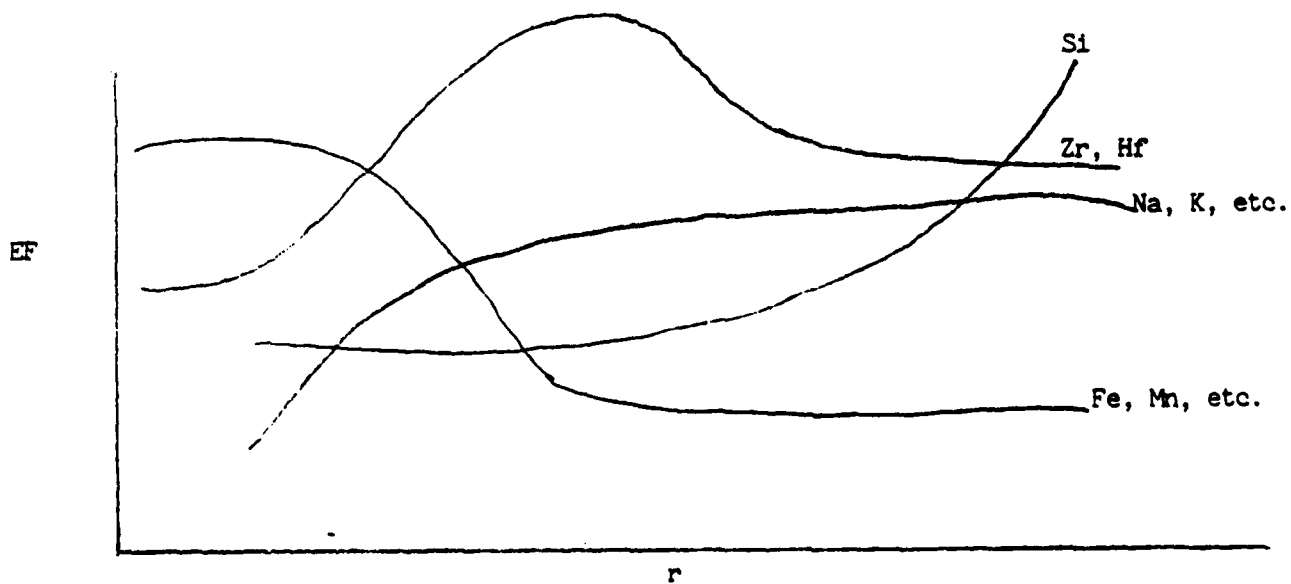
4. <u>Interest to the Navy</u>.

A.   Desert aerosols are found in atmospheres all over the world including those over oceans and those over the Arctic. The amount of aerosol released annually from deserts has been traditionally underestimated. For example the SMIC report assigned a source strength of 5×10^6 tons aerosol per year to the world's deserts, whereas Lothar Schütz, in the New York Conference, proposed $1,000 - 3,000 \times 10^6$ tons as a better figure, and this was only for the fraction of desert aerosol which was transported over long distances. Since the NY conference in 1979 there has been another upward revision: Lothar estimated 3×10^6 tons of Saharan dust transported annually from the Sahara eastward toward the Caucasus; the proper figure now seems to be $20-30 \times 10^-$ tons per year.

This desert dust has a variety of potential effects in the atmosphere. It is an absorbing aerosol and so heats the layer of air in which it is found. This can affect not only temperatures but stability structure within the atmosphere. Desert aerosol, even though quite large in size, is so abundant in the atmosphere that it makes the atmosphere quite turbid. Lastly, desert aerosol, through its nucleational properties, may effect cloudiness and precipitation over large regions of the world.

B.   Potential use of desert aerosol as a tracer of individual deserts.

One major surprise in this work was that desert soils, even those from different deserts, were surprisingly similar in composition, especially for particles smaller than $60 \mu m$ radius. A good example of this is seen in the plot for Hf. Consequently, major variations in bulk desert soils do not necessarily imply that aerosols produced from these soils are correspondingly different in composition. As a result of this we can draw several conclusions.

(1) There seems to be only little hope of using the chemical composition of desert dust, especially of the fraction which is transported over long distances, to discriminate between deserts. This conclusion has been derived from analyzing 6 soils from the Libyan Sahara, 2 soils from North American deserts and 2 cultivated Sudan soils. Even though this sample set is not particularly large, the correspondence between the aerosol-size fractions of these soils is great enough to suggest to us that analysis of further soils is not likely to change this conclusion. Of course, this conclusion has been based only on those elements which we have been able to analyze so far. It is possible that exotic enriched trace elements for this would include gold, silver, antimony, etc. These elements are most abundant in the smallest particle sizes. Future investigations aimed at deduction of sources should concentrate on enriched elements like these. There is a problem here, however: these elements are enriched in the fine fraction of the aerosol by other atmospheric processes. Because of this, these elements may not necessarily be uniquely attributable to deserts, even in areas of concentrated desert dust.

(2) The concept of a single crustal reference material for the atmosphere and aerosol is strengthened by the homogeneity for this size range in desert soils. We need to look further for the basic explanations of this homogeneity. Lothar intends to follow this up. There may well be exceptions to this homogeneity, however, especially in regions close to single major sources of desert dust.

(3) Mean enrichment factors of Sahara soils versus those of Mason's crustal rock. Which features of the compilation does this work seem to explain? Much of this information is contained in Lothar's big table of mean enrichment factors.

.
. (to be developed)
.

Note from this table that both the bulk soils and the fraction of radius less than 16μm have about the same concentrations when all samples are grouped together as when only the Sahara samples are considered. Lothar will send another big table in which he will calculate mean enrichment factors and compare them with those in the compilation.

(4) The future.

(a) One of the principal uses of this huge data set will be to refine and streamline future analysis of desert soils, by eliminating redundant or uninteresting elements.

(b) Most future work will take place at Institut für Meteorologie, not at URI.

(c) It is planned to analyze more soils from various deserts. Analysis will probably be restricted to two size ranges, 0-10μm and the bulk. They will be analyzed chemically, possibly by X-ray fluorescence. One goal is to get a reliable average concentration for Saharan soils. A second goal is to see stability (?) of ranges of elemental enrichment factors. Lastly, fewer samples will be used from the other deserts. Other deserts to be investigated include those in Australia and China.

5. New global reference material?

Most elemental concentrations in the bulk soils are close to those in Mason's rock data. They are however, quite variable from sample to sample, with standard deviations up to or exceeding a factor of 10. For the r<16μm, the concentrations are closer to those of Mason and have a smaller standard deviation from soil to soil, more like a factor of 3. Concentrations in the r<16μm range are generally greater than those for the bulk soils. Elements which deviate most strongly from Mason's data are Si (2-2.5 x low), Ca, K (a bit low), Na (5x lower), Ti (2x higher; artifact of including radius 10-16μm in the calculations), Mn (70% of Mason's), Sr (4x low), and Zr (?). Elements systematically higher than Mason include Ag (30x), Au (15x) and Sb (4x). It should be noted that:

A. These deviations will vary from aerosol to aerosol depending on the proportion of large particles in the aerosol. For fresh desert aerosol the proper reference material should go up to radius 20μm, for aged continental or desert aerosol the reference material need only extend to radius 5μm.

B. The data generated in this project represent strongly weathered soils rather than weakly weathered soils.

C. In general this work confirms the use of Mason's rock as crustal reference material. It supports the basic underpinings of the compilation. In other words, we were very lucky in the past.

6. <u>Miscellaneous remarks.</u>

    A.   The wet-dry test. There appeared to be minor dissolution of elements, on the order of 20-30%. Curiously, this effect seemed to be independent of the element. The reason for this is not yet known. This dissolution does not affect any of the basic conclusions from the work.

    B.   Silicon/Al ratio. This ratio decreases to values of approximately 1-2 for radii less than 10μm. This ratio is in basic agreement with that found for remote aerosols.

    C.   The enrichment factors of some elements tend to increase with the amount of mass in the small size range of the soil (radius less than 16μm), so that the greater weathering is higher enrichment factors. See plot below.
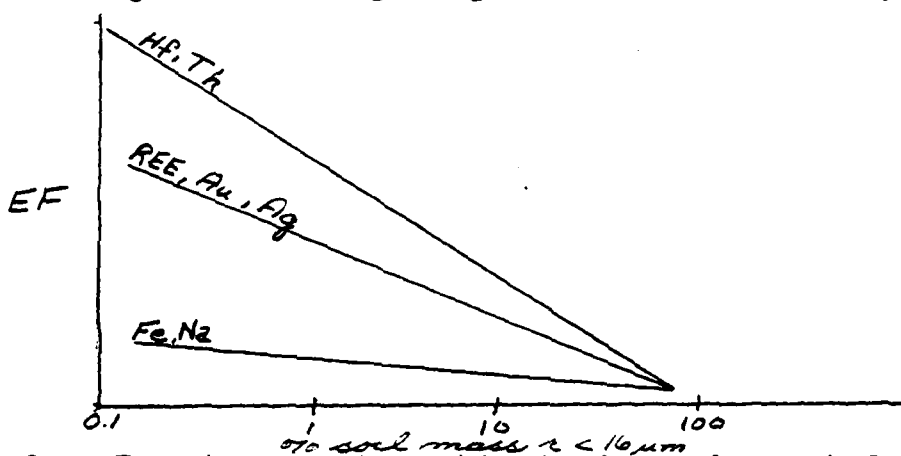
    D.   The moderate to high enrichments of most elements in Sahara aerosol now seem to be explained by Sahara soil. Indications are that aerosol-soil fractionation is primarily one of selection of certain radii when aerosol is generated from the soil.

    E.   Comparison of fallout with parent soil (S-39 vs. S-2). The major results from this study was that there seemed to be no fractionation within a given size range. High concentrations of antimony in the fallout seem to be contamination from the paint of the ping-pong table rather than positive aerosol-soil fractionation.

10.   Particle-size distribution of the Arctic aerosol (UA)

The particle-size distribution of an aerosol is one of its most basic physical properties, yet this has never been directly measured for the Arctic aerosol. An indirect estimate of the size distribution can come from the inversion of various optical properties of the aerosol that G. Shaw measures (Shaw, 1979; Section I.E.3 here). Preliminary results for 4-5 test periods in Fairbanks suggest that the size distribution there is approximately log-normal, with the maximum particle concentration at radius roughly 0.1 µm. The total columnar aerosol mass, derived from the columnar size distribution, is 10^{-6} to 10^{-7} g cm^{-2}.

11.   Climatology of Arctic haze (UA)

Study of the climatology of Arctic haze, the Ph.D. thesis of Mr. W.E. Raatz at the University of Alaska, has already yielded results in two of its three areas (see Section I.B.13 above for an outline of his thesis). A historical study of cloudiness in the Arctic since 1920 has shown that cloudiness there has remained generally unchanged since 1920, except for a slight increase in Alaska in the early 1940's. From this he has concluded that pollution aerosol in the Arctic, which has presumably increased greatly since 1920, does not affect the overall cloudiness. (It may well affect the microstructure and radiative properties of the clouds, however; the Ph.D thesis of R.D. Borys may deal with this topic.)

Mr. Raatz has begun the second phase of his thesis, a climatological study of the weather observations from the Ptarmigan reconnaissance flights of 1949 through 1967. Record sheets from these flights were obtained on four reels of microfilm during the last contract period from the National Climate Center in Asheville, NC. Mr. Raatz has examined the records for reports of haze, with some surprising and discouraging results. From 1949 through 1961 there were 3274 Ptarmigan flights; in the late 1950's they operated almost daily (see Table 2). There were three different forms used to record the observations, in five periods between January 1949 and December 1967. After July 1963 the forms used did not have a place for visibility, so visibility records are only available from January 1949 through June 1963. Haze was reported from 1949 through the mid-1950's, in a total of 114 flights (see Table 3). After this, haze was not reported any more, even though the corps of observers remained about the same. The reason for this is not known. Thus, there are basically only seven years of haze observations available from the Ptarmigan flights. These years, though, are enough to establish a clear maximum in March-May (Table 3) with a nearly constant "background" during the other nine months. This spring maximum agrees well with the March-April maximum of aerosol at the surface in Barrow (as seen for SO$_4^=$ and V, for example), and with an apparent April maximum of turbidity at Barrow. The question of possible winter haze is left unanswered by the Ptarmigan observations, however. We believe that there must be frequent winter haze in the Arctic, which cannot be observed visually. The Ptarmigan data do not support this idea unless a correction for poor winter lighting is applied.

No results are yet available from the third phase of Mr. Raatz's thesis, the statistical evaluation of chemical data from the Arctic Air-Sampling Network. The 5-day means are calculated; first statistical results should be ready soon. UA has recently begun to receive hemispheric meteorological maps from the German Weather Service, to aid in correlating patterns of Arctic haze with synoptic configurations in the Arctic.

Table 2. Number of Ptarmigan Weather Reconnaissance Flights
per month for the period 1949-1961.

	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Total
1949	11	13	15	14	14	15	14	16	13	10	9	0	144
1950	2	9	3	4	8	11	9	13	15	12	10	10	106
51	2	4	15	24	27	17	17	15	13	16	15	12	177
52	5	5	9	2	3	13	16	18	22	14	27	28	162
53	23	23	27	28	31	27	32	30	24	13	21	22	301
54	14	18	31	32	32	30	32	31	29	29	26	27	331
1955	32	27	33	31	31	30	31	32	31	31	31	31	371
56	31	29	33	30	32	31	31	31	31	31	32	33	375
57	25	8	0	0	19	15	20	22	14	27	27	30	207
58	30	27	11	25	31	31	31	2	30	39	29	31	317
59	31	27	31	30	31	29	33	31	30	31	31	31	366
1960	31	20	19	16	0	2	5	16	12	11	13	31	176
61	3	12	31	20	25	30	26	10	37	19	13	15	241
Total	240	222	258	256	284	281	297	267	301	283	284	301	3274

Table 3. Number of Ptarmigan Weather Reconnaissance Flights per month when haze was reported in the form as an additional comment on the data sheet.

	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Total
1949	3	4	2	4	0	1	0	2	1	0	2	–	19
1950	1	0	0	2	5	1	0	2	3	4	2	2	22
51	1	2	4	8	7	2	1	0	1	0	1	0	27
52	0	0	1	0	0	0	0	0	0	0	0	0	1
53	0	0	1	2	7	1	3	3	1	0	0	3	21
54	0	0	5	2	0	1	0	0	0	0	0	1	9
1955	1	0	3	1	3	0	0	0	0	0	0	0	8
56	0	0	0	1	1	0	2	0	0	0	0	0	4
57	0	0	–	–	0	0	0	0	0	0	0	0	0
58	0	0	0	0	0	0	0	0	0	0	0	0	0
59	0	0	2	0	0	0	0	0	0	0	0	0	2
1960	0	0	0	0	–	0	0	0	0	0	0	0	0
61	0	0	0	0	1	0	0	0	0	0	0	0	1
Total	6	6	18	20	24	6	6	7	6	4	5	6	114

D.   Publications resulting at least in part from this project,
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WMO Symposium on the Long-Range Transport of Pollutants and its Relation
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In preparation

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devoted to the Second Symposium on Arctic Air Chemistry, held at the
University of Rhode Island, 6-8 May 1980):

Rahn K.A., Progress in Arctic Air Chemistry 1977-80.

Patterson E.M. and Rahn K.A., Visible absorption coefficient of the Barrow
Alaska aerosol: Seasonal variations and compositional relations.

Shaw G.E., Eddy diffusion transport of Arctic haze: A preliminary model.

McCaffrey R.J. and Rahn K.A., The role of ^{210}Pb and ^{222}Rn in understanding
sources and transport of the Arctic aerosol.

Borys R.D. and Rahn K.A., Measurements of cloud-active aerosol in the Arctic.

Rahn K.A. and Shaw G.E., Potential direct radiative effects of the Arctic
aerosol.

Rahn K.A., On the atmospheric deposition of trace elements to the Arctic
Ocean.

Rahn K.A., The Mn/V ratio as a tracer of large-scale sources of pollution
aerosol for the Arctic.

Rahn K.A., The Arctic Air-Sampling Network in 1980.

Rahn K.A. Evidence for North America vs. Eurasia as a source of Arctic
aerosol.

Rahn K.A., Conway T.J. and McCaffrey R.J., Annual cycles of constituents
of the Barrow aerosol.

(To be presented at the General Motors Research Symposium <u>Particulate</u> <u>Carbon: Atmospheric Life Cycle</u>, 12-14 October 1980, Warren, Michigan):

Rahn K.A., Brosset C. and Ottar B., Long-range transport of elemental carbon from Eurasia to the Arctic.

E. REPRINTS OF SELECTED PUBLICATIONS

I.E.1.

Compositional differences between Arctic aerosol and snow

UNIQUE information on trace elements in polar atmospheres is available through records of deposition on snow and ice. Proper interpretation of these data requires a knowledge of the mutual chemical relationship between the deposition and its parent aerosol. Because several complex processes determine the trace-element content of precipitation, polar ice and snow cannot be considered *a priori* to have the same composition as polar aerosol. In most of the current literature, such an assumption is usually made. Within the past year or so, the first tests of the long-term relationship between aerosol and deposition have been (un)effectively produced for the Antarctic and the Arctic. The results are different for each region, and illustrate the great caution that must be exercised in this entire field. At the South Pole, snow from 1973–74 compares very well with aerosol from 1974–75 — both media give the same impression of the environment. In the Arctic, on the other hand, opposite conclusions about the atmosphere have been drawn from snow and aerosol, and this problem is discussed here.

Weiss *et al.* have used crustal enrichment factors of trace elements in recent Barrow, Alaska, snow and pre-1900 Greenland ice to conclude that (1) the lithospheric elements Al, V, and Mn are not enriched at Barrow and Mikcent (Greenland), and hence are derived from a (naturally) windborne lithospheric component; (2) the enrichments of Na, Zn and Cd are similar at Barrow, Mikcent, and other Greenland sites [which implies that these elements are natural or unique at Barrow]; (3) other enriched elements such as S, Se, Hg, Cd, As and Sb are naturally enriched in pre-1900 Greenland deposits. The overall impression given by Weiss *et al.* is of the natural character of polar deposition in both Greenland before 1900 and Barrow today. Weiss *et al.* refer interchangeably to 'atmosphere' and 'aerosol', 'snow', 'ice', and 'deposit', implying that this natural character also holds for the polar atmosphere.

In contrast, we have recently concluded from chemical analysis of two years' samples of the Barrow aerosol that it is strongly pollution-derived, particularly during winter, to the extent that abundances of even lithospheric elements can be affected. This deduction is based on actual V/Al and Mn/Al ratios, which are more characteristic of mid-latitude pollution than of crust. It is supported by ancillary evidence such as abundant grey, sooty

carbon and high concentrations of sulphate (1–5 μg m⁻³ in the aerosol (both of which imply pollution), and high concentrations of ²¹⁰Pb in the aerosol (which indicate that it is continental in origin). Tests at the surface (spring 1979) and North Slope, it is associated with air coming to Alaska from North Slope. In analogy to the similarity of snows from various parts of the Arctic reported by Weiss *et al.*, we find a similar (pollution) aerosol at the widely dispersed Arctic sites of Spitsbergen, northern Norway, and northern Greenland during water.

We therefore have two lines of evidence (aerosol and snow) which have arisen independently, are both solid and defensible, but offer divergent pictures of the Arctic environment. How can they be reconciled? Part of the difference for Barrow can be explained by recalculating snow-crust enrichment factors with Al rather than Mn as crustal reference element. Weiss *et al.* used Mn because it was common to all their snow samples. In general, however, Mn is enriched by factors of 3–10 in urban and rural atmospheres, presumably by pollution sources, and therefore should not be used as a crustal reference element wherever it can be avoided (Of course, without knowing how much the Barrow aerosol is affected by pollution, it would seem reasonable to use Mn.) Concentrations and revised enrichment factors for the Barrow winter aerosol and the near-Barrow snows are shown in Table 1. These calculations, show that which can be justified from many aerosol samples, show that bulk V and Mn have modest but definite enrichments in the near-Barrow snows. But this recalculation removes only part of the discrepancy between Barrow aerosol and snow — V and Mn are still several times more enriched in the aerosol than in the snow, and all other elements except Ca have greater enrichments in aerosol than in snow. For Na, Mg and Ca, this difference may be because related, because the aerosol sampling site was closer to the sea than were the snow-sampling sites. The other elements, however, are not marine in origin, their different enrichments must have another explanation.

We believe that the broad discrepancy between Arctic aerosol and Arctic snow is a genuine feature of the present Arctic environment. Possible explanations for it include: (1) formation of snow from a parent aerosol aloft whose composition differs from the surface aerosol, and (2) aerosol–snow fractionation during or after snowfall. We favour the second explanation for two reasons: first, we see little reason that the composition of the aerosol in such a remote, source-free area should change markedly with altitude. Second, formation of snow crystals by nucleation on clay minerals, such as are found at the centre of many or most polar snow crystals, could fractionate snow chemically relative to aerosol. A central clay nucleus which contained 90% of the (inorganic matter) if the ice crystal (estimated by M. Kumai, personal communication) would decrease enrichments in snow by the factor of 10 that is observed. We stress, however, that aerosol–snow fractionation in the Arctic is only a hypothesis.

We emphasise that whatever the eventual explanation, aerosol–snow differences in the Arctic imply, care is needed in interpreting ice and snow data atmospherically. Modern snows may not be automatically equated with modern aerosols, historical trends of deposition may not be immediately equated with historical trends of parent aerosol. Until thorough studies of the factors controlling the elemental composition of polar snow and ice are undertaken, these uncertainties will remain.

Table 1 Elemental concentrations and crustal enrichment factors in Barrow aerosol and snow

Element	Aerosol Concentration (ng m⁻³)	EF_crust	Snow Concentration (μg l⁻¹)	EF_crust
Na	770	74	144	159
Mg	160	21	41	61
Al	30	1.0	26	1.0
Ca	350	4.1	100	86
V	0.65	130	0.086	7.0
Mn	1.12	32	0.79	26
Zn	1.45	580	0.16	52
Cd	0.375	5,000	0.047	730
Hg	<0.04	<14,000	0.0115	430

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I.E.2.

LONG-RANGE TRANSPORT OF POLLUTION AEROSOL
TO THE ARCTIC: A PROBLEM WITHOUT BORDERS

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Overview of the Arctic aerosol

Recent research has revealed the presence of unexpectedly large amounts of aerosol in the Arctic atmosphere during winter[1]. At Barrow, Alaska, for example, the aerosol undergoes a rapid change in amount and composition during October and November. Before this it is low in concentration, colorless, and composed primarily of natural sea-salt and soil particles. After this it is much more abundant, is gray on filters, and shows a marked pollution component. In contrast to the giant-particle sea-salt and soil components, which change little from summer to winter, constituents such as V, Mn, sulfate, ^{210}Pb (the first three of which we feel are pollution-derived), increase by factors of 10 to 30 from summer to winter. The period of high concentration is November through April, after which concentrations rapidly decrease to summer values. Recent tests have established that this winter submicron aerosol (called simply "aerosol" in the rest of this paper) is not significantly influenced by Barrow or the rest of the North Slope of Alaska, including Prudhoe Bay. Our samples are truly regional in nature, and represent the aerosol of at least the central Polar Ice Cap. Similar concentrations, compositions, and seasonal variations have been found for the aerosols of Spitsbergen, Bear Island, Thule, and Northern Norway by a series of cooperative programs with the Norwegian Institute for Air Research and the Danish Air Pollution Laboratory.

A significant feature of the Arctic aerosol is its secondary nature. The three secondary constituents of the aerosol which we have measured to date (secondary aerosol is formed in the atmosphere from a gas-phase precursor, primary aerosol is injected directly into the atmosphere as an aerosol), sulfate, organics, and ^{210}Pb, are all enriched relative to primary constituents by an order of magnitude compared to midlatitude aerosol (organic data from J. Daisey, New York University, personal communication). This suggests that the Arctic aerosol is very aged, and cannot be explained by local sources. In more practical terms, the bulk of the mass of Arctic aerosol is secondary.

A major motivation for our study of the Arctic aerosol was Arctic haze, abnormally high turbidities in the Arctic atmosphere during winter and spring[2]. This haze was first reported by the Ptarmigan weather reconnaissance missions in the Alaskan Arctic, and was observed to extend poleward of 80°N[3]. Since then we have received reports from widely scattered locations such as Mould Bay (Canada), Thule (Greenland), and Ny Alesund (Spitsbergen). The haze thus seems to be a general feature of the Arctic atmosphere, and seems to be several times more intense in spring (probably during winter as well) as in summer. Although the haze extends at times to the tropopause, near Barrow its greatest intensity is normally

between about 1 and 2 km altitude, near the top of the surface inversion layer. At this level the relative humidity is near 100%; we postulate that the haze is caused as hygroscopic aerosol particles (mostly sulfates) take up large amounts of water and grow into optically-active submicron size range. Here the enrichment of secondary material in the Arctic aerosol is critical - concentrations of sulfate and organics at Barrow during winter are only about 3 times less than in midlatitude polluted areas such as Europe or the northeast United States, and the concentration of ^{210}Pb is actually double that of the northern United States during the winter. With such high concentrations of sulfate (which seems to be the main agent of light scattering in various regions of the world), and a persistent near-saturated layer over the entire ice cap, it is no wonder that high turbidities are found during spring. With the onset of summer and the decrease of the submicron aerosol turbidities simultaneously decrease.

The origin of this winter Arctic aerosol is of prime importance to any evaluation of its environmental significance. It is highly continental in character, containing some of the highest ^{210}Pb concentrations ever measured. Of the trace elements, at least V and Mn have sufficiently high aerosol-crust enrichments that they must be considered of pollution origin. Many other trace elements, such as As, Sb, Cd, Fe, Cr, In, etc., also seem to be pollution-derived at Barrow during winter, but have been less completely measured than V and Mn and so will not be considered further here.

V is of particular interest, because its major pollution source is combustion of heavy residual oil[3]. Because this oil is very viscous, it is burned almost exclusively in midlatitudes. We are not aware of the use of residual oil in any Arctic community. The extremely high concentration of V at Barrow must, therefore, have come from midlatitudes. Because of the extremely good correlation between sulfate and V at Barrow (see Figures 1 and 2 below), we feel that the sulfate is probably also basically pollution-derived. Because sulfate is probably the most abundant constituent of the Barrow aerosol, we feel that much of the Barrow winter aerosol is pollution derived. We stress, however, that sulfate can have natural sources such as the stratosphere, volcanoes, and the biosphere, so that the above conclusion is only tentative.

Detailed interpretation of the Barrow aerosol data

During the last two years a comprehensive explanation for the Barrow aerosol data has evolved, and will be presented here. Although it makes good physical and meteorological sense, it involves several concepts not yet widely accepted and is largely unquantified. Because such quantitative proof (via trajectories, complete transport models, etc.) is still years away, we feel that a discussion of our semi-quantitative hypothesis, together with its uncertainties, is appropriate.

The important chemical data for Barrow (and for much of the rest of the Arctic, we feel) are summarized in Figures 1 and 2. Figure 1 shows monthly mean concentrations of ^{212}Rn, ^{210}Pb, sulfate, and V at or near Barrow, together with monthly means of several meteorological parameters for the Arctic. Figure 2 shows actual data for V, sulfate, ^{210}Pb in individual samples during 1977-78,
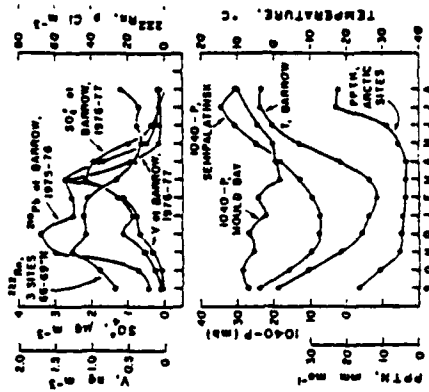

Figure 1. Seasonal variations of various chemical and meteorological parameters the Arctic.

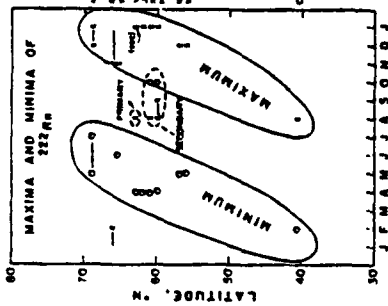The seasonal pattern of ^{210}Pb is similar to that of its precursor ^{222}Rn, but there are systematic differences. The $^{210}Pb/^{222}Rn$ ratio increases sharply in October, remains high November through April, then decreases during May to low values which remain through September and part of October. The high ratios coincide well with the November-April minimum in precipitation in the central Arctic, also shown in Figure 1. These precipitation values are the means of 11 Arctic sites on the edge of the ice sheet, forming a rough ring from Alert to Barrow to Novaya Zemlya[9]. The relation between precipitation and $^{210}Pb/^{222}Rn$ ratio suggests that the low Arctic precipitation in winter gives rise to very long residence times there, which can enhance long-range transport of all aerosol, something we have suspected for some time from other lines of evidence. The effects of these long residence times are especially great on secondary aerosol formed within the Arctic, which can then build up to unusually high concentrations, being limited mainly by the transit time of an air parcel through the Arctic rather than removal processes. Independent calculations show that at Barrow has been formed from ^{222}Rn within the Arctic. If this is indeed so, then it should be possible to relate Arctic ^{210}Pb to its parent ^{222}Rn in a simple way, because the seasonal patterns of northern ^{222}Rn are known and the residence times of aerosol over the Arctic are uniformly long during winter. We have attempted such a quantitative treatment of ^{210}Pb at Barrow; the results are shown in Figure 4. The upper curve (a) is the mean northern seasonal pattern of the parent ^{222}Rn. Curve (b) is an arbitrary residence-time function for the Arctic, the reciprocal of the mean monthly precipitation over the 11 sites mentioned above. Curve (c) is merely the product of curves (a) and (b). It should approximate ^{210}Pb at Barrow if seasonal changes in speed do not seriously affect the balance between ^{222}Rn and ^{210}Pb, and if the removal of ^{210}Pb formed within the Arctic is small compared to its generation. These two conditions are apparently met because curve (c) bears a remarkable resemblance to the actual pattern of ^{210}Pb at Barrow, shown in curve (d).

There are several further remarks that can be made about this plot. First, ^{210}Pb in the Arctic seems to be explained by northern ^{222}Rn. But little of the ^{222}Rn observed near 70° North may have originated there, because of the nearly universal presence of permafrost at these latitudes. Although there is little hard evidence concerning emanation of ^{222}Rn from permafrost, there is a general feeling that it must be attenuated relative to most unfrozen soils. For purposes of calculation, Wilkening[9] assumed rates of emission of ^{222}Rn from permafrost to be one-tenth of those from midlatitude soils. If this figure is accurate, the ^{222}Rn observed over...

Figure 4. Calculated and actual seasonal variations of ^{210}Pb at Barrow, Alaska.
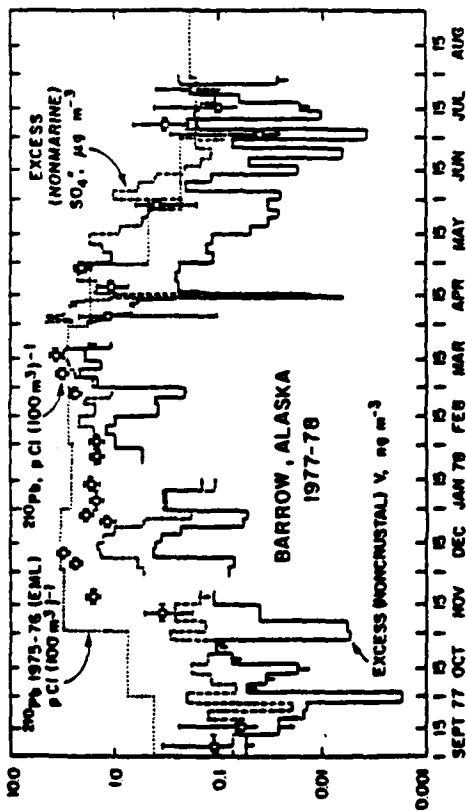
Figure 2. Concentrations of noncrustal V, nonmarine sulfate, and ^{210}Pb in individual filter samples at Barrow, Alaska during 1977-78. Monthly mean concentrations of ^{210}Pb at Barrow during 1975-76 are given for comparison.

gether with monthly means of ^{210}Pb from 1975-76 for comparison[5]. The seasonal pattern of ^{210}Pb of Figure 1 is a mean of 3 northern sites, none of which is Barrow, and is discussed etem.

From Figure 1 it can be seen that there are two distinct seasonal patterns present in the arrow aerosol record, one for ^{222}Rn and ^{210}Pb, another for sulfate and V. ^{222}Rn and ^{210}Pb ave a peak in November-December followed by a broad plateau for the rest of the winter, and cline rapidly during April and May. Sulfate and V have nearly the opposite pattern, a road winter plateau beginning in November with a strong peak in March and April, followed y the same rapid decline in April and May. Winter maxima for all 3 aerosol species are 10-0 times their summer values; for ^{222}Rn the figure is roughly a factor of 4. The natural ontinental ^{222}Rn and ^{210}Pb thus peak in early winter at Barrow; the pollution-derived sulate and V peak in late winter. Alternatively stated, Barrow is influenced by relatively mpolluted continental air in early winter, followed by progressively more polluted, less ontinental air as winter advances. At the end of winter there is a pulse of much more polluted, less continental air which lasts for about two months just before the summer clean-air situation begins.

Both the ^{222}Rn and ^{210}Pb at Barrow seem to be derived from the northern parts of Asia nd North America. For ^{222}Rn this is a consequence of the progressive delay in the times f maximum and minimum concentrations with increasing North latitude (Figure 3), which suggests that ^{222}Rn concentrations are related to local emission conditions, although transport effects cannot yet be ruled out. From the regular seasonal patterns seen in Figure 3, nd the fact that ^{222}Rn has a November maximum in both Alaska and extreme northern Finland[6,7] the mean northern seasonal pattern shown in Figure 1 was derived by averaging data from Nales Alaska[8] with Kevo and Rovaniemi, Finland[7]. These three sites are between 66 and 69° North. No ^{222}Rn data are available for Barrow yet.

continents at 70° North must have come from farther to the south, at least as far as the sub-Arctic and possibly in part from midlatitudes as well. Such transport of ^{222}Rn, seldom considered in the literature, may be important in the North.

A second implication of Figure 4 is that Arctic residence times may be 7 times more in winter than in summer. If residence times are inversely proportional to rate of precipitation, as assumed here, the relative lack of precipitation (roughly 10 times less) in the Arctic indicates that aerosol residence time there may average an order of magnitude more than in midlatitudes (about 5 days). Winter Arctic residence times, however, are estimated to be 50-100 days or more. Such seasonal variations of residence times and the large absolute values presumed here for the lower troposphere are not commonly recognized.

The calculations in Figure 4 assume summer and winter transport times within the Arctic to be equal, as mentioned above. This is almost certainly not the case, because the Arctic circulation is more sluggish in summer. Our calculations suggest that ^{210}Pb should be sensitive to the intensity of the Arctic circulation. If so, then the transit time between continents and Barrow must be nearly the same in summer and winter, for reasons yet unknown. The big seasonal difference in Arctic ^{210}Pb concentrations thus seems to be principally caused by variations in source and precipitation functions rather than by variations in transport.

Lastly, there is some evidence of a time delay of about a month between parent ^{222}Rn and daughter ^{210}Pb, both in the actual measurements of Figure 1 and in the simulated ^{210}Pb of Figure 4. In fact, there must be some delay caused both by transport (which we estimate at roughly 10 days within the Arctic) and any "reservoir effect" of a sluggish pool of Arctic air responding only slowly to inputs of air from the south. This time-delay aspect of the Barrow aerosol is not well understood, however.

Both ^{222}Rn and ^{210}Pb at Barrow probably originate from both Asia and North America. Figure 1 shows that the buildup of these constituents at Barrow (and also V and sulfate) during October and November coincides with the buildup of the pressure in the Svalbard region decreases, so that the pressure gradient between the Norwegian Arctic and eastern Siberia increases. As a result, northward flow of air across the Arctic border of the USSR increases at this time. This basic pressure pattern and resultant air flow continue throughout much of the winter. The mean northward flow of Siberian air into the Arctic can be seen in a number of ways, for example from the January streamlines of surface air flow or from the October and January surface pressure maps. The January pressure map is reproduced here as Figure 5, from which it can be seen that the mean flow enters the Arctic between Novaya Zemlya and the New Siberian Islands. Although this path surely varies from day to day and from season to season, the October map suggests that on the mean the entry to the Arctic is systematically farther to the east, between the Taymyr Peninsula and the New Siberian Islands. Evidence will be presented below that in March and April the path is temporarily much more to the west, perhaps between northern Scandinavia and Novaya Zemlya. The flow of air after it enters the Arctic has been described as follows: "Over the central Polar Ocean, the main air stream is directed from the middle and western Siberian coast toward the Pole and thence southward across the Greenland-Spitsbergen area. On the Bering Strait side of the Pole the Siberian air masses cross the Polar Ocean and invade the
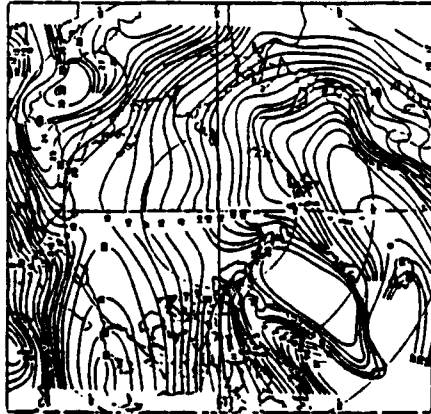
Figure 5. Mean surface pressure (mb) in January (ref. 8).

Canadian Archipelago, mixing with Alaskan-Yukon air."

Inspection of the daily pressure maps of early winter 1977-78 confirms this mixed North American-northern Asian origin of ^{210}Pb-rich air reaching Barrow. Air of 3-12 December was Siberian in origin; air of 23-27 December and 31 December-10 January had a greater North American influence. During January it is common to have flow of Pacific air from the south into Barrow; the Alaskan continental influence should also be felt in such cases. It cannot be excluded, however, that air most recently from northern North America or the Gulf of Alaska may have previously come from Siberia, hence some of the high ^{210}Pb may be attributable to this more distant source rather than the more obvious nearer ones. In this regard the November maxima of ^{222}Rn at Kodiak and Wales, Alaska, which are marine or semi-marine sites, may well be Asian in origin.

The other main seasonal pattern of aerosol at Barrow to be interpreted is the spring maximum of the pollutants V and sulfate, which we take to indicate maximum in pollutants in general. V and sulfate have basically the same seasonal pattern at Barrow, and will be considered together for the rest of this discussion. Their behavior is puzzling. On the one hand, they respond quickly to the change of the general circulation and the decrease of precipitation in October and November, by increasing in concentration to roughly 10 times their summer values. This shows that the Arctic atmosphere becomes chemically linked to midlatitudes at this time. On the other hand, their absolute maximum comes about 2 months later than that of ^{210}Pb, in March and April. (The monthly mean concentrations are higher in March than April, but the frequency of very high concentrations is about the same in two months, as shown in Figure 2). Without the spring maximum the seasonal patterns of V and sulfate would be very similar to that of ^{210}Pb.

Like ^{222}Rn and ^{210}Pb, small but significant differences can be seen between the seasonal patterns of V and sulfate. The sulfate/V ratio is roughly inversely proportional to the concentrations of each, with a maximum in the summer about 3 to 4 times greater than the spring minimum. Because this seasonal variation seems to be greater than that of SO_2/V in urban source regions, which is roughly a factor of 1.5, it appears to reflect the effect of differing path lengths to the Arctic from midlatitude polluted areas, with higher sulfate/V ratios associated with longer path lengths or transit times, which provide more time for SO_2 to be converted to sulfate. According to this idea, path lengths or transit times to the Arctic are longer in summer than in winter. This makes sense because the summer circulation is more sluggish than the winter circulation, and because the path to the Arctic in winter is more direct due to the greater meridional circulation over Asia.

As noted above, there is significant evidence accumulating that both sulfate and vanadium are mostly pollution-derived in the Arctic during winter. Aerosol-crust enrichment factors of V are 10-15 at Barrow during winter, which means that more than 90% of it is pollution-derived. An analogous calculation for sulfate is not possible because of the variety of natural sources that can affect its abundance, but its similarity to V, its high concentrations in winter, and its compatibility with known sources, residence times, and transport paths all strongly suggest that much of it is pollution-derived as well.

The major pollution sources in the Northern Hemisphere which could be potential sources of Arctic aerosol are Europe/European USSR, the northeast United States, and Japan/Korea/China. The possible atmospheric pathways from these sources to the Arctic are shown in Figure 6. For comparison, mean winter concentrations of V and sulfate in the Arctic and selected environs are shown in Figures 7 and 8. Little aerosol seems to reach the Arctic from Japan/Korea/China via the Pacific pathway,

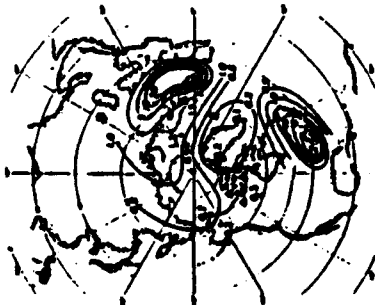Figure 6. Possible pathways of aerosol from midlatitudes to the Arctic.

The details of the path from Eurasia to Barrow are not yet clear. In fact, there must be a variety of paths. As mentioned above, the mean flow during early winter seems to pass over more of the Asian continent than does the January flow. There is now further evidence that the path is still shorter in spring: (1) In spring the sulfate/V ratio is the lowest of the year at Barrow, which we interpret to mean that the path length is the shortest; (2) In late March and early April there is a temporary minimum in ^{210}Pb at Barrow, which comes simultaneously with the peak concentrations of V and sulfate. This is consistent with a less Asian, more northward path from Europe to the Arctic; (3) In spring a closed high is established over the central Polar Ocean, which is reflected in the March-April-May pressure maximum at Mould Bay shown in Figure 1. This pressure system intensifies the circulation southwestward along the Canadian Arctic Archipelago, which is then easily connected to the air flow northward from Europe; (4) During spring the Asiatic high migrates westward as it breaks up, and during April becomes temporarily established with its center in northeastern Kazakhstan[11]. This provides a logical physical mechanism for shunting air flow more northward from Europe. Individual pressure maps from Spring 1977 and 1978 have shown that such a high was in fact present for 1-2 weeks just before the highest aerosol concentrations of the season were observed at Barrow; (5) A spring maximum of aerosol is indeed observed along the proposed short path north of Europe, at northern Norway, Bear Island, and Spitsbergen. During the majority of cases which we have accumulated to date; (6) Lastly, we have an argument by default: A short path during spring is needed to explain the spring maximum at Barrow because it can't be explained in the same way as was done in Figure 4 for ^{210}Pb.
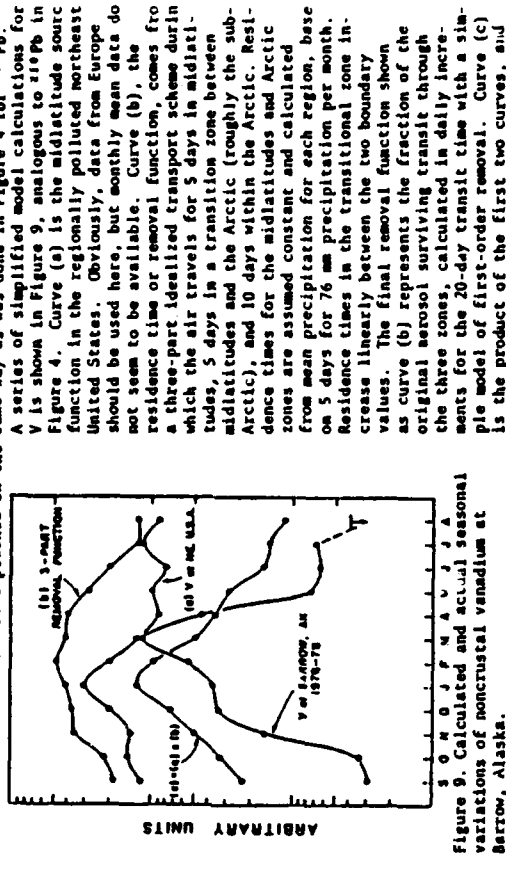
A series of simplified model calculations for V is shown in Figure 9, analogous to ^{210}Pb in Figure 4. Curve (a) is the midlatitude source function in the regionally polluted northeast United States. Obviously, data from Europe should be used here, but monthly mean data do not seem to be available. Curve (b), the residence time or removal function, comes from a three-part idealized transport scheme during which the air travels for 5 days in midlatitudes, 5 days in a transition zone between midlatitudes and the Arctic (roughly the sub-Arctic), and 10 days within the Arctic. Residence times for the midlatitudes and Arctic zones are assumed constant and calculated from mean precipitation for each region, base on 5 days for 76 mm precipitation per month. Residence times in the transitional zone increase linearly between the two boundary values. The final removal function shown as curve (b) represents the fraction of the original aerosol surviving transit through the three zones, calculated in daily increments for the 20-day transit time with a simple model of first-order removal. Curve (c) is the product of the first two curves, and represents the expected seasonal variation of V at Barrow when only the source and removal functions are allowed to change. Unlike ^{210}Pb, however, the maximum is predicted two months early and the overall amplitude of the curve is 3 to 4 times too small. One gets the impression that curve (c) reasonably accounts for the basic features of the actual V pattern, shown in curve (d), except for the missing spring maximum, of which there is no hint. Were a maximum during March and April to be added to the plot, the simulation would be acceptable. Our deduction from this is that the missing spring maximum is a result of the factors not considered in these calculations, most probably a change in the transport path.

Figure 9. Calculated and actual seasonal variations of noncrustal vanadium at Barrow, Alaska.

Figure 7. Winter concentrations of noncrustal vanadium, ng m^{-3}.

Figure 8. Winter concentrations of noncrustal sulfate, µg m^{-3}.

as evidenced by both consistently lower concentrations of V and sulfate at Fairbanks than at Barrow during winter, and by the fact that strong incursions of warm southern air into Barrow inevitably bring concentrations of aerosol greatly reduced from those found when the colder air comes from the north. A good, though perhaps extreme, example of this is shown for mid-April 1978 in Figure 2, when a strong current of southern air to Barrow decreased concentrations of V and sulfate by more than an order of magnitude for about two days. The pathway from the northeast United States via Iceland and Spitsbergen seems also to be an ineffective one, as evidenced by lower concentrations of V, sulfate, and ^{210}Pb along this path also showed that southern Greenland than in the Arctic. Earlier simulations of V, sulfate, and ^{210}Pb found in the Arctic. It probably could not produce the high concentrations of sulfate and ^{210}Pb found in the Arctic. The general ineffectiveness of this path seems physically reasonable, because a great deal of the pollutants should be removed over the stormy North Atlantic. In this sense, the Atlantic and Pacific paths are similar. In individual cases, more the exception than the rule, aerosol may be transported along this pathway to the Arctic.

The short version of this path, via the western side of Greenland and Baffin Bay, is also possible but unlikely to dominate the mean. As seen in Figures 7 and 8, concentrations of pollutants are less at Thule than at Barrow during winter. Further, there seems to be no major driving force connecting Baffin Bay to Barrow atmospherically. As with the North Atlantic, this pathway may well be effective occasionally, possibly during spring.

Figures 7 and 8 seem to indicate that Eurasia is the dominant source of aerosol for the Arctic in winter. A broad tongue of high concentration of aerosol, well-defined on the western side but ill-defined on the eastern side, extends from Europe through Scandinavia all the way to Barrow. The simplest possible interpretation of these figures is that this is in fact the dominant pathway for aerosol to the Arctic, northward or northeastward from Eurasia, then northward to the Arctic. Such a path is physically reasonable, because it is continental near the source and for quite some distance downwind, hence has less precipitation and cleansing than the Atlantic and Pacific pathways. It is also significantly shorter than the North Atlantic pathway via Iceland. To the extent that it passes northeastward over part of European Russia before being deflected northward, it can produce the high ^{210}Pb concentrations needed for Barrow in winter. The previous calculations showed that the V, sulfate, and ^{210}Pb observed at Barrow, and a path could account quantitatively for the V, sulfate, and ^{210}Pb observed at the high ^{210}Pb. In fact seemed to be required to account for the high ^{210}Pb.

Conclusions

In conclusion, then, much or all of the Arctic has high concentrations of aerosol in winter. No such phenomenon occurs in the Antarctic, to the best of our knowledge. At least some of this excess aerosol is pollution-derived, for example the excess V, Mn, and probably a number of other trace elements as well. Sulfate, the most abundant constituent so far measured in the submicron aerosol, reaches monthly mean concentrations of greater than 2 ug m^{-3}, with individual samples approaching 5 ug m^{-3}. These high concentrations, together with the similarity of seasonal variation of V, suggest but do not prove that much or most of the winter sulfate in the Arctic is pollution-derived. Such a conclusion seems physically tenable, because 90% of the world's pollution is released in the Northern Hemisphere, and most of that during winter in midlatitudes, which are meteorologically connected to the Arctic from November through April or May.

A number of pieces of evidence all suggest that the dominant source for this pollution aerosol is Eurasia, that is, Europe and the European USSR, although other polluted regions such as the northeastern United States and the Japan region may contribute from time to time. The pathways from Europe to the Arctic seem to vary with the season, entering the Arctic at progressively more western longitudes as winter progresses. Nearly all paths seem to pass over some portion of the USSR, though.

Although the environmental effects of this excess Arctic aerosol are not known with certainty, it is reasonable to assume that they exist. Arctic haze is a result of these high concentrations. Depositional effects are also of interest, particularly for acidity, nutrients and trace metals. Nucleational effects may also be significant, but have not yet been studied.

One thing seems clear, however: the Arctic aerosol may be of international importance. It shows evidence of being derived largely from (polluted) midlatitudes, with transport distances of approximately 10,000 km. Its precursors apparently originate in western and eastern Europe and the European USSR... pass over more of the central USSR, then over the international Arctic, and eventually reach northern Canada and Alaska. Thus, at least 10-20 countries are involved with the Arctic aerosol or its precursors at some stage of its development and transport.

A brief calculation can illustrate the magnitude of aerosol transport to the Arctic. We assume that during January Arctic air systematically enters the Arctic from northern Asia between longitudes 45°E and 135°E, at latitude 70°N, according to Figure 5. We further assume that the major transport is at low levels, between 0 and 2 km altitude (the OECD model for Europe uses 0 to 1.5km). From Figure 5 the mean geostrophic northward wind in this corridor and between 65° and 75° North can be estimated at 2.8 m s^{-1}. During January, therefore, .1 x 10^{15} g of air is transported into the Arctic via this corridor. The sulfur content of this air can be estimated at 1.42 ug m^{-3} (0.75 ug m^{-3} from SO$_2$ and 0.67 ug m^{-3} from sulfate) from measurements in northern Norway, Bear Island, and Spitsbergen. Therefore, the total S entering the Arctic via this corridor in January is 0.073 MT. Eurasian emissions of S for January can be estimated at 2.71 MT. Thus, 2.7% of the S emitted in Europe in January enters the Arctic. But only about 20% of the S emitted in January actually leaves Europe[12]. Therefore, 13.4% of the S leaving Europe in January enters the Arctic. These calculations are of course rough and preliminary, because we have no S data available from the main part of the transport pathway over the USSR. But they do serve to illustrate the importance of the Arctic as a receiver of midlatitude aerosol during winter.

Concerning the international aspects of studying the Arctic aerosol, we wish to point out that because the atmospheric residence times of aerosols in general are short compared to hemispheric mixing times, aerosols tend to be heterogeneously distributed in the atmosphere. When a region of high concentration is found, such as the Arctic, specific sources and well-defined transport paths can be expected, whose systematic study requires an area-wide or network approach. This is in great contrast to longer-lived atmospheric constituents such as CO$_2$, which become nearly homogeneously distributed and can be studied anywhere. Because the Arctic aerosol appears more and more to involve both eastern and western countries in its origin, transport, and environmental effects, it would seem to present an ideal

opportunity for a truly international cooperative study with potentially great global environmental significance. The Arctic Air-Sampling Network, a loosely bound alliance of several western Countries interested in Arctic air chemistry[13], now has nearly 15 sampling sites in operation but its regional coverage (Figure 10) is limited to only one-half the Arctic.
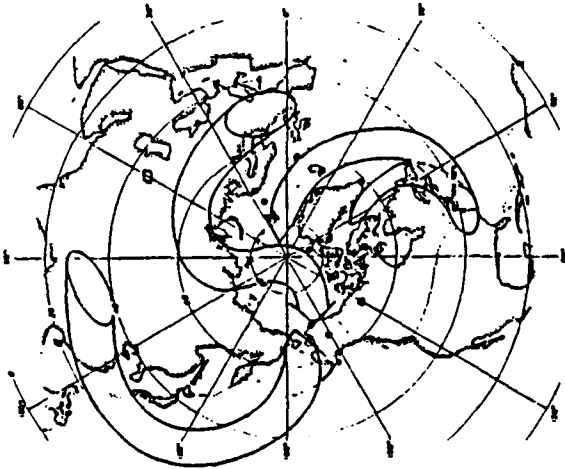

Figure 10. Sites of the Arctic Air-Sampling Network.

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Inversion of optical scattering and spectral extinction measurements to recover aerosol size spectra

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B 489

Fairbanks, Alaska 99

Aerosols interact with optical radiation in a complicated way, but in a way that can be described through the methods of electromagnetic theory. By applying constrained linear inversion methods one can recover an estimate for the size spectrum of aerosols in the micron and submicron size range from measured optical data. The amount of information that can be secured is increased by combining measurements of spectral extinction with measurements of optical scattering. From the combined measurements one can obtain about seven pieces of information about the particle size spectrum, provided optical parameters are measured to 1.5% accuracy. The application of linear constrained inversion on simulated noise degraded data successfully recovers major features, such as the presence of modes, in the aerosol size distribution function.

Introduction

When a sample of turbid air is intensely illuminated, the scattered light from the sample and the wavelength *dependence of optical spectral extinction through the sample provide information that, in principle, can be used to estimate the size and number concentration of the suspended particulates. The technique of recovering the particle size distribution function by combining measurements of scattered radiation and spectral extinction is the subject of this paper. It will be shown that by using experimental data taken to an accuracy of 1.5%, the major features of an aerosol size distribution function (in the particle size range $r > 10^{-5}$ cm) can be derived to an accuracy acceptable for many scientific purposes.

The problem of inversion being addressed differs fundamentally and philosophically from the usual analysis problem encountered in electromagnetic theory or optics, where one calculates scattered radiation or spectral extinction of light caused by an arbitrary but prespecified distribution of particles. A unique solution exists for the analysis problem. In the inverse problem, however, it is by no means obvious that a unique solution exists, especially when it is considered that the required solutions are functions of variables not just

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Mie's theory is used for solving problems of light scattering by aerosols, though it has to be stipulated that it applies in the strict sense only to spherical particles, which real aerosols often are not. But provided the aerosols themselves are nearly spherical (not disklike or rodlike, for example) and provided they do not have preferential orientations in space, Mie theory provides an acceptable approximation for the optical scattering and attenuation from a cloud of heterodispersed particles. In the case where there are many particles, the radiation fields or attenuation coefficients are gotten by summing the contribution from each particle. This works only provided the particles are distributed randomly in space.

The scattered light or the spectral extinction can be written as

$$g(x) = \int K(x,r)f(r)dr. \qquad (1)$$

where $f(x)$ represents the scattered radiation field at scattering angle $x = \theta$ and/or the optical extinction through a volume of particles evaluated at wavelength $x = \lambda$. In general g varies with x. $K(x,r)$ is an optical scattering or optical extinction cross section; it is evaluated with Mie theory and is dependent upon scattering angle $x = \theta$ (or wavelength $x = \lambda$) and particles radius r. The function $f'(r)$ is the particle size distribution function dN/dr or simply the number concentration of particles within size range r to $r + dr$ taken as a function of r; it is the term that represents the solution to the problem being treated. The kernel K depends also on the composition of the particle (more particularly on the particle's refractive index R); in this paper R is assumed known.

Taken as a strict mathematical exercise, Eq. (1) can, in principle, be solved by Fourier or Laplace transformation, but problems arise that can be best illustrated by transforming the equation into a set of first-order linear equations by expressing the integral as a summation:

$$x_i + c_i = \sum_{j=1}^{n} w_{ij}A_{ij}f_j; \qquad (2)$$

where g_i is understood to represent g at one of m specific scattering angles $x_i = \theta_i$ (in the case of scattering) or at a specific wavelength $x_i = \lambda_i$ (in the case of extinction), w_{ij} is a quadrature weighting coefficient, and f_j is the particle size spectrum evaluated at one of the n r_j values. In Eq. (2), g_i corresponds to the exact (error free) optical scattering or extinction coefficient, and c_i is an error term resulting from experimental uncertainty in practice, errors introduced into the equations by approximating the integral by quadrature expansion are usually small in comparison with random errors associated with experimental measurements.

Equation (2) may be expressed in matrix notation:

$$[g] + [c] = [A] \cdot [f] \qquad (3)$$
$$(m \times 1) \quad (m \times n)(n \times 1)$$

where, as a reminder, it is implicitly understood that

there are m measured values of optical parameters, and the problem is to evaluate the vector $f'(r) = dN/dr$ at n different particle sizes.

For the entirely hypothetical case of g being known exactly ($c_i = 0$), when quadrature errors can be neglected, and when $n = m$, the solution to Eq. (3) may be written simply as

$$[f] = [A]^{-1}[g]. \qquad (4)$$

In Eq. (4), A^{-1} is the inverse matrix of A. The solution exists, of course, only provided $DET(A) \neq 0$. If $m \neq n$, a least-square solution to Eq. (3) is straightforward to derive and can be written as

$$[f] = [A^tA]^{-1}A^tg. \qquad (5)$$

where A^t is the transpose of A (columns and rows interchanged), but again it has to be assumed that $DET(A^tA) \neq 0$.

When it is realized that an actual experimental determination of the vector g will always contain error ($c_i \neq 0$), a linear combination of the rows of A that does not vanish but is less than the error itself is hardly any better than a linear combination that vanishes and would lead to $DET(A) = 0$. This situation could come about if there was a large degree of interdependence among the rows of A, which in fact there is for the problem being treated. It is because of this fact that direct inversion or least-squares inversion by Eqs. (4) or (5) generally are not successful; usually one recovers wildly oscillating estimates for the vector f'.

Twomey[1] has discussed the reasons for the instabilities that arise when one attempts to use Eq. (4) or (5) for inversion. The problems that arise can be shown to be associated with the presence of small eigenvalues in the symmetric matrix A^tA; the small eigenvalues are the consequence of the near lack of linear independence of the equations. Of the possible manifold of functions f' allowed by $g + c$ through the integral transform, one must attempt, by some method, to select reasonable solutions. This can be done by applying constraints on the solution.[1]

The constrained solution to Eq. (1) is derived as follows: Consider first of all the quadratic form given by

$$Q = f^tH f = \sum_i \sum_j h_{ij} f_i f_j \qquad (6)$$

The value of Q obviously is dependent on the vector f and on the elements in the $n \times n$ matrix H. In Eq. (3) it can be assumed that an upper bound can be specified for the norm of the vector c, i.e.,

$$\sum_i c_i^2 \leq E^2 \qquad (7)$$

Now let the vector c vary within the limits prescribed by Eq. (7), and consider the minimum attained by the quadratic form in Eq. (6), m,c traces out its space. This is equivalent to finding an f such that $E^2 = (Af - g)^t(Af - g)$ while being subjected to the constraint that Q takes on its minimum value.[1] The Lagrange multiplier solution to the problem is

Mathematical Formulation

The solution to Maxwell's equations for electromagnetic radiation interacting with spherical bodies of arbitrary size and composition was first formulated by Mie in 1908 (for a discussion see Deirmendjian[5]).

numbers. A final complication comes about by realizing that experimentally determined parameters, optical ones in this case, are only estimates of real parameters and are always, therefore, uncertain.

The commonly used transform methods (Fourier, Laplace, etc.) for solving the inverse problem tend to give solutions that oscillate wildly and bear no resemblance at all to actual aerosol size spectra. All these problems with mathematical inversions arise because the kernel functions in the integral equations relating aerosol size spectra to optical vectors are poorly conditioned, as has been discussed extensively by Twomey.[1,3]

Even though inversion theory is fraught with problems of instability, there are ways of obtaining acceptable solutions; one way is to apply numerical smoothing constraints. When smoothing constraints are used in the inversion process, some but not all information pertaining to the aerosol size spectrum can be successfully recovered. Dave[3] has described the problem of recovering aerosol size spectra by constrained inversion of scattered light measurements. Yamamoto and Tanaka[4] have discussed inversion of spectral optical extinction to derive aerosol size spectra. In this paper the two kinds of optical information (scattering and extinction) are combined into a single matrix; this has increased the amount of detail that can be derived about the size spectrum of particulates.

	Size distribution vector						Optical vector				

Fig. 1. Kernel function in the integral Eq. (10); $i = 1, II = 10^4$ cm; $r = 50 \times 10^{-12}$ g cm^{-3}; $\rho = 1$ g cm^{-3}, and $\lambda_0 = 700$ nm.

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Fig. 5. Recovered aerosol size spectra for three independent noisy input vectors with 2.5% random noise. The error bars are the standard deviations for f, computed from twenty inversions of noise-degraded measurements.

Fig. 2. Examples of the effect of varying the smoothing parameter γ on the recovered aerosol size distribution function. The optimum γ for the modeled experiment (2.5% rms) is about 10^{-4}. Too small a γ overconstrains the solution and causes oscillations in the inverted f, while γ being too large results in oversmoothing and the suppression of high frequency structure.
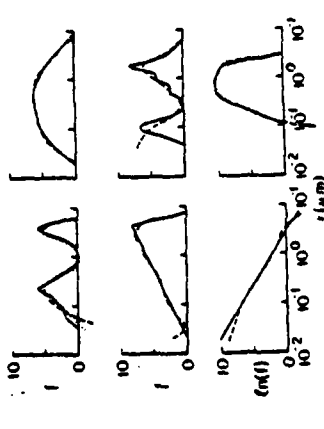
Fig. 3. Examples of recovered aerosol size distribution functions. The optical vectors were degraded with 2.5% rms noise before being inverted. Solid line indicates the actual function f; dotted line indicates f derived by inversion (The upper left hand block shows two inverted noisy vectors).
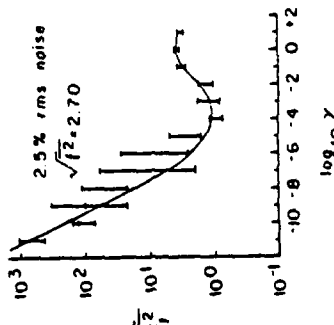
Fig. 4. The rms deviations $(E)^{1/2} = (f - \bar{f})^{1/2}$ between actual and inverted size distribution vectors as a function of the smoothing parameter γ. The error bars show the range of rms deviations recovered when twenty independent noise-degraded (2.5% rms noise) optical vectors were inverted. The norm of the vector f is 2.7. Most of the deviation in the recovered solution occurred in the small particle size range, $r < 0.1\,\mu m$, where the information content is poor. Values of γ near 10^{-4} provide the most accurate inversions.

I.E.4.

ON THE ORIGIN AND TRANSPORT OF THE WINTER ARCTIC AEROSOL*

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MIDLATITUDE POLLUTION AEROSOL IN THE ARCTIC

The study of Arctic air chemistry is still in its infancy. Until very recently, only fragmentary data on trace gases and aerosols from the Arctic were available. During the last 2-3 years, however, a small but systematic study of the Barrow, Alaska aerosol has been undertaken at the University of Rhode Island. The results have shown, contrary to all expectations, that the atmosphere of Arctic Alaska contains an abundance of pollution-derived aerosol, particularly during the winter half-year. This is in great contrast to the Antarctic, which has one of the cleanest atmospheres yet studied. In this article we first summarize the new Arctic data, which have totally altered our understanding of the Arctic atmosphere, and then attempt to demonstrate that they are quantitatively consistent with a midlatitude origin followed by rather direct transport to the Arctic. We conclude with a series of arguments that Europe is the main source region for the winter Arctic aerosol.

Since September 1976 we have been continuously sampling the Barrow aerosol, in conjunction with the University of Alaska and the National Oceanic and Atmospheric Administration. High-volume samples are taken for 3-7 days on 20 × 25-cm (8 × 10-inch) Whatman No. 41 cellulose filters, which are then analyzed chemically for a variety of natural and anthropogenic constituents. In this article we concern ourselves with the results of three constituents— V (more properly excess, or noncrustal, V), sulfate (more properly excess, or nonmarine, sulfate), and ^{210}Pb. Noncrustal V is used as an indicator of midlatitude pollution emissions. Nonmarine sulfate, a major constituent of the Barrow aerosol, has a variety of sources, but the sulfate reaching Barrow during winter seems to have predominantly anthropogenic sources. ^{210}Pb is a natural daughter product of the radioactive decay of gaseous ^{222}Rn emitted from continents.

Barrow is a typical Arctic village. It has a population of about 2,500 and is situated at 71° north latitude on the shore of the Arctic Ocean. The Barrow area is snow covered for roughly nine months each year, September through May. Mean temperatures are about -25°C in winter and about 3-5°C in summer. The nearest town of any size is Prudhoe Bay, about 300 km to the east-southeast.

The first two years of data from Barrow have shown that its aerosol changes drastically from summer to winter in a reproducible fashion. The Arctic winter aerosol is at least an order of magnitude more concentrated than the summer aerosol. The precise winter-summer ratio depends on the site and chemical species; at Barrow it is

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about 15 for ^{210}Pb, 20 for sulfate, and nearly 50 for V. Monthly mean concentrations of sulfate and V for 1976-77 are shown in FIGURE 1. The overall pattern is one of a rapid increase during fall to a general winter maximum, followed by a similarly rapid decrease in late spring. Other Arctic sites, for which we have less detailed information, such as Spitsbergen, Bear Island, and Greenland, also show winter maxima, which indicates that this phenomenon is Arctic-wide. Where systematic, multielemental data are available, as at Barrow, further details within the broad winter maximum may be resolved. The strikingly similar patterns of sulfate and V, each with an early winter plateau and spring maximum in March, are, we believe, typical of the Arctic winter aerosol as a whole, at least at this site.
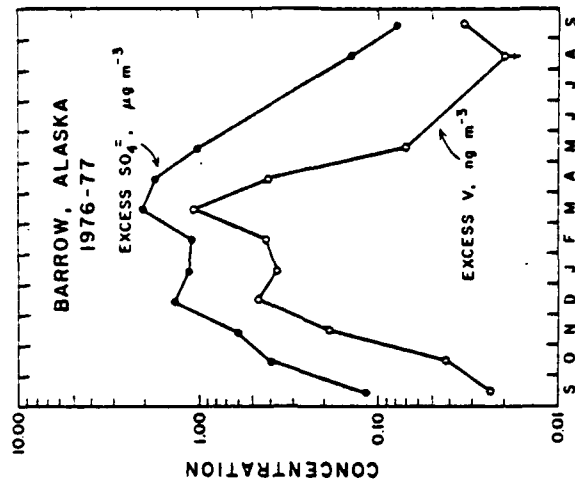
FIGURE 1. Monthly mean concentrations of excess sulfate and vanadium at Barrow, Alaska, 1976-77.

The winter-summer contrast of the Barrow aerosol manifests itself in other ways as well. Unpublished data from J. Rosen and D. Hofmann of the University of Wyoming show that the concentrations of all particles larger than radius 0.15 μm increase by roughly an order of magnitude throughout the depth of the Arctic troposphere during winter, which is a valuable extension of our surface data. The color of the Arctic aerosol changes in winter—summer filters are typically colorless, whereas winter filters are gray. Winter is also the season of Arctic haze, but in summer the Arctic skies are extremely clear, as one might intuitively expect for the entire year.

The winter-summer contrast of the Arctic aerosol is much greater than in
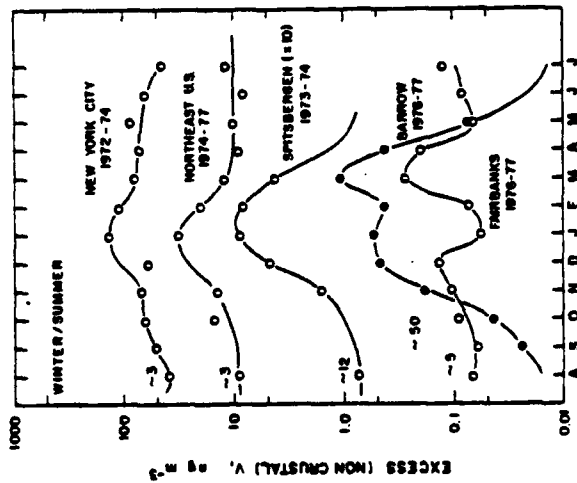
FIGURE 2. Monthly mean concentrations of excess vanadium at five Arctic and non-Arctic sites.

midlatitudes. This is shown for V in FIGURE 2, where seasonal patterns from New York City, the Northeast United States, Spitsbergen, Barrow, and Fairbanks are given. Variations at the midlatitude sites (including Fairbanks) are factors of 3–5, whereas the two Arctic sites have factors of 10–50. This tells us that transport from midlatitudes to the Arctic (which we show below must occur) is more effective in winter than in summer. Another interesting feature of this plot is the nature of the airbanks aerosol, which, in spite of being only 800 km south of Barrow, appears to be much more midlatitude than Arctic in character.

The winter Barrow aerosol appears to be much more pollution-derived than its summer aerosol. This is shown in FIGURE 3, where aerosol-crust enrichment factors of V and Mn are seen to have a strong maximum in winter. Interestingly, the enrichment factors of both elements are 2–3 times lower in Barrow during winter than they are in the suspected midlatitude source regions, a change which seems to indicate that the aerosol picked up some crustal material en route.

The midlatitude origin of the Barrow aerosol is attested to by the presence of excess V. This element is a most useful one in atmospheric chemistry, because its pollution component (as measured at Barrow) comes nearly exclusively from combustion of residual oil,[1] which is too viscous to be easily handled in cold climates and so is burned nearly exclusively in temperate latitudes.

The pollution character of the Barrow aerosol is also indicated by the high absolute concentrations of sulfate. Each spring the monthly mean concentrations of sulfate

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reach or exceed 2 μg m^{-3}, a remarkably high value. By comparison, mean winter sulfates in the northeastern United States are only about 5–6 μg m^{-3}. Similarly high sulfate concentrations are also observed at Spitsbergen.

Another interesting feature of the Barrow aerosol is its highly secondary nature. This is shown here by the sulfate/V ratio, which, as shown in FIGURE 4, is one order of magnitude higher in the Arctic than in typical midlatitude source regions. Interestingly, the values for the intermediately located northern Norway seem to be also intermediate. In our opinion, the source of the excess sulfate of the Arctic must be oxidation of the gaseous SO$_2$ of midlatitude polluted air masses. This oxidation can occur along the path to the Arctic, and shows that the Arctic aerosol is an aged version of the midlatitude aerosol. The factor of increase in the sulfate/V ratio is also reasonable, because midlatitude polluted air masses have several times more SO$_2$-S than sulfate-S. This point will be explored in more detail in the second half of this article.

In general, ^{210}Pb collected on our Barrow filters has three components: a "supported" fraction, which is in equilibrium with nuclides of the ^{238}U series of the soil fraction of the Barrow aerosol; an "unsupported," or excess, component, which was also present in those same parent soils and was simply carried to Barrow with them; and an excess component formed from ^{222}Rn en route. Because the specific activity of total ^{210}Pb that we measure in the Barrow aerosol (15 × 10^6 dpm g^{-1} of soil aerosol) is much larger than the specific activity of supported ^{210}Pb in soils (about 1 dpm g^{-1}) or

FIGURE 3. Monthly mean aerosol crust enrichment factors for V and Mn at Barrow, Alaska, 1976–78.

concentrations are nearly double the New York values, a completely unexpected result. We suggest that the most likely explanation for the high ^{210}Pb concentration at Barrow is an unusually long residence time for the Arctic aerosol, so that ^{210}Pb produced in the Arctic (from parent ^{222}Rn, which must be about an order of magnitude less concentrated there than near New York) accumulates with little loss due to precipitation. We return to this point later.

One can immediately see the parallels between the SO_2-sulfate and ^{222}Rn-^{210}Pb systems: both have gaseous precursors with residence times of a few days that are emitted over continents, and both are converted into submicron secondary aerosols. Both daughter products are enriched in the Barrow winter aerosol by an order of magnitude relative to what was expected. The parallelism between ^{210}Pb and sulfate at Barrow is even greater: they are extremely well correlated (FIGURE 6), with a

correlation coefficient of 0.95 or greater. This strongly suggests that sulfate, itself a major fraction of the Arctic aerosol, is serving as a substrate for the ^{210}Pb. But we still do not understand why the correlation is so good.

In summary, then, the Barrow winter aerosol seems to be derived from midlatitude polluted air masses that are efficiently transported to the Arctic. Primary and secondary aerosol components are both observed, with the secondary components being enriched relative to the primary ones by an order of magnitude compared to midlatitudes. Sulfate is the principal secondary component measured so far. Because it may represent most of the mass of the aerosol, the Arctic winter aerosol may – to a first approximation – be considered secondary, that is, derived from midlatitude pollution gases.

typical excess ^{210}Pb in soils ($\leq$100 dpm g^{-1}), we believe that essentially all of the total ^{210}Pb that we measure has been formed by decay of ^{222}Rn en route.

FIGURE 5 shows seasonal patterns of total ^{210}Pb at New York City and Barrow.³ At New York, the mean ^{210}Pb concentration is about 15 fCi m^{-3}, with no strong seasonal variation. At Barrow, its annual mean is lower; but during winter the ^{210}Pb

FIGURE 4. Winter $SO_4^=$/V ratios at Arctic and non-Arctic sites.

FIGURE 5. Monthly mean ^{210}Pb concentrations, New York City and Barrow, Alaska.³

FIGURE 6. ^{210}Pb vs. excess sulfate, Barrow, Alaska.

TRANSPORT OF AEROSOL TO THE ARCTIC DURING WINTER

In the second half of this article we propose a simple transport model for the winter Arctic aerosol and its precursor gases. Because the study of the Arctic aerosol is yet so new, too few data exist to warrant the development of a detailed model. We believe, however, that the available data do place reasonable constraints on the origin and pathway of the Arctic winter aerosol and gases. The spirit of the following section is then to extract the maximum from the present data and to use these first semiquantitative conclusions as a guide for future studies.

In general, the concentration of a constituent of the Arctic aerosol will depend on its concentration in the midlatitude source region, the speed of transport to the Arctic, and the rate of removal along the way. Each of the above quantities will have seasonal variations. As has been seen in FIGURE 2, the fact that the seasonal variation of V at Barrow is a factor of 50, as compared to a factor of only 3–4 at the source, implies that the seasonal variation of transport plus removal is more than an order of magnitude, i.e., several times greater than the source variations. Thus, transport and removal are important in determining the concentration of the aerosol at Barrow. We will now examine these two processes in more detail, in particular for the winter half-year.

Consider first the speed of transport from midlatitudes to the Arctic. The best indicator of this parameter is the concentration of ^{222}Rn in the Arctic. We are aware of data from Spitsbergen,[4] and from Kodiak and Wales, Alaska,[5] but have none from Barrow itself. These data all show that ^{222}Rn concentrations are several times higher in winter than in summer, the actual factors being 10 for Spitsbergen and 4 for the Alaskan sites. This suggests that the speed of transport from continental regions to the Arctic is faster in winter than in summer. On the basis of this and the timing of the onset and disappearance of the winter maximum of aerosol at Barrow, a very general air-mass picture of transport to the Arctic was proposed,[6] which has been called the coupling-decoupling hypothesis.

This picture considers the Northern Hemisphere atmosphere to be comprised of polar and tropical air masses, which are separated by the polar front. Air flow within each air mass is less restricted than air flow across the polar front, which by its very nature represents a barrier to exchange of air. In summer, when the polar front is located in its most northerly position, it is between the midlatitude source areas and Barrow, so that Barrow and the rest of the Arctic are cut off, or decoupled, from the midlatitudes. Air reaching the Arctic from midlatitudes is very aged and cleansed of contaminants. In winter, on the other hand, the polar front lies just south of the midlatitude source areas, so that they and the Arctic are essentially in the same air mass. Air flow between the two areas is much freer than in the summer, and Arctic aerosol concentrations are consequently much higher.

It is interesting that the polar front does not migrate smoothly between its summer and winter positions. Rather, its motion is quite sudden, during roughly October–November and May–June. These are just the times when the Arctic aerosol changes its character.

Obviously, such a simple, broad picture of the basic forces controlling aerosol transport to the Arctic cannot supply any details of the specific sources or of the mechanics of transport, nor is it intended to. But it is useful as a first picture of the seasonal influences on transport.

Concerning the effect of removal rates, we have already seen that the high concentration of ^{210}Pb at Barrow during winter almost certainly has to be caused by longer residence times in the Arctic than in midlatitudes. The same argument applies to the sulfate-SO_2 system. The main points about residence times in the midlatitudes vs. those of the Arctic can be derived, we feel, from FIGURE 7, which gives monthly mean precipitation rates for a typical midlatitude site (Providence, Rhode Island) and Barrow. (We assume that precipitation controls the residence time of atmospheric aerosols.)

FIGURE 7 has two main features. First, Providence has 10 times the annual precipitation that Barrow does. For this reason, we believe that the residence time of aerosol near Barrow ought to be approximately 10 times longer than in midlatitudes. Second, the seasonal patterns of precipitation at the two sites are quite different. Providence has its precipitation distributed quite evenly over the year, with only gentle

PRECIPITATION, mm mo⁻¹

120 80 40 0

PROVIDENCE, RI (1090 mm)

BARROW, AK (104 mm)

J F M A M J J A S O N D
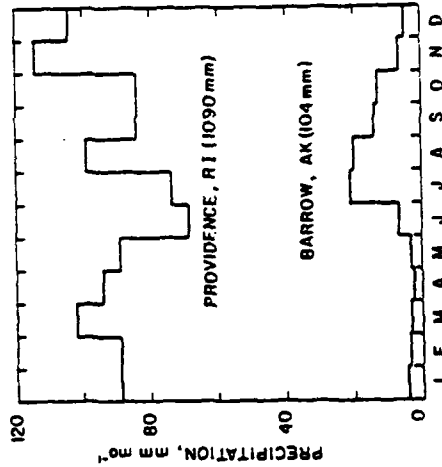
FIGURE 7. Monthly mean precipitation, Providence, R.I., and Barrow, Alaska.

maxima in spring and fall. Barrow, by contrast, has a broad winter and spring minimum followed by a strong summer maximum. The difference between spring and summer precipitation rates is about an order of magnitude. According to FIGURE 7, winter Arctic residence times ought to be roughly 20 times higher than in the midlatitudes, if precipitation is indeed the controlling factor. In summer, on the other hand, Arctic residence times ought to be only 4 times higher than those of midlatitudes.

An equally important question is the midlatitude source area for Arctic aerosol. Regions to be considered include Japan/Korea/China, the northeastern United States, Europe/UK, and possibly central Siberia. Likely pathways from these sources, based on major air flow patterns, are shown in FIGURE 8. The pathway from eastern Asia corresponds to that derived for an episode of Asian dust at Barrow in the spring

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We are thus left with the northeastern United States and Europe, the two sources that from the beginning seemed most likely. Very recent evidence suggests that the United States may be a relatively weak source compared to Europe. For one thing, transport from this source to the Arctic via Iceland and Spitsbergen is quite analogous to the Asia/Pacific case. If the cleansing effect of the Icelandic low is at all similar to that of the Aleutian low, air from North America ought to be very clean by the time it reaches the Arctic. This indeed seems to be the case. Recent data for northern and southern Greenland during the winter of 1977–78 (obtained in cooperation with the Danish Meteorological Institute) show that mean concentrations of V at both sites were several times lower than in Spitsbergen (data obtained in cooperation with the Norwegian Institute for Air Research) and Barrow (FIGURE 9). Thus, the source of V at these latter sites does not seem to be North America. Another piece of evidence is the proportions of V and Mn. At Spitsbergen and Barrow, Mn is more abundant than V, as it is in the European aerosol. In the northeastern United States, on the other hand, V is several times more abundant than Mn. But perhaps the most powerful evidence for a European source is a series of calculations of the evolution of the composition and concentration of the aerosol as it moves from midlatitudes to the Arctic, our transport model, which is presented below.

These calculations were performed to see whether the actual concentrations of V.
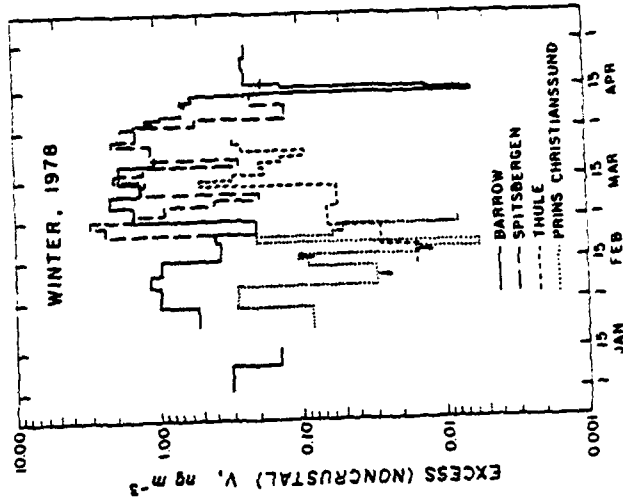
FIGURE 9. Concentrations of excess vanadium at four northern sites, 1977-78.

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FIGURE 8. Possible main pathways of aerosol transport to the Arctic.

of 1976,[1] but would probably be about the same for transport of pollution aerosol. The pathway from the northeastern United States basically follows the jet stream through the Icelandic low, with a branch around the western side of Greenland corresponding to periods when the Baffin Island low is strong. The significance of this latter branch is not yet certain. The pathway northward from central-Europe is roughly that followed by pollution episodes observed throughout Scandinavia. Lastly, the hypothesized pathway from central Asia is based on motions observed around the Siberian high. This path is also highly uncertain.

Until recently, we had no clear indication of which source or combination of sources might be most important for the Arctic during winter. Within the past few months, however, several pieces of data have emerged that seem to be narrowing the field. First, we see no evidence that central Siberia can be an important source, in spite of the rapid industrialization of the region. Population densities are simply too low there. Second, it now appears that air masses coming up to Barrow and Fairbanks from the south, which have come from eastern Asia via the Pacific, are very clean compared to Arctic air coming from the north (see episode during mid-April 1978 in FIGURE 9). We interpret this to mean that Japan/Korea/China is not an important source region, at least during winter. This is reasonable in light of the fact that air from this region has traveled about 7,000 km over the stormy Pacific Ocean, where removal of aerosol must be rapid.

sulfate, and 210Pb at Barrow could be accounted for by reasonable source concentrations, transport times, oxidation and decay rates, removal rates, and dilutions. The results showed that this was indeed possible. Before presenting these calculations, we wish to make it clear that they are quite elementary and will doubtless be revised. No vertical motions are incorporated, and many of the parameters are imprecisely known. Yet even this simple approach yields essentially only one "solution," i.e., the combination of source area, transport time, pathway, and rate constants that effectively reproduces the Barrow data. All parts of this solution are physically reasonable. Further, there exist sufficient constraints to argue effectively against other solutions that we have tried.

Our model starts with an air mass whose bulk chemical characteristics are derived from surface measurements in a particular midlatitude polluted region, normally either the northeastern United States or western Europe. This air mass is then allowed to age for a period of time designed to simulate transport to the Arctic. During this time, gases are allowed to decay and/or react, particles and gases are removed, and the air mass is steadily diluted by external air having no aerosol or trace gases. For ease of calculation, discrete one-day time steps are used. The chemical characteristics of the air mass after a preset number of days are compared with the observed winter means for Barrow.

The controlling equations are:

$$V_{n+1} = V_n e^{-(k_p)_n \Delta t}(1/D_{n+1})$$

$$(SO_4^-)_{n+1} = e^{-(k_p)_n \Delta t}\left[(SO_4^-)_n + \frac{1.5(k_m)_n(SO_2)_n}{\lambda_n}(1 - e^{-\lambda_n \Delta t})\right](1/D_{n+1})$$

$$(^{210}Pb)_{n+1} = e^{-(k_p)_n \Delta t}\left[(^{210}Pb)_n + \frac{8.41 \times 10^{-5}(Rn)_n}{[0.1824 - (k_p)_n]}(1 - e^{-[0.1824 - (k_p)_n]\Delta t})\right](1/D_{n+1})$$

$$(SO_2)_{n+1} = (SO_2)_n e^{-(k_m + k_w)_n \Delta t}(1/D_{n+1})$$

$$(Rn)_{n+1} = (Rn)_n e^{-k_{Rn} \Delta t}(1/D_{n+1})$$

where:

$$\lambda_n = (k_m)_n + (k_w)_n - (k_p)_n$$

and:

k_m – rate constant for combined wet and dry removal of SO2;
k_w – rate constant for dry oxidation of SO2;
k_p – rate constant for removal of (submicron) aerosol;
Δt – time step of 1 day;
D_n – dilution factor ($D_n \geq 1$).

The subscripts n, $n + 1$, etc., refer to the number of days of transport.

Calculations are executed as follows: Initial values for sulfate, SO2, V, 222Rn, and 210Pb are entered, together with initial and final values for k_p, k_m, k_w, and D_n ($D_0 - 1$). The k's may decrease exponentially between initial and final values or remain

constant, and D_n may increase exponentially or remain constant. First, D_{n+1} is calculated as follows:

$$D_{n+1} = \sqrt{1/D_n}$$

where N is the number of days of transport. Next, V_{n+1}, $(SO_4^-)_{n+1}$, and $(^{210}Pb)_{n+1}$ are calculated, followed by $(SO_2)_{n+1}$ and $(Rn)_{n+1}$. Finally, values of k_{n+1} are calculated using one of the following equations:

$$k_{n+1} = k_n e^{[\ln(k_0/k_N)]/N} - k_n e^{[\ln k_0 - \ln k_N]/N} \quad \text{(if exponential decay)}$$

$$k_{n+1} = k_n - \frac{(k_0 - k_N)}{N}\Delta t \quad \text{(if linear decay)}$$

This sequence is repeated N times.

We now present a series of typical results of these calculations, designed to illustrate in a simplified way how European, but not North American, air masses can account for the Barrow winter aerosol. For all these calculations we have used a value of 20 days for the transport time to Barrow—a somewhat arbitrary figure. At present, the best that can be said is that the travel time is somewhere between 10 and 30 days. The best paths that we can construct from both the United States and Europe to Barrow seem each to be about 13,000 km long (see below for a discussion of the best European path). For mean wind speeds of 7 and 10 m s⁻¹, this corresponds to travel times of 21 and 15 days, respectively. Most importantly, though, it will be seen below that the total travel time to the Arctic is not an important variable, because the composition of the aerosol, i.e., the ratio between primary and secondary aerosol, becomes fixed after about 10-15 days of transport.

FIGURE 10 shows results for air masses of the northeastern United States. This first calculation is particularly simple, with the three k's constant at 0.2 day⁻¹ (corresponding to residence times of 5 days for particle removal, oxidation of SO2, and removal of SO2) and no dilution during transport. These assumptions are of course not justified, because at least the two k's that depend on precipitation and turbulence (k_p and k_w) will decrease during transport, and there is sure to be a dilution as well. Initial conditions were taken as 222Rn - 170 pCi m⁻³ (winter mean for Chester, N.J.);[8] 210Pb - 15 fCi m⁻³ (winter mean for New York City);[7] SO2 - 16 μg m⁻³ (survey of available literature); SO4⁻ - 6 μg m⁻³ (also available literature); and V - 22 ng m⁻¹ (our own data).

FIGURE 10 shows that V is slightly underestimated (0.4 ng m⁻¹ instead of about 0.6 ng m⁻¹), but that sulfate and 210Pb are seriously underestimated. To remedy this, one can incorporate exponentially decreasing k's (initial and final residence times of 5 and 50 days for k_m and k_w, and 4 and 50 days for k_p) into a second series of calculations, starting with the same initial conditions.

The Barrow aerosol cannot be derived from midlatitude aerosols by using constant aerosol residence times. The residence time required to reproduce V and SO4⁻ is about 10 days, which is too long to be defended for midlatitudes. Further, use of this value produces a 210Pb/SO4⁻ ratio that is too low. Testing a series of possible constant residence times has shown that no value can produce both the correct concentrations of V, sulfate, and 210Pb as well as their correct ratios. On the other hand, residence

Left column (page 498)

Figure 10 labels: INITIAL CONDITIONS: NE USA — WINTER VALUES AT BARROW — ^{210}Pb — ^{222}Rn — SO$_4^=$ — V — ^{210}Pb, fCi m^{-3} — ^{222}Rn, pCi m^{-3} — SO$_4^=$, μg m^{-3} — V, ng m^{-3} — NO DILUTION — SO$_2$ — SO$_2$, μg m^{-3} — CONCENTRATION — TRAVEL TIME, days
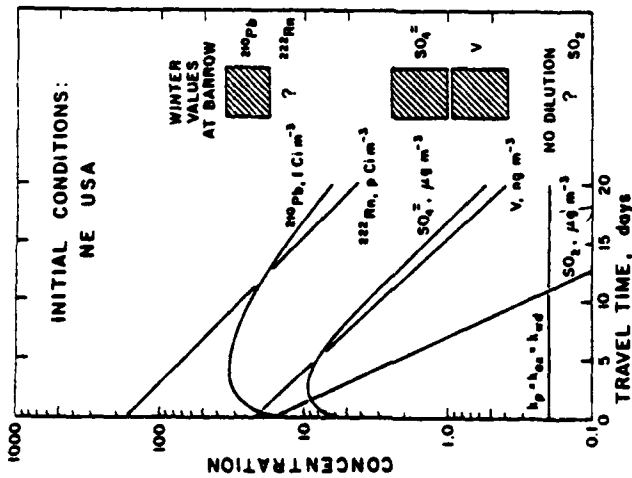
FIGURE 10. Modeled transformation of a polluted air mass as it travels from the northeastern United States to the Arctic.

times that increase with travel time satisfy these conditions rather easily, as will be seen below. This approach also makes good physical sense: (a) precipitation at Barrow is much less than in midlatitudes; (b) our preliminary calculations of residence times for the Barrow aerosol, based on trace elements in aerosol and snow there, yield residence times of roughly 30–100 days; and (c) calculations of deposition of ^{210}Pb in the Arctic and in midlatitudes show that its residence times are about an order of magnitude longer in the Arctic.

In order to adjust the final V value to 0.6 ng m^{-3}, a dilution factor, or D_N, of 6 can be used. These calculations, displayed in Figure 11, show that sulfate and ^{210}Pb are still much too low. Sensitivity calculations have shown that the results of this kind of calculation are very insensitive to the values of k_w and only moderately sensitive to k_w. We have therefore concluded that the only way that the Barrow values for sulfate and ^{210}Pb can be reached is by altering the source conditions, i.e., starting with an air mass with more SO$_2$ and ^{222}Rn. (Initial values of sulfate and ^{210}Pb are unimportant to these calculations, because they are diminished by factors of 20–30, as is V.)

To match the sulfate at Barrow, or more properly stated, to match the SO$_4^=$/V ratio at Barrow, initial SO$_2$ has to be doubled to about 32 μg m^{-3}. This is very similar to European conditions, where area-wide SO$_2$ is roughly 30 μg m^{-3} during winter.

Right column (page 499)

Indeed, taking European initial conditions (^{222}Rn estimated at 100 pCi m^{-3}; ^{210}Pb estimated at 15 fCi m^{-3}, SO$_4^=$ – 32 μg m^{-3}, SO$_2$ – 6 μg m^{-3}) and letting all 3 k's decrease exponentially from 0.2 day^{-1} to 0.02 day^{-1} after 20 days, and keeping the dilution factor at 6, as before, both the V and sulfate of Barrow are reproduced. ^{210}Pb, however, is still several times too low.

In order to account for the actual ^{210}Pb found at Barrow, its initial European value would have to be set at about 600 pCi m^{-3}, a value that is unacceptably high. The problem is intensified when one realizes that Europe, being essentially a broad peninsula, probably has ^{222}Rn concentrations considerably lower than the 100 pCi m^{-3} used in Figure 12. We found, however, that a prolonged source of ^{222}Rn (keeping its initial value constant for the first few days), or still better, a source that increased the ^{222}Rn during the first few days, greatly increased the final ^{210}Pb concentrations at Barrow. One somewhat arbitrary system that worked is shown in Figure 13. Here, an initial ^{222}Rn concentration of 50 pCi m^{-3} was taken for the first 5 days, followed by 150 pCi m^{-3} for the next 5 days. After this, the ^{222}Rn decayed normally. This system reproduced concentrations of V, SO$_4^=$, and ^{210}Pb, as well as their ratios.

This suggests to us that the polluted European air mass passes over a much less polluted continent on its way to the Arctic. The physical interpretation is obvious from

Figure 11 labels: INITIAL CONDITIONS: NE USA — WINTER VALUES AT BARROW — ^{210}Pb — ^{222}Rn — SO$_4^=$ — V — ^{210}Pb, fCi m^{-3} — ^{222}Rn, pCi m^{-3} — SO$_4^=$, μg m^{-3} — V, ng m^{-3} — SO$_2$ — SO$_2$, μg m^{-3} — DILUTION · 0.944 PER DAY · 6 FOR 20 DAYS — CONCENTRATION — TRAVEL TIME, days
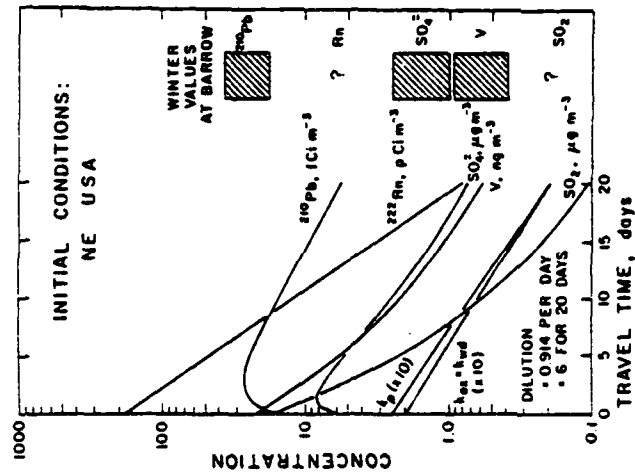
FIGURE 11. Modeled transformation of a polluted air mass as it travels from the northeastern United States to the Arctic.

over Novaya Zemlya, then splitting (in the mean) into two branches, one that flows southward along the east coast of Greenland and another that flows westward along northern Greenland and the Canadian Arctic Islands, eventually reaching Barrow.

Here we emphasize again that the above picture must be regarded with extreme caution. It is a first approach only, and is intended only to test whether it is possible, using the best available information, to synthesize a mean pathway from Europe to Barrow that makes good physical sense and that satisfies the aerosol data at both places. The truth is undoubtedly much more complicated. For example, the European air probably rises during its passage northward, and by a height of about 700 mb (3 km), the Arctic high has disappeared, having been replaced by an Arctic-wide low. At this level, European air will come to Barrow from the west, not the east.

In summary, then, we have attempted to demonstrate that observed winter concentrations of V, sulfate, and ^{210}Pb are indeed consistent with a midlatitude pollution source and transport for roughly 20 days with reasonable values of transformation rate constants. Considering the relatively small number of measurements available, there seems to be a remarkable set of constraints imposed upon the system, such as the need for increased residence times in the Arctic and, perhaps most
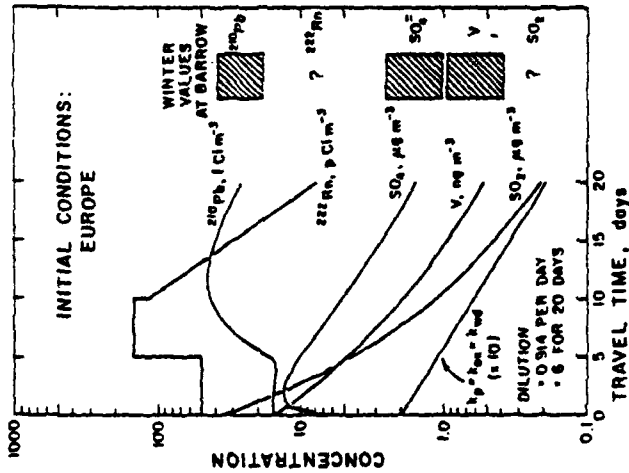
FIGURE 13. Modeled transformation of a polluted air mass as it travels from Europe to the Arctic.

FIGURE 12. Modeled transformation of a polluted air mass as it travels from Europe to the Arctic.

a map and from knowledge of mean air flows over Europe—the air mass moves to the northeast, into European Russia (where population density is a few times less than in central Europe), then at some point turns northward to the Arctic.

Several pieces of independent evidence seem to confirm this view. First, the mean European winter 850-mb wind, as calculated by Bolin and Persson[10] for air over northern England, is indeed to the east-northeast. For air leaving central Europe, this would correspond to a mean path somewhere between Moscow and Leningrad. For evidence that the air flow after this should curve more to the north, we invoke the blocking influence of the winter Siberian high, which diverts this air almost straight to the north (see Figure 3.30 in Trewartha[11]). Thus, we envision this aged European air leaving the Eurasian landmass somewhere between northern Scandinavia and the Taimyr Peninsula, i.e., in the general vicinity of Novaya Zemlya.

After this, we can only guess about the trajectory. According to FIGURE 14, which shows the annual mean surface pressure distribution for the Arctic (after Dunbar and Wittman[7]), surface flow in the Arctic is broken into two distinct regions— the Greenland-Spitsbergen-Novaya Zemlya region, which is dominated by the Icelandic low and where cyclonic flow is the rule, and the Alaskan side, where the Arctic high dominates and anticyclonic flow is the rule. It is easy to imagine European air passing

FIGURE 14. Annual mean surface pressure distribution for the Arctic (after Dunbar and Wittman[12]).

important, the apparent identification of Europe rather than the United States as the prime source, with a mean winter transport path over the European part of the Soviet Union.

So far, we have been unable to find any other realistic solution to the equations of transport than the source and path just proposed. We will continue to search for others, however, and at the same time will be carefully studying the model for its sensitivity to variations in the different parameters. These results already suggest further experiments that should be carried out to test our hypothesis.

SUMMARY

Recent research on the Arctic aerosol has revealed that it is an order of magnitude more concentrated in winter than in summer throughout most of the troposphere, is gray in winter, but colorless in summer (on filters), and is much more enriched in pollution-derived constituents like V, Mn, and sulfate in winter than in summer. The winter Arctic aerosol also appears to be highly secondary in nature, containing unusually high concentrations of gas-to-particle conversion products, such as sulfate and ^{210}Pb. On this basis, then, the Arctic aerosol in winter seems to be strongly pollution-derived. A simple transport model is presented, from which it is shown that the actual concentrations of V, sulfate, and ^{210}Pb at Barrow, Alaska are quantitatively consistent with polluted European air masses as the source, followed by transport over European Russia and then to the north.

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I.E.5.

ARCTIC HAZE: PERTURBATION OF THE POLAR RADIATION BUDGET

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ARCTIC HAZE: PERTURBATION OF THE POLAR RADIATION BUDGET*

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INTRODUCTION

Arctic haze refers to turbid air in the northern polar regions. It was first noticed and commented on in 1956 by Murray Mitchell, Jr.[1] who more or less described it as being an amorphous haze extending from the northern Alaskan coasts to the highest polar latitudes. In the period 1971-75, Shaw and Wendler,[2] Shaw,[3] and Holmgren et al.[4] measured anomalously high values of optical extinction through the atmosphere at McCall Glacier in remote northeastern Alaska and at Point Barrow on the northern tip of Alaska. The high values of extinction (haze optical depth ~ 0.1-0.2 at 500 nm wavelength) were extremely puzzling, especially since the atmospheric optical extinction underwent a seasonal variation in the opposite direction from what one would expect: the values of optical extinction were large during the months March-April, when the oceans and surface are covered with ice and snow, but were smaller in summer. This behavior is distinctly anomalous, since turbidity at most locations is at its greatest in summer[5,6,7] and at its minimum in middle or late winter.

Since 1975, arctic haze has been studied extensively in Alaska by Rahn et al.[8] Its composition, size distribution, and other information indicate that it may not be natural but may be associated with pollution emitted at the middle latitudes. Chemical studies indicate that sulfates and carbonaceous particles are present in the haze. Vanadium and other elements indicating a pollution source are strongly enriched, for example. There are other indications that the haze is pollution-derived (see Rahn and McCaffrey[9] for an account of the properties and probable origin of the haze).

The subject of this paper is a somewhat specialized topic pertaining to arctic haze—we will consider the perturbation that arctic haze makes on the polar radiation balance. It will be shown that the presence of haze at the time of the spring equinox has a dramatic effect on the arctic radiation balance.

THE RADIATION BALANCE CALCULATION

The objective of the study was to theoretically model the response of the atmospheric and earth-atmosphere radiation budget when a homogeneous layer of haze is introduced into the air poleward of the Arctic Circle. We used a complete

solution to the equations of transfer that accounted for high-order scattering and for the reflection of light from the surface ice and snow. The reported solutions apply only to visible-band radiation; the effects of arctic haze on infrared heat balance will later be shown to be small.

The solutions reported here were obtained for a monochromatic wavelength band centered at 500 nm. We made the assumption that 500 nm would be a representative wavelength to use for the visible-spectrum radiation balance. This is an approximate technique, but one that will be rectified in the future by weighting multiwavelength calculations against the solar spectrum. The numbers will change slightly when a more exact treatment is given, but the basic conclusions reached will remain the same.

COMPUTATIONAL SCHEME

The transfer of radiation through an absorbing/scattering atmosphere is described by the radiative transfer equation; in this case, for a plane-parallel, horizontally homogeneous geometry. An azimuthally integrated version of the transfer equation is:

$$-\mu \frac{dI(\tau,\mu)}{d\tau} = I(\tau,\mu) - \frac{\omega(\tau)}{2} \int_{-1}^{1} \bar{P}(\tau,\mu,\mu') I(\tau,\mu') d\mu' \quad (1)$$

where I is the azimuthally averaged intensity, μ is the cosine of the zenith angle, ω is the single scattering albedo, and τ is the optical thickness. The azimuthally integrated phase function is:

$$\bar{P}(\tau,\mu,\mu') = \frac{1}{2\pi} \int_{0}^{2\pi} P[\mu\mu' + (1-\mu^2)^{1/2}(1-\mu'^2)^{1/2} \cos\phi] d\phi$$

where P is the phase function satisfying the usual normalization:

$$\frac{1}{2} \int_{0}^{\pi} P(\cos\theta) \sin\theta \, d\theta = \frac{1}{2} \int_{-1}^{1} \bar{P}(\mu,\mu') d\mu' = 1 \quad (2)$$

In general, the single scattering albedo and the phase function depend on the optical depth, τ. For the purpose of this preliminary study, we assume a constant mixing ratio of aerosols to molecules. This approximation has the effect of making ω and P independent of τ. Herman and Browning[10] showed that the vertical inhomogeneity has very little effect on the reflected flux. We conjecture that if altitude profiles of the energy deposition (heating/cooling rate profiles) are not required, the vertical distribution of the aerosols will not be important. Thus, our model is adequate for determining the integrated quantities: albedo, transmissivity, and total absorption within the atmosphere.

The discrete ordinate approximation[11,12] consists of replacing the integral term in (1) by a numerical quadrature containing $2n$ terms. This leads to a system of $2n$ coupled differential equations for which eigen solutions can be obtained. As shown by Stamnes,[11] this system of equations can be written in matrix form, and the symmetry properties of the phase function can be used to reduce the order of the resulting algebraic eigenvalue problem by a factor of 2.

The numerical implementation is accomplished by using algebraic eigenvalue routines. The program is rapid and reliable.

MODEL ATMOSPHERE

To estimate the effect of adding aerosols of given optical thicknesses and absorbing/scattering properties to a Rayleigh-scattering atmosphere, we make the following assumptions:

1. Constant mixing ratio of aerosols to molecules (see discussion above).
2. The ground is a "Lambert reflector" with albedo A.
3. The aerosols scattered according to a Henyey-Greenstein phase function:

$$P_A(\cos\theta) = P_{HG}(\cos\theta) = \frac{1-g^2}{[1+g^2-2g\cos\theta]^{3/2}} \quad (3)$$

with asymmetry factor:

$$g = \langle\cos\theta\rangle = \frac{1}{2} \int_{0}^{\pi} \cos\theta \, P(\cos\theta) \sin\theta \, d\theta \quad (4)$$

Denoting the Rayleigh scattering optical thickness τ_R and the aerosol optical thickness $\tau_A = \tau_{A,S} + \tau_{A,A}$, where $\tau_{A,S}$ and $\tau_{A,A}$ refer to scattering and absorption, respectively, the single scattering albedo is:

$$\omega = \frac{\tau_R + \tau_{A,S}}{\tau_R + \tau_A} \quad (5)$$

Similarly, the phase function becomes:

$$P(\cos\theta) = (1-b)P_A(\cos\theta) + bP_R(\cos\theta) \quad (6)$$

where $P_R(\cos\theta)$ is the Rayleigh scattering phase function and:

$$b = \frac{\tau_R}{\tau_R + \tau_{A,S}} \quad (7)$$

Clearly, $b = 1$ and $\omega = 1$ refer to a nonabsorbing Rayleigh-scattering atmosphere.

DETERMINATION OF THE OPTICAL PARAMETERS OF ARCTIC HAZE

The necessary parameters to solve the transfer equation for arctic haze conditions are: τ, the optical thickness; ω, the albedo of single scattering; $\langle\cos(\theta)\rangle$, the asymmetry factor; and A, the underlying surface albedo. These four parameters were estimated from multiple-wavelength optical measurements during haze episodes in March-April 1978 at Fairbanks and Barrow. The following experimental measurements were made: (1) the intensity of direct solar radiation at the surface with a precision sun photometer; (2) the sky radiance along the solar almucantar with a photoelectric coronameter and directional-scanning spectrometer; (3) the monochro-

metic downwelling and upwelling diffuse radiation fluxes with specially developed flux sensors; and (4) reflectivity of exposed millipore filters."

Optical thickness was derived according to standard sun photometry methods." The albedo of single scattering, ω, was determined in two ways: (1) by use of Kubla-Monk theory applied to the measurements of exposed filters,[1,3,5] and (2) by comparing measured downwelling diffuse radiation with that calculated by radiative transfer theory for a series of trial values of $\tilde\omega$." The asymmetry factor was determined from the scattering phase function, which itself was derived from the scans of sky intensity along the solar almucantar." There are uncertainties in the derived optical parameters, because some of the techniques were approximate and the optical parameters fluctuated with time. Nevertheless, the values are accurate enough to do preliminary modeling.

During times of moderately heavy haze, the optical parameters were: τ ($\lambda \sim 500$ nm) ~ 0.1–0.2; $\tilde\omega$, 0.5–0.8; (cos (θ)), 0.6–0.8; A, 0.6–0.8. The low values of $\tilde\omega$ indicate

TABLE 1

VISIBLE-BAND RADIATION BALANCE AT 65 AND 75° NORTH LATITUDE DURING VERNAL EQUINOX[a]

	Net Radiation Absorbed (kcal month⁻¹ cm⁻²)					
	65° N			75° N		
	CLR[b]	NAH[c]	AH[d]	CLR[b]	NAH[c]	AH[d]
Earth-Atmosphere	2.13	1.98	3.68	1.22	1.13	2.24
Atmosphere	0	0	1.94	0	0	1.32
Surface	2.13	1.98	1.75	1.22	1.13	0.91+

[a] Solar Constant: 1.96 cal cm⁻² min⁻¹ (8.7 kcal cm⁻¹ month⁻¹) ~ 136 mw cm⁻². Haze optical thickness, 0.2; albedo of single scattering, 0.8; surface reflection, 0.8; haze scattering asymmetry factor, 0.75.
[b] CLR, uncontaminated atmosphere.
[c] NAH, atmosphere charged with nonabsorbing haze.
[d] AH, atmosphere charged with absorbing haze.

that the haze is an absorbing haze, as is evident also from the gray color of the exposed filters and from the presence of what appeared to be carbonaceous particles when the filters were inspected with a scanning electron microscope.

EVALUATION OF THE MODEL

The radiative transfer model was evaluated for latitudes at 65° and 75° N, assuming the haze to be homogeneous with $\tau_H \sim 0.2$, $\tilde\omega \sim 0.8$, $A \sim 0.8$, and (cos (θ)) ~ 0.75. Comparative calculations were also made for a pure Rayleigh-scattering atmosphere. Results are tabulated in TABLE 1.

Arctic haze heats the atmosphere and forces a slight cooling at the surface. The earth-atmosphere system radiative heating increases (when haze is added) by 73% at latitude 65° north and 84% at latitude 75° north. The dramatic change in the amount of visible-band radiation absorbed by the arctic regions, by simply introducing an

optically thin haze layer, can be understood by realizing that (a) the actual amount of radiation absorbed by the Arctic during haze-free periods is small to begin with, for the pack ice is highly reflecting and the sun angle is low, and (b) the long atmospheric ray paths and the multiple reflections between ice and haze both enhance the amount of radiation absorbed by the atmosphere. The net effect is to heat the atmosphere by an amount quite comparable with heating of ice by the sun in spring. At the same time, net radiation absorbed at the surface of the Arctic is decreased (by 22% and 32% at 65 and 75° N, respectively) when haze is introduced. The deficit is mostly absorbed in the atmosphere.

So far, nothing has been said about the interaction between haze and emitted thermal radiation. The optical thickness of arctic haze decreases with wavelength in the visible spectrum as λ^{-2}; by extrapolation, this would suggest a haze optical thickness of only 5×10^{-4} at $\lambda \sim 10$ μ, or 400 times smaller than the optical thickness in the visible region of the spectrum. If the haze is entirely absorbing in the long wave infrared region (as it probably is), the atmosphere would gain 0.01 kcal cm⁻² month⁻¹ (for a radiating temperature of 250°K)—a negligible amount in comparison with the energy deposited by absorption of visible-band radiation.

DISCUSSION

The radiation term represents only a small fraction of the arctic heat budget except for about two months centered around summer solstice (during which time there is no haze). Essentially, arctic heat transfer is a balance between advected heat and emitted thermal radiation. The distribution of radiant fluxes is, however, obviously disturbed by clouds.

It is difficult to state exactly what the influence of arctic haze is, or what it could be, on climate. There are secondary effects that must be considered: (a) The redistribution of radiant heating terms in the atmosphere would increase dynamic stability, thereby decreasing cloudiness. (b) If the haze is confined to the Arctic only, the model predicts that the energy balance contrast (which drives the general circulation) between high and low latitudes would decrease; this would probably also act to reduce cloudiness because of smaller amounts of heat and moisture entering by advection. On the other hand, the net increase in the meridional gradient of radiation balance in the Arctic itself may increase horizontal scale dynamical turbulence. In summary, cloudiness would be predicted to decrease in the Arctic during haze episodes, and circulation patterns might alter, though in ways entirely unknown at this stage of the study.

Heating of the hazy atmosphere from 1.94 kcal cm⁻² month⁻¹ of absorbed radiation (see TABLE 1) in a 2-km-thick layer would occur at a rate of 1.35°C day⁻¹. If the haze were uniformly mixed through the atmosphere, the heating rates would have to be reduced by a factor of four, but these are still substantial heating rates.

The most important effect on climate of the heating of the atmosphere probably would be to decrease cloudiness. Decreased cloudiness, lowered surface temperatures

It is not actually necessary to suppose that the haze is maintained only in the Arctic. The heating becomes smaller, or even turns to cooling, when arctic haze is placed over temperate latitudes, which in general have lower surface reflectances.

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(because of loss of infrared radiation), and dryer air masses would be expected to be caused by arctic haze. Clear skies, lower than normal surface temperatures, and lower than normal air humidities are exactly what were found at Barrow during arctic haze episodes in April 1978. The correlations suggest that arctic haze could have substantial influences on arctic climate.

The actual climatic alterations caused by arctic haze will have to be evaluated with numerical grid climate models. The parameters that would have to go into the models are, however, known only poorly. It is only recently that arctic air has been suspected of being contaminated. This first study indicates that significant climatic alterations may be brought about by the haze.

SUMMARY

Air in the northern polar regions is sometimes charged with an enigmatic scattering-absorbing haze that cuts visibility and alters the amount and distribution of radiant energy flowing through the atmosphere and the absorption at the terrestrial surface. The perturbation of the radiation energy balance caused by the haze over the reflecting pack ice has been estimated by solving the equation of transfer with a discrete ordinate method. By using what are believed to be representative values for the optical parameters of arctic haze (albedo of single scattering ~ 0.8, optical thickness in visible spectrum ~ 0.20, and an asymmetry ratio of 0.75), we calculate that in spring the haze will introduce a heating of the earth-atmosphere system of 1.3–1.9 $kcal\ cm^{-2}\ month^{-1}$, a heating of the atmosphere of 1.3–1.9 $kcal\ cm^{-2}\ month$, and a cooling at the surface of 0.3–0.4 $kcal\ cm^{-2}\ month^{-1}$. The changes in the radiation budget will tend to introduce air subsidence and decrease cloudiness. Arctic haze may therefore have a substantial influence on arctic climate.

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Abstract

During a study of cloud-condensation nuclei (CCN), condensation nuclei (CN) and various chemical constituents of the aerosol at Irafoss, Iceland, an episode of long-range transport of CCN and pollution aerosol from Eurasia was observed. This air was enriched in CCN by factors of 2-5 and in pollution aerosol by factors of 5-10 compared to near-background air from the west Atlantic and Arctic. CCN spectra of the aged European air were enhanced considerably in the largest particles, those active at the lowest supersaturations. The CCN appeared to be explained primarily by nonmarine $SO_4^=$ and some yet-unidentified background constituent of the aerosol, possibly organic matter. From these results, one may predict that anthropogenic CCN can be transported throughout the Arctic in a fashion similar to that already observed for $SO_4^=$.

Long-range atmospheric transport of cloud-active aerosol to Iceland

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Introduction

Recent studies of aerosol and precipitation in and around the Arctic have revealed trace-element concentrations considerably greater than those of typical remote regions, particularly during the winter half-year. In most of these cases, the high concentrations are attributable to long-range transport of anthropogenic aerosol from midlatitudes (Rahn et al., 1977; Gjessing, 1977; Rahn and McCaffrey, 1979a,b, 1980; Rahn, 1980; Larssen and Hanssen, 1980; Ottar, 1980). The main source regions and meteorological aspects of transport to the Arctic have been dealt with by Rahn and McCaffrey, 1979b, 1980; Rahn, 1980, 1981a,b). Eurasia seems to be the principal source region; transport to the Arctic appears to be primarily between the Icelandic low and Asiatic high, at low elevation (well below 5 km), and of duration 10-20 days.

Anthropogenic aerosol in the Arctic atmosphere may have radiative, depositional and nucleational effects. Direct radiative effects have been dealt with elsewhere (Shaw and Stamnes, 1980; Rahn and Shaw, 1981); the nature and amounts of deposition of aerosol in the Arctic are also under study (Weiss et al., 1978; Ra   and McCaffrey, 1979a; Rahn, 1980; Rahn, 1981c). One of the most efficient ways for an aerosol to affect the radiation balance of an area, hence affect its climate, is to interact with water vapor at the very low supersaturations found in clouds. These so-called "cloud-active aerosols", particles that readily interact with water to initiate the growth of droplets or ice crystals in the atmosphere, are of two types, cloud-condensation nuclei (CCN) and ice nuclei (IN). Changes in their concentrations have been related to changes in the microstructure of clouds in an air mass (Twomey, 1959; Fitzgerald and Spyers-Durbin, 1973; Hobbs et al., 1980). Such changes would be expected to change the size distributions of cloud droplets as well as

the relative numbers of ice crystals in the cloud, which in turn should change the radiative properties of the cloud. Inasmuch as one of the major components in the aradiation balance of the Arctic is the persistent stratus cloud of summer (   ), any modification of the radiative properties of Arctic clouds by anthropogenic aerosol may have palpable climatic consequences.

Very little is known about cloud-active aerosol in the Arctic. Radke et al. (1976) measured CCN and IN at Barrow, Alaska during March 1970 and found, to their surprise, that much of the time the concentrations of CCN were very high ($100-500 \ cm^{-3}$ at 1% SS), more like those of urban areas than like other remote areas they had studied. There were no obvious local sources for these CCN. Other than this report, we are not aware of any CCN or IN data for the Arctic. In an effort to remedy this, we have begun a systematic, large-scale study of CCN and IN at selected sites in and around the Arctic. Summer and winter experiments have been carried out at six sites: Barrow, Alaska; Pangnirtung, NWT; and Ny Ålesund, Spitsbergen (Arctic); Poker Flat, Alaska and Irafoss, Iceland (sub-Arctic); and Narragansett, Rhode Island (midlatitude source region). Locations of sites are shown in Figure 1. The Barrow site was chosen to represent the North American Arctic and, on occasion, aerosol entering the Arctic from over the North Pacific. A portion of this North Pacific aerosol may be ultimately traceable to sources in eastern Asia. Poker Flat, in interior Alaska, represents conditions at an earlier stage of this Pacific pathway. Spitsbergen was chosen to represent the Eurasian Arctic, as well as aerosol entering the Arctic along various pathways from Eurasia, which seems to be the major source of Arctic aerosol (Rahn and McCaffrey, 1979b, 1980; Rahn 1980, 1981a,b). The inclusion of Barrow

and Spitsbergen to represent different sides of the Arctic was seren-dipitous, for subsequent studies have revealed systematic, previously unrecognized differences in the chemical composition of aerosols from these two areas, which seem to imply different mixes of sources for each (Rahn, 1981b). The data of this study should show whether there are corresponding differences in the cloud-active aerosol as well. Pangnirtung was chosen to represent the eastern Canadian Arctic, as well as aerosol along the proposed transport path from eastern North America via the Davis Strait. The Iceland site was chosen to represent aerosol along North Atlantic pathways from North America and from Europe. The Rhode Island site represented conditions in a typical midlatitude source area, albeit one which is now not considered of major importance.

One of the important features of this study was the concomitant measurement of trace constituents of the aerosol, such as $SO_4^=$, V, Mn, Al, etc., whose long-range transport in and around the Arctic has now been studied considerably. By relating the behavior of the cloud-active aerosol to these other constituents, its origin, transport, and perhaps also chemical identity should be revealed with additional clarity.

In summary, then, the sites of this study were chosen and the measurements were carried out in a way designed to determine the importance of long-range transport from midlatitudes to the budget of cloud-active aerosol in the Arctic.

In this article we report results from the summer study in Iceland, which illustrate the approach taken at the other sites, and serve as a case study of long-range transport. Results from other sites and other seasons will be reported as they become available.

Sampling and analysis

The sampling site was Irafoss hydroelectric station, about 30 km east of Reykjavik, Iceland. Local sources of aerosol were minimal, and were limited to sparse traffic and small deserts 10 km to the north. Iceland as a whole has nearly no heavy industry to produce aerosol; the only significant fossil-fuel combustion is by automobiles, because virtually all domestic and industrial energy comes from hydroelectric and geothermal sources. Thus, Iceland is an excellent location from which to study long-range transport of aerosol over the North Atlantic.

CCN were activated with a Mee Cloud-Condensation Nucleus Counter, Model 130, and were photographed with a He-Ne laser and 35-mm camera. CCN concentrations were determined visually from the negatives. Spectra for supersaturations of 0.05, 0.10, 0.20, 0.50, 1.0 and 2.0 percent were determined each 12 hours. The actual supersaturation in the thermal diffusion chamber at very low values is quite uncertain—Mee claims an accuracy of only 0.02%—due to 0.1°C uncertainties in the temperature difference between the plates. Thus, uncertainties at the low-supersaturation portion of the CCN spectra are considerable. IN concentrations were determined from particles collected on 0.2-μm Nuclepore filters, using a modified static diffusion chamber and a technique similar to that described by Stevenson (1968). In addition, condensation nuclei (CN) were measured with a Gardner Small Particle Counter several times per day.

Aerosol for chemical analysis was collected on continuous 24-h high-volume filters. A series of elements (Na, Mg, Cl, Br, I, Al, Ti, Mn, V, etc.) was determined by nondestructive neutron activation; sulfate was determined by a modified version of the turbidimetric technique of Tabatabai (1974). In addition, 0.2-μm Nuclepore filters were taken to

determine particle-size distributions by scanning electron microscopy and elemental composition of single particles by energy-dispersive X-ray spectrophotometry.

In this article, CCN, CN and trace-element results will be discussed. Results of the other analyses will be reported later.

Results

Figure 2 gives 24-h means of some of the data obtained for 17-26 August 1979 at Irafoss. CCN concentrations are given for the intermediate supersaturation of 0.2%; counts at other supersaturations were generally parallel. Three indicators of natural aerosol are given -- marine Na for sea salt, Al for soil dust, and I for secondary marine aerosol. Marine, or noncrustal, Na was derived from total Na and Al by the following equation

$$Na_{noncrustal} = Na_{total} - \left(\frac{Na}{Al}\right)_{crust} Al_{total} \tag{1}$$

where Al and noncrustal Na are presumed to be derived exclusively from the crust and the sea, respectively. The Na/Al ratio of bulk Icelandic rock, 0.2, was used (C.K. Unni, personal communication). [Selection of the proper Na/Al ratio for use in Equation (1) was critical for samples 4-6, which had low Na and high Al. A value of 0.25, for example, was too high, because it gave unrealistically low concentrations of marine Na. Thus, the Na/Al ratio of mean crustal rock, 0.35 (Mason, 1966) could not be used here.] The sea salt obviously came from the North Atlantic, but without size-distribution data for the salt particles it is impossible to say from how far away they originated. The soil dust may have had a local or a distant origin; V/Al ratios which supported a local origin are presented below. The significance of the I data is also discussed below.

Two indicators of pollution-derived aerosol, nonmarine $SO_4^=$ and noncrustal V, are also shown in Figure 2. Nonmarine $SO_4^=$, which in many areas is pollution-derived, was calculated from total $SO_4^=$ by the following equation:

$$SO_4^=\big|_{nonmarine} = SO_4^=\big|_{total} - \left(\frac{SO_4^=}{Na}\right)_{sea\ salt} (Na)_{noncrustal} \tag{2}$$

The $SO_4^=$/Na ratio of sea-salt aerosol used here was 0.25, the value for bulk seawater (Pytkowicz and Kester, 1971). Laboratory generation of sea-salt aerosol from seawater (Gravenhorst, 1978) has yielded values between 0.22 and 0.28 for this ratio, confirming that there is no fractionation of $SO_4^=$ relative to Na at the source. Nonmarine $SO_4^=$ was 37-98% of the total $SO_4^=$.

Noncrustal V in atmospheric aerosol has a single major (pollution) source, combustion of residual oil (Zoller et al., 1978), and has been used extensively as an indicator of transport of aerosol from midlatitudes to the Arctic (Rahn and McCaffrey, 1979b, 1980; Rahn, 1980). The crustal component of V is subtracted from the total V in a parallel fashion to noncrustal Na:

$$V_{noncrustal} = V_{total} - \left(\frac{V}{Al}\right)_{crust} Al_{total} \tag{3}$$

V/Al ratios for global average crustal rock are normally used for this type of subtraction, but in these samples the V/Al ratio never fell below about double the bulk crustal ratio, even when the concentrations of V and Al were high enough that we were sure they were both derived from local soils (see Table 1). This is one of the first problems we

was at near-background levels of 0.04 to 0.3 ug m^{-3}, and V was below its detection limit of 0.03 to 0.13 ug m^{-3}. CCN concentrations were low and steady at 70-90 cm^{-3}; these values are similar to those found in other remote regions and considered to be background (Radke et al., 1976; Hoppel, 1979). Interestingly, they are also roughly the concentrations of cloud droplets in Arctic summer stratus ( ).

CN had their highest concentrations of the experiment during this first period. This was probably an effect of Reykjavik, which was generally upwind of the sampling site during this period. Thus the CN count seems to be a very sensitive indicator of local sources but an insensitive indicator of long-range transport. A similar effect has been reported recently for Barrow, Alaska (Peterson et al., 1980), where transport from the north could be seen clearly in light scattering (a measure of the submicron aerosol) but not in CN count. This behavior of CN is probably a result of their short atmospheric lifetimes, caused by rapid coagulation of the Aitken nuclei which make up the vast majority of CN numbers.

The second period was less well defined than the first. It included roughly the fourth through the sixth days, when the winds were from the north. This period was more continental, with the Na/Al ratio less than unity. Concentrations of aerosol were generally low, and began to rise at the end. CN and CCN had their absolute minima on the first day of this period; SO$_4^=$ had a relative minimum on the second day. V continued below its detection limit throughout this period.

The third period comprised the last three days of sampling, when winds were SE-SW. This period can be classified mixed marine-continental, because Na and Al had relatively high concentrations of variable dominance. The high Al of the last day was probably a local effect,

been compared with global crustal ratios applied to remote-area aerosol. The consistency of the V/Al ratio in samples 1-6 implied that this. The consistency of the V/Al ratio in these early samples and that the V/Al ratio of the Icelandic soils was more than double that of global crust. This was perhaps reasonable, because Icelandic rock is basaltic and V is generally enriched in basalts by a factor of two compared to mean crustal rock (Gmelin, 1968). For the V/Al ratio in Equation (3), we used the mean from samples 1-6, (3.64±0.21) x 10^{-3}; this ratio is close to that of bulk Icelandic rock, 4.25 x 10^{-3} (C.K. Unni, personal communication).

Noncrustal Mn (hereafter referred to simply as Mn) is often a good indicator of pollution aerosol; its values were calculated as well (see Table 2), but it was not plotted in Figure 2 because only sample 7 had a detectable value. The local Mn/Al ratio also appeared to be significantly different from the global value; a value of (22.8±2.5) x 10^{-3} was derived from samples 1-6 and 8-9 and used in the subtraction. This value compared well with the 19.6 x 10^{-3} for Icelandic rock; it was roughly double the global crustal ratio.

Three representative CCN spectra are presented in Figure 3, each the mean of three determinations. Error bars represent experimental spread of the data.

... f data shown in Figure 2, the Iceland experiment can be a... e periods. The first, which comprised roughly the first thr.. .. of sampling, had 850-mb winds from the S-W and was relatively mar.. character, as evidenced by a Na/Al ratio greater than unity. During this period, nonmarine SO$_4^=$ and noncrustal V (hereafter referred to simply as SO$_4^=$ and V) were very low in concentration: SO$_4^=$

from weekend traffic on dirt roads near the sampling site. Concentrations of CCN, $SO_4^=$ and V had their maximum during this period, and CN concentrations had a relative maximum. CCN, $SO_4^=$ and V had a temporary minimum on the second day. For V the decrease from the first day was a factor of 4, for $SO_4^=$ it was a factor of 2, and for CCN it was only 10%. The absolute maximum of $SO_4^=$ and V on the first day of this period, 1.34 µg m^{-3} and 1.21 ng m^{-3}, respectively, are high enough to strongly suggest pollution sources. They are also quite compatible with atmospheric transport on the scale of several thousand km (Rahn and McCaffrey, 1979b). On this day, haze was also observed at the site.

Trajectory analysis supported the division into three periods, and offered information about potential source areas. During the first period, air at 850 mb (1.5 km) came to Iceland from the southwest, under the combined influence of a trough over southern Greenland and the Bermuda high, which extended quite far north at that time. By the second day of sampling, a trough was beginning to form over Iceland; it deepened and progressed eastward, so that by the fourth day of sampling the air-flow to Iceland came from the north. The trough remained stationary and deepened into a closed low during the fourth and fifth days of sampling, during which time it advected air around its backside from Europe towards Iceland in a manner very similar to that proposed for flow into the Arctic around the more northerly placed Icelandic low of winter (Rahn, 1980). By the last three days of sampling, the low had dissipated and high pressure, low-wind conditions dominated. Trajectories for each of the nine days are shown in Figure 4. They were calculated in 12-h time steps at 850 mb, using observed wind speeds and analyzed contours for wind direction. Open circles are given each 24 h.

Thus, air of the first and third periods was from midlatitudes, whereas that of the second period was derived from the Arctic. Because trajectories of the second period passed over most of Iceland, we refer to this air as Arctic/Icelandic in Figure 2. Its basic cleanliness is shown by the low CN and CCN concentrations, particularly on the first day of the period. The low Na concentrations, consistently the lowest of the experiment, were probably due to a combination of relatively short path lengths over open ocean before reaching Iceland (only 300-500 km from Greenland) and dilution/removal of marine aerosol at the north coast of Iceland. The moderate-to-high concentrations of Al during this period were probably Icelandic influences, from small, stony deserts 50-60 km N-NW of the sampling site (dust clouds were occasionally observed in the direction of these deserts during the sampling).

The contrast between air of the first and third periods was striking. Even though each was a product of midlatitudes, air from the western Atlantic was very clean, and probably of near-background character, whereas that from the eastern Atlantic showed strong influences of distant pollution sources. The near-background character of the western Atlantic air could have been caused either by marine origin in the subtropical Bermuda high, or by rapid cleansing and dilution of more-polluted North American air; at present we do not know which is the correct explanation. Transport from North America would have taken 5-7 days at 850 mb to cover the 5000-6000 km. While such distances for efficient transport of aerosol in midlatitudes are not impossible, even in summer, they may be infrequent over oceans (Rahn, 1980). In this case, the V and Mn generated from Icelandic soils effectively masked any small residues of trans-Atlantic transport from North America; the concentrations of $SO_4^=$ were

low enough so that they alone could not be used to discriminate between tropospheric and North American pollution sources. The Icelandic crustal aerosol could not mask the much stronger transport from Eurasia, however.

There were far fewer ambiguities concerning the origin of the aerosol of the third period. The trajectories clearly pointed to a Eurasian origin for this pollution event, although at these distances (3000-4000 km) horizontal resolution could not be better than a few hundred km. Confirmation of a Eurasian origin was provided by the noncrustal Mn/noncrustal V ratio of sample 7, the day of haze over Iceland, which was the only day on which both noncrustal Mn and noncrustal V could be detected above the high local background. Rahn (1981b) has shown that the Mn/V ratios of eastern North American and Eurasian aerosols are systematically different, with Eurasian values 7 ± 4 times higher than those of eastern North America. When proper allowances are made for seasonal variations in this ratio (factors of 2-4 in both North America and Eurasia) and its systematic changes during long-range transport (decreases by factors of 2-3 over the first few thousand km), this ratio can be a powerful indicator of the origin of an aerosol or an air mass. There is now both theoretical and observational evidence that the Mn/V ratio of North American aerosol is 0.2-0.3 after transport to the vicinity of Iceland during summer, whereas Eurasian aerosol near Iceland in August should have and actually has a Mn/V ratio an order of magnitude higher, approximately 2-3 (Rahn, 1981b; Rahn et al., 1981). (The great difference in these ratios is because the greater aging of the North American aerosol, already several times richer in V than is the Eurasian aerosol, enriches the V still further.) By comparison, the Mn/V ratio of sample 7 of this study was 4.2 $\pm$1.9 (Table 1); this value is completely compatible with the Eurasian ratio cited above but not with the North American ratio.

These high Mn/V ratios are almost certainly a seasonal effect related to high ratios at the source in summer. Rahn (1981b) has noted that the Mn/V ratios of New York City and central Europe are 3-4 times greater in summer than in winter, which is consistent with the ratios in Iceland during August being 3-6 times higher than they are after transport to the Norwegian Arctic in winter. The ratios in Iceland are also more directly consistent with a Eurasian source: data from the Jungfraujoch (Dams and DeJonge, 1976) can be used to derive a Mn/V ratio of 6-7 for summer. If this aerosol is one-half as aged as is the Icelandic aerosol of sample 7 here, and if transport of a few thousand km decreases the Mn/V ratio by a factor of 2-3 (as it seems to for both Eurasian and North American aerosols; Rahn, 1981b; Rahn et al., 1980; Rahn et al., 1981), the Icelandic Mn/V ratio should be 3.5-5.0. Thus, chemistry and meteorology concur that Eurasia was the source of the aerosol of sample 7.

As noted above, the aerosol of samples 8 and 9 was probably also derived from Eurasia, but not so directly as was sample 7. This is confirmed by the $SO_4^=/V$ ratios of samples 7-9. The $SO_4^=/V$ ratio has been shown to be a very sensitive indicator of the degree of aging of a pollution-derived aerosol, because it increases by severalfold during the first few days of transport (Rahn and McCaffrey, 1980; Rahn et al., 1980). $SO_4^=/V$ ratios for this experiment are derived in Table 2. For samples 1-6 they were $>10^3$, consistent with summer Arctic values of $(1-10) \times 10^3$. This appears to confirm the conclusion that this early aerosol was not derived directly from a polluted air mass. In samples 7-9, on the other hand, $SO_4^=/V$ was much lower, roughly $(1-2) \times 10^3$. This indicates rather direct transport from Eurasia. Note that $SO_4^=/V$ of sample 7 was a factor of two lower than that of samples 8 or 9; this supports the impression

from the meteorological maps that the transport of sample 7 was more rapid and direct than was that of the other two samples.

We were surprised by the low $SO_4^=/V$ ratios after long-range atmospheric transport in summer. On the basis of winter ratios in the Norwegian Arctic ($2-3 \times 10^3$) and the more rapid oxidation of SO_2 in summer, we had expected to find ratios more like the $(2-10) \times 10^3$ observed in the Arctic in summer. At present, the explanation for the low Icelandic ratios is not clear. They may imply that dry deposition of SO_2 is considerably greater in summer than in winter, as would seem reasonable in view of the inertness of snow surfaces. Careful examination of data from Bear Island revealed, in agreement with the Iceland situation, that $SO_4^=/V$ of high-concentration episodes during summer is $(1-3) \times 10^3$, which is considerably lower than the $(2-10) \times 10^3$ for other times. Thus, summer transport of Eurasian aerosol over land or water seems to result in a $SO_4^=/V$ ratio of $(1-3) \times 10^3$, comparable to or slightly lower than the $(2-3) \times 10^3$ from winter transport. In the future, we will examine the chemometeorological aspects of summer transport in more detail, in the same way as has been done for winter transport by Rahn and McCaffrey (1980) and Rahn et al. (1980).

Figure 3 shows that the cloud-active character of aerosol from Europe is markedly different from that of the near-background aerosol of the earlier periods. The CCN spectrum from 23-24 August, at the height of the transport from Europe, is elevated in concentration and more nearly flat than are the two other spectra. Not only are there more CCN present than earlier, they are more active at lower supersaturations as well. These two features are indicative of continental aerosol, i.e. large numbers of large CCN, as would be expected with transport from Europe.

Taken together, then, the physical, chemical and meteorological evidence presented here reveals an Icelandic aerosol which was generally near-background in character during summer, except when the air came from Eurasia. A three-day high-concentration event of CCN and pollution aerosol was observed in connection with this Eurasian air, during which the CCN counts were elevated by 2-3 times and the pollution aerosol by up to an order of magnitude.

At this point one may inquire about which constituents of the aerosol contributed most to the CCN. In Figure 2 there is an obvious similarity between the temporal variations of CCN and $SO_4^=$, particularly during the last half of the experiment. This should come as no surprise, for it has been shown previously that $SO_4^=$ is an important component of CCN and is the most abundant trace constituent of cloud water and precipitation (Junge, 1956; Dinger et al., 1970; Twomey, 1971; Petrenchuk, 1975). $(NH_4)_2$ SO_4, NH_4HSO_4, and H_2SO_4 are all highly hygroscopic, and would be expected to readily form haze and cloud droplets at high relative humidities and low supersaturations, respectively. But the CCN and $SO_4^=$ are not perfectly parallel; something else must have contributed CCN.

We have used elementary regression analysis to determine whether any of the other major constituents of the Icelandic aerosol represented by elements we measured could have contributed significantly to the CCN; the results are shown in Tables 3 and 4. Fitting $SO_4^=$ alone plus a constant background to the CCN data explained 75% of the variance. The resulting background of CCN was 61 cm^{-3}; over the 9 samples, $SO_4^=$ and the background contributed roughly equally to the CCN. Interestingly, adding Al (for the crust), Na (for sea salt), or CN (for local pollution) to the fit changed the results only slightly; a background CCN between 57 and 61 cm^{-3} was always required, and the coefficients of

$SO_4^=$ ranged between only 0.09% and 0.121 (for the case of Na, its coefficient was negative). Thus, neither the Icelandic crust, nor sea salt, nor local Icelandic pollution seemed to contribute substantially to the CCN: relative concentrations of background, $SO_4^=$, and other constituents ranged from 51-55%, 40-52%, and -6 to 6%, respectively.

The chemical identity of this background contributor to the CCN is not yet known. It may have been NO_3^-, organics, or some secondary marine derivative. As a test of this last possibility, I, a secondary marine derivative, which was more nearly constant during this experiment than anything else that was measured, was regressed along with $SO_4^=$ against the CCN. It went much farther toward explaining the CCN (34% contribution) than did the Al, Na, or CN. But this was probably only because of its near-constancy; its actual concentrations (~1 ng m⁻³) were far too low to contribute the required numbers of CCN. The most likely source of the background CCN was organic material, which is abundant (~1 µg m⁻³ in marine aerosol) and nearly constant over various oceans (Ketseridis et al., 1976; Duce, 1978).

Other researchers have found background CCN concentrations over oceans, but without accompanying chemical measurements. This work appears to go a step farther and suggests that there are at least two primary constituents of marine background CCN, nonmarine $SO_4^=$ and something else.

Summary and Conclusions

In summary, then, we have observed a case of strong transport of CCN some 3000-4000 km from Eurasia to Iceland during summer. The CCN increased in parallel with the pollution-derived fraction of the aerosol, particularly with the $SO_4^=$, and may be identified in large measure with it. Thus, the CCN concentration in remote regions may prove to be a

very useful indicator of the presence of anthropogenic aerosol. It also can provide a direct measurement of a property that is important climatically.

These results from Iceland also allow us to speculate on CCN in the Arctic. It is now known that pollution aerosol is transported from midlatitudes to all reaches of the Arctic during winter, and possibly during summer as well (Rahn and McCaffrey, 1979b, 1980; Rahn, 1980). Our measurements in Iceland have shown that CCN are transported over comparable distances from the same source, at least in summer. CCN should thus also be dispersed over the Arctic in winter. According to Fitzgerald and Spyers-Duran (1973) and Hobbs et al. (1980), increased CCN from pollution sources shifts the size distributions of cloud droplets toward smaller sizes, with consequent changes in the radiative properties of the clouds. If our measurements of CCN in the Arctic document higher-than-expected concentrations, a similar effect on radiative properties of Arctic clouds, hence on the radiation balance of the Arctic, may be inferred.

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Figure 2. Atmospheric concentrations of Cn, CCN, and several chemical constituents of the aerosol at Irafoss, Iceland, August 1979. All data are 24-h means.

Figure 1. Sampling sites for study of cloud-active aerosol in the Arctic.
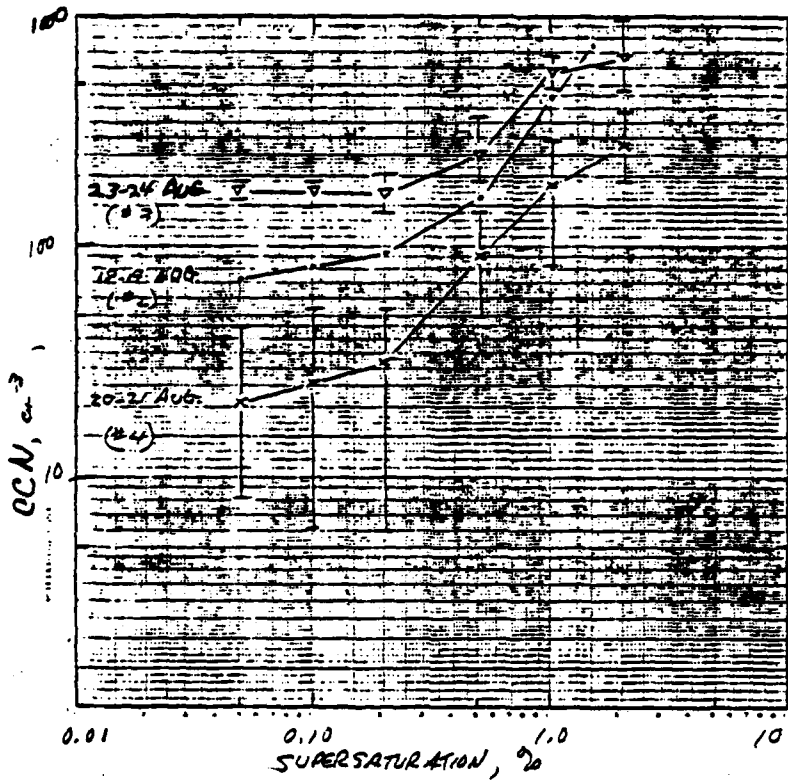
Figure 3. Three typical CCN spectra from Irafoss, Iceland, August 1979. Each spectrum is the mean of three determinations during 24 hours. Error bars are based on experimental scatter.
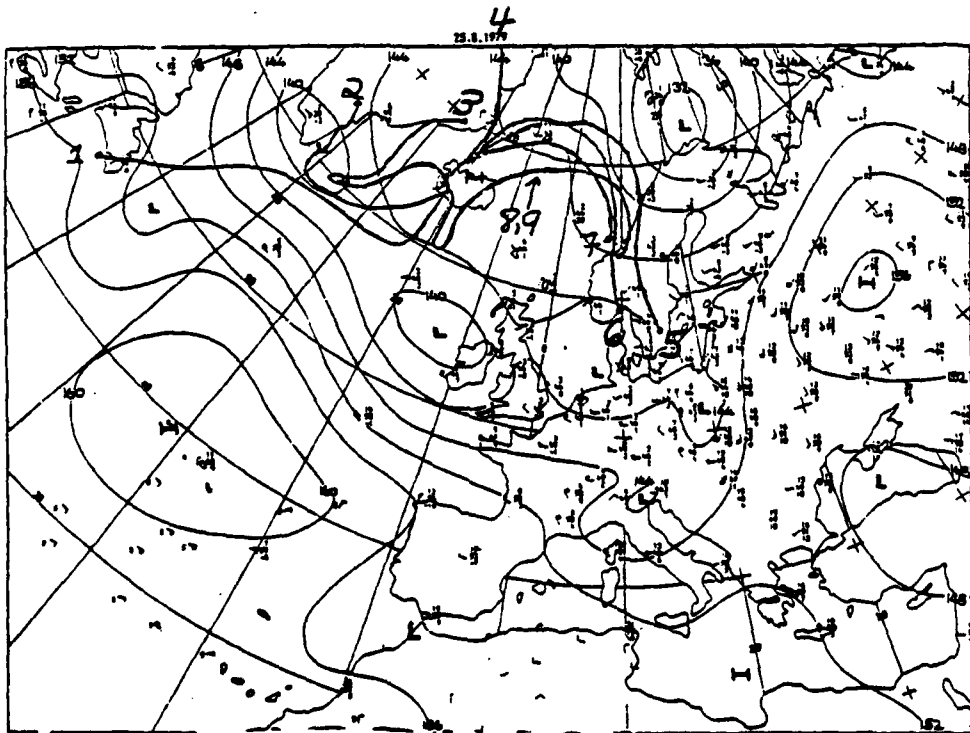
Figure 4. 850-mb air-mass trajectories to Iceland from each of the days of this study. Open circles each 24 hours. Dates are for arrival at Iceland.

Table 1. Concentrations (ng m^{-3}) and ratios of Al, Mn and V in the Icelandic aerosol of August 1979. All uncertainties 1σ.

Sample	Dates (Aug.1979)	Al	Mn	Mn/Al (x10^{-3})	V	V/Al (x10^{-3})	Noncrustal Mn	Noncrustal V	Noncrustal Mn / Noncrustal V
1	17-18	346±20	8.37±0.60	24.2±2.2	1.22±0.09	3.53±0.33	<0.9	<0.13	—
2	18-19	97±8	2.68±0.20	27.6±3.1	0.338±0.030	3.48±0.42	0.47±0.28	<0.05	—
3	19-20	38±5	0.81±0.15	21.2±4.8	0.148±0.020	3.89±0.73	<0.2	<0.03	—
4	20-21	168±10	3.96±0.30	23.6±2.3	0.642±0.060	3.82±0.42	<0.5	<0.08	—
5	21-22	155±10	3.09±0.20	19.9±1.8	0.524±0.050	3.38±0.39	<0.4	<0.07	—
6	22-23	807±50	16.7±1.0	20.7±1.8	3.02±0.20	3.74±0.34	<1.7	<0.30	—
7	23-24	752±40	22.2±1.4	29.5±2.4	3.95±0.30	5.25±0.49	5.05±1.80	1.21±0.35	4.2±1.9
8	24-25	126±9	3.02±0.20	24.0±2.3	0.744±0.060	5.90±0.64	<0.4	0.28±0.07	<1.5
9	25-26	1196±60	26.6±1.5	22.2±1.7	4.83±0.30	4.04±0.32	<2.3	0.48±0.42	<4.8
Mean of 1-6, 8-9				22.8±2.5	Mean of 1-6	3.64±0.21			
Mean Icelandic rock (C.K. Unni, Pers. Comm.)				19.6		4.25			
Mean crustal rock (Mason, 1966)				11.7		1.66			
Mean Soil (Vinogradov, 1959)				11.9		1.40			

Table 2. SO$_4^=$/V ratios in Icelandic aerosol of August 1979.

Sample	Dates, August 1979	Nonmarine SO$_4^=$, ng m^{-3}	Noncrustal V, ng m^{-3}	SO$_4^=$/V (x 10^3)
1	17-18	184±33	<0.13	>1.42
2	18-19	40±12	<0.05	>0.80
3	19-20	250±30	<0.03	>8.33
4	20-21	212±17	<0.08	>2.65
5	21-22	157±13	<0.07	>2.24
6	22-23	500±37	<0.30	>1.67
7	23-24	1343±106	1.21±0.35	1.11±0.33
8	24-25	636±51	0.28±0.07	2.27±0.60
9	25-26	999±68	0.48±0.42	2.08±1.83

Table 3. Regression analysis for CCN.

Sample	CCN (cm^{-3}) at 0.2% SS					
	Actual	$SO_4^=$ + background	$SO_4^=$ + Al + background	$SO_4^=$ + Na + background	$SO_4^=$ + CN + background	$SO_4^=$ + I + background
1	86	80	83	76	85	89
2	92	65	65	61	65	75
3	76	87	84	87	92	86
4	33	83	82	85	80	81
5	74	78	77	78	75	71
6	116	114	120	120	111	105
7	178	202	198	192	203	202
8	160	128	121	129	127	121
9	189	166	173	177	166	176
r^2	—	0.75	0.76	0.77	0.76	0.77

Table 4. Regression coefficients and relative mean contributions
of several possible contributors to the CCN over Iceland.

Component of CCN	Mean values	Regression coefficients					Relative mean contribution to CCN, %				
Nonmarine $SO_4^=$ (secondary nonmarine)	480 ng m^{-3}	0.105	0.094	0.121	0.105	0.065	45	40	52	45	28
Al(crust)	409 ng m^{-3}	—	0.017	—	—	—	—	6	—	—	—
Noncrustal Na (primary marine)	412 ng m^{-3}	—	—	-0.017	—	—	—	—	-6	—	—
CN (local contam.)	730 cm^{-3}	—	—	—	0.0054	—	—	—	—	4	—
I (secondary marine)	1.06 ng m^{-3}	—	—	—	—	36.17	—	—	—	—	34
Background	—	61.1	59.8	60.2	57.1	41.9	55	53	54	51	38
CCN	112 cm^{-3}	—	—	—	—	—	—	—	—	—	—

I.E.7.

Abstract

The ratio of noncrustal Mn/noncrustal V is 7 times greater for Eurasian aerosol than for eastern North American aerosol. When a decrease of this ratio during transport and aging is allowed for, the aerosol of the Norwegian Arctic in winter is seen to be compatible with a Eurasian source and incompatible with an eastern North American source. Winter aerosol of the North American Arctic seems to be affected by an additional source of high Mn/V ratio.

The Mn/V ratio as a tracer of large-scale sources of pollution aerosol for the Arctic.

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Introduction

Research since 1976 into the chemical composition of the Arctic aerosol has established that it contains a sizeable pollution component, particularly during the winter half-year. Primarily because of the presence of large quantities of noncrustal V (emitted by combustion of residual oil in midlatitudes), but also from the high concentrations of $SO_4^=$, Pb and other pollutants, the major sources of this Arctic aerosol are considered to be in midlatitudes (Rahn et al., 1977; Rahn and McCaffrey, 1979a,b; Rahn and McCaffrey, 1980; Rahn, 1980; various articles in this issue).

The question of which midlatitude source region, if any, is most important to the Arctic aerosol of winter is an intriguing one, and has been debated over the last 2-3 years. The major sources of air pollution in the Northern Hemisphere are eastern Asia (Japan, Korea, China, etc.), eastern North America, and Eurasia (the United Kingdom, western and eastern Europe and the western USSR). Inasmuch as air from each of these regions reaches the Arctic along reasonably well-defined pathways (via the North Pacific and the Bering Strait for eastern Asia, via the North Atlantic and the Norwegian Sea for eastern North America, and via the Norwegian Sea, Scandinavia, or the northwestern USSR for Eurasia), some aerosol will reach the Arctic from each of these sources. Because these pathways will probably have different transport efficiencies for aerosol, some source regions may be more important than others.

At present there are no established techniques of dealing with long-range transport of pollution aerosol on the scales implied here (5,000 to 15,000 km, 10 to 30 d). Consequently, in order to evaluate the various possible sources of Arctic aerosol and their pathways, we have constructed a simple model of long-range transport (Rahn and McCaffrey, 1979b, 1980;

Rahn, 1980). In it, rates of removal of aerosol are made proportional to mean precipitation, because of the very good inverse relation between aerosol and precipitation in the Arctic. Once this principle is accepted, it follows that continental pathways should be more efficient transporters of aerosol during winter than marine pathways should be, because there is much less precipitation over the interiors of continents during winter than over most oceans. Simple model calculations indicate that aerosol traveling to the Arctic via the North Atlantic will be removed with nearly an order-of-magnitude greater efficiency than will the same aerosol traveling the same distance over Eurasia. Similarly rapid removal would be expected for eastern Asian aerosol traversing the North Pacific.

This hypothesis seems to be borne out by observations. Air masses reaching Barrow, Alaska from the south during winter are much cleaner than those coming from the north (Rahn and McCaffrey, 1979b; Peterson et al., 1980); mean concentrations of aerosol pollutants in interior Alaska, 800 km south of Barrow, are 2-3 times less than at Barrow (Rahn and McCaffrey 1979b); no SO_2 and only small amounts of sulfate at the west coast of Scandinavia appear to come from North America (Rahn, 1980). Conversely, an unbroken zone of concentrated aerosol seems to extend from Eurasia northward to the Arctic (Rahn and McCaffrey, 1979b).

On the basis of this developing picture, Eurasia has been proposed as the major source of Arctic pollution aerosol during winter (Rahn and McCaffrey, 1979b, 1980; Rahn, 1980). But because the arguments behind this idea are mostly meteorological, hence indirect, or else based on seasonal-mean distributions of aerosol, they are supporting evidence rather than conclusive proof; the dominance of Eurasian sources is not yet universally accepted. The arguments against eastern Asia as a source are quite strong, however; we accept them and do not consider eastern Asia

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further here. What is then needed is a chemical tracer which can discriminate between North American and Eurasian aerosol in the Arctic. Such information would provide direct evidence about the relative importance of these two sources and give a clear direction to future studies in the Arctic.

Enrichment factors of V and Mn as tracers

At this early stage of Arctic air chemistry, the elements which are most commonly measured in aerosol samples are those coming from short neutron irradiations (Al, V, Mn, Ti, Ba, Na, Cl, Br, I), additional elements from atomic absorption and PIXE (Pb, Cd), and $SO_4^=$ by a variety of methods. Of these, the neutron-activation elements and $SO_4^=$ are the only ones common to all stations so far. Among these species, noncrustal V and noncrustal Mn are the most usable indicators of pollution aerosol, both in terms of their sources and their quality of analyses. With time, data for other indicator elements of equal reliability (such as Pb) are sure to be generated, but at present, V and Mn seem to be the best.

Of these two, V is the better indicator. V has combustion of residual oil as its only significant pollution source (Zoller et al., 1973), whereas Mn apparently has a variety of sources (Evendijk, 1974); V is more highly enriched in the atmosphere relative to the rust than is Mn (aerosol-crust enrichment factor defined for element X as $EF_X = (X/Al)_{aer}/(X/Al)_{crust}$), hence less subject to interference by crustal aerosol (Rahn, 1976). This is mostly because Mn has a severalfold higher concentration in the crust than does V, but comparable atmospheric concentrations.

Unfortunately, however, the enrichment factor of V cannot be used to distinguish North American from Eurasian aerosol, because its value in eastern North America is only 50% greater than its value in Eurasia (30 vs. 20; derived from data of Table 1). Furthermore, the V enrichment factor of an aerosol should increase greatly as it ages, because it is derived from

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large-particle Al and small particle V, as shown in Figure 1 for the Liège (Belgium) and New York City aerosols. If an aerosol ages by progressive loss of the largest particles (by sedimentation and precipitation scavenging), the V/Al ratio, hence the V enrichment factor, would be expected to increase with time. Figure 2 shows what the chemical consequences of such (simplified) aging might be. If particles are progressively removed down to radius 1 μm, the V enrichment factor would increase by factors of 2 to 5, i.e., by amounts larger than the original difference between the air masses. Variations in the aging process could thus completely obscure any remaining distinction between North American and Eurasian air masses.

A similar argument can be advanced against the use of the Mn enrichment factor to discriminate between North American and Eurasian air masses. The lower enrichments of Mn (3 for North America vs. 6 for Eurasia) make it even harder to use.

The Mn/V ratio as a tracer

Aerosols are best traced by using two elements with the same mass-size function, so that their ratio will remain nearly the same during transport. For tracking pollutants, an ideal pair would be V and Pb, which are strongly pollution-derived and are mostly submicron in urban aerosols (Rahn, 1976). When more Pb data become available for the Arctic aerosol, these elements are sure to be used extensively as tracers. But for the moment the best choice is noncrustal Mn and noncrustal V, hereafter referred to simply as Mn and V. We have previously recognized that the relative proportions of Mn and V differ to some extent between North America and Eurasia, and that their ratio in the Arctic agrees better with Eurasia than with North America (Rahn and McCaffrey, 1980). This article examines the Mn and V data more systematically, notes the

problems associated with using Mn/V ratios, and explores the limits to which one can draw conclusions from their values.

Table 1 gives concentrations of Mn, V and Al in various eastern North American, Eurasian and Arctic aerosols. Ideally, all data would have been for the winter only, the season of greatest aerosol transport to the Arctic, but some annual or nonwinter data from midlatitude source regions had to be used. Mn/V ratios from periods other than winter appear to be systematically higher than winter ratios in both Eurasia and North America. The magnitude of this effect may depend on the site, however. For example, Kretzschmar et al. (1978) found that the total Mn/total V ratio of the Belgian aerosol was the same for heating periods (November through March) and nonheating periods (June through August); Everdijk (1978) found the same result for noncrustal Mn/noncrustal V of the winter half-year vs. the summer half-year in two of three Dutch cities he studied. He did find a 50% higher ratio during the winter half-year in the third city, however. On the other hand, data of Dams and DeJonge (1976) for the Jungfraujoch, which presumably represents aged European aerosol, can be used to derive monthly mean Mn/V ratios which are 3-4 times higher in summer than in winter. Six-month means of winter vs. summer are less than a factor of two different, however. We thus conclude that use of annual or summer means from Eurasia may overestimate the true winter Mn/V ratio by up to 50 or 100%, respectively.

The only detailed seasonal data on Mn and V in North America which we could find were for total Mn and total V in New York City (Eisenbud and Kneip, 1975). Because of the relatively high enrichment factors of Mn and V in New York City (Bernstein and Rahn, 1979), the ratio of total Mn/total V is probably only about 20-30% higher than the noncrustal Mn/noncrustal V ratio, with the greater overestimate in summer. The monthly mean Mn/V

ratios for New York have an annual cycle of nearly a factor of four, with lowest values in mid-winter and highest values in mid-summer. Six-month means are only a factor of two higher in summer, however, which is similar to the Eurasian situation.

From the total Mn and V, noncrustal concentrations of each element were calculated, by the following formula:

$$X_{aer,noncrustal} = X_{aer,total} - \left(\frac{X}{Al}\right)_{crust} Al_{aer} \qquad (1)$$

where X and Al stand for concentrations of element X and Al, respectively, in the aerosol or in the crust. For crustal concentrations, data of Mason (1966) were used (Mn = 950 ppm, V = 135 ppm, Al = 81,300 ppm). These proportions of V and Al are very close to what is actually observed in crustal aerosol, but for Mn they may be 10-30% too high (Rahn, 1976). Because the proportions of Mn and Al in true crustal aerosol of midlatitudes are not yet known accurately, however, we continue to use the Mason (1966) data; the conclusions of this article are not significantly affected. Within 1-2 years the concentration of Mn in crustal aerosol should be better known.

The resulting concentrations of noncrustal Mn and V are listed in the last two columns of Table 1, and are plotted as a scatter diagram in Figure 3. The most striking feature of this plot is that the diagonal 1:1 line almost perfectly separates the Eurasian points from those of the northeast U.S., Chilton (UK) being the only exception. The Mn/V ratio is greater than unity in Eurasia and less than unity in the northeast U.S. Mean ratios for the two areas are 2.2±0.9 and 0.32±0.16, respectively, a factor of 7±4 different. The Mn/V ratio would thus seem to be a clear discriminator between these aerosols.

At present we do not know with certainty why these two aerosols should have such different Mn/V ratios. One reason may be the relatively great reliance on oil as an energy source in the United States compared to Eurasia. According to Shah et al. (1970), Mn/V ratios of 10 crude oils from California, Libya, Louisiana and Wyoming ranged from 0.003 to 0.253, i.e., lower than all the Eurasian points and most of the North American points of Figure 3. In addition, the Venezuelan residual oil burned in much of the northeast United States is unusually rich in V (Zoller et al., 1973). Because the variability of the Mn/V ratio in crude oils is almost entirely due to the variability of the V concentrations, combustion of Venezuelan oils should produce aerosol with an extremely low Mn/V ratio.

Another factor contributing to the different Mn/V ratios may be the relative lack of heavy ferrous and nonferrous industry, major sources of Mn to the atmosphere, in eastern North America compared to Eurasia. The preferential enrichment of Mn relative to V in aerosols from such sources can be seen in data from Liège, Belgium (Figure 3) and northwest Indiana (Harrison et al., 1971), both of which have large amounts of ferrous and nonferrous industry. Concentrations of noncrustal Mn in Eurasian cities are several times higher than in North American cities, whereas the concentrations of noncrustal V are about the same (Figure 3).

Arctic Mn/V ratios are greater than unity in the North American sector (Barrow, AK and Mould Bay, NWT) and less than unity in the Norwegian sector (Spitsbergen and Bear Island). In general, Arctic Mn/V ratios are higher than almost all the North American values; the ratios of the Norwegian Arctic fall roughly midway between North American and Eurasian ratios, and the North American Arctic ratios are very similar to the Eurasian ratios.

Does this mean that aerosol of the Norwegian Arctic is derived in roughly equal proportions from North America and Eurasia, and that aerosol of the North American Arctic comes mainly from Eurasia? We think that neither of these conclusions necessarily follows, because the Mn/V ratio of a pollution aerosol will probably decrease significantly during transport and aging, in the same way that the Al/V ratio decreases. V in pollution aerosol is found at systematically somewhat smaller particle sizes than is Mn (Rahn, 1976). An example of this, the aerosol of Liège, Belgium, is shown in Figure 4. Also shown here is a simplified simulation of how the Mn/V ratio of this aerosol would change during aging, assuming that aging removes the largest particles progressively. If particles were removed from the Liège aerosol from the upper end down to radius 0.3 μm (as would seem reasonable for extensive aging), the Mn/V ratio would decrease by roughly a factor of 2. Note that the decrease is much less than that that of the V/Al ratio, which from Figure 2 amounts to factors of 8 to 25 for the same change in particle size. This is of course because Mn and V have much more nearly the same particle size than do Al and V. Similar analysis of a cascade-impactor sample of New York City aerosol of August 1976 suggests that the Mn/V ratio should decrease by a little less than a factor of two during aging.

On the other hand, there is now empirical evidence for somewhat larger decreases in the Mn/V ratio of both Eurasian and North American aerosols during transport, for reasons not yet understood. The Mn/V ratio of Eurasian air seems to decrease by a factors of 3-4 during winter transport to the Norwegian Arctic (Rahn et al., 1980) and by factors of 2-4 during summer transport to Iceland (Borys and Rahn, 1981; Rahn et al., 1981). The Mn/V ratio of eastern North American air seems to decrease by factors of 3-4 during summer transport to the vicinity of Iceland (Rahn et al.,

1961). [These latter data also show that the Mn-rich aerosol of mid-western North America (Rahn, 1976) continues to be masked by aerosol from eastern North America even after transport of more than 2000 km across the North Atlantic.]

It would thus appear that the Mn/V ratio of the Arctic aerosol is inconsistent with an eastern North American origin. The aerosol of the Norwegian Arctic is probably compatible with a Eurasian source, because the Mn/V ratio of the former is about 3-4 times lower than that of the latter. Also, the Mn/V ratio at northern Norway (point 7 in Figure 3), which is geographically between Eurasia and the Norwegian Arctic, falls between the Eurasian and Norwegian Arctic ratios. A Eurasian source for the winter aerosol of the Norwegian Arctic is in accord with the considerable indirect evidence alluded to above and addressed separately in this issue (Rahn, 1981a).

But what about the aerosol of the North American Arctic? Its Mn/V ratios, after aging, are still as high as those in Eurasia. If an aging factor of 2-4 is applied to aerosol of the North American Arctic, one must conclude that it has been influenced by a previously unrecognized precursor with a very high Mn/V ratio, probably higher than almost all of the Eurasian data cited here. The identity of this precursor is not yet known. An unlikely alternative is that, for reasons unknown, the aerosol of the North American Arctic is actually less aged, hence of greater mean particle size, than that of the Norwegian Arctic. In view of the longer transport distances from suspected sources to the North American Arctic, this possibility seems doubtful, but it will be checked experimentally.

[An interesting point is that this high Mn/V ratio of the North American Arctic also shows up in snows there, as can be seen from data of

Weiss et al. (1978) cited by Rahn and McCaffrey (1979a) and Rahn (1981b): Mn/V of North Slope snows was 10. Weiss et al. used Mn as a crustal reference element; this is now seen to be untenable. Rahn and McCaffrey regarded the high Mn as a curiosity; its significance is only now becoming clear.]

In conclusion, then, the Mn/V ratio seems to be a powerful tool in discriminating between North American and Eurasian precursors of the Arctic aerosol. Present evidence suggests strongly that eastern North America is not a major source, that Eurasian aerosol affects the Norwegian Arctic strongly, and that the North American Arctic is influenced by a third source which is unusually rich in Mn.

Acknowledgements

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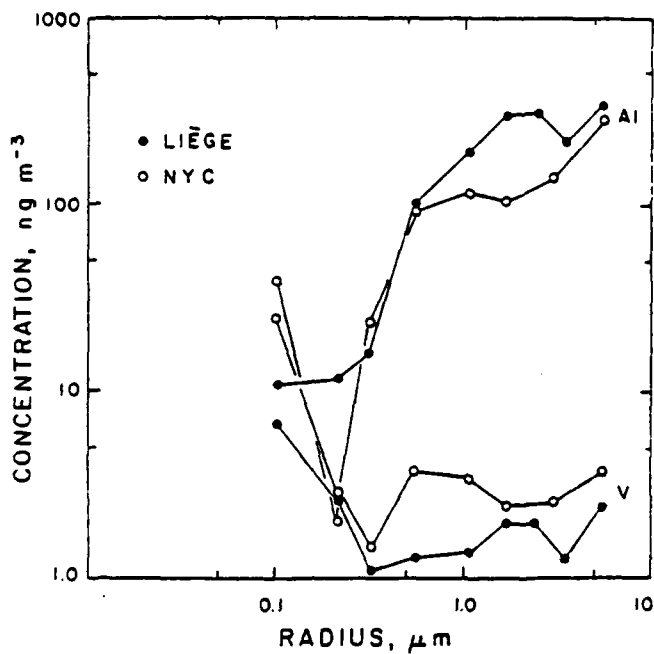
Figure 1. Mass-size functions for V and Al in the aerosols of Liège, Belgium (Rahn et al., 1972) and New York City. Both aerosols samples with low-volume Anderson cascade impactors. Liège sample from 19 April - 9 March 1972; New York sample 10-16 August 1976.
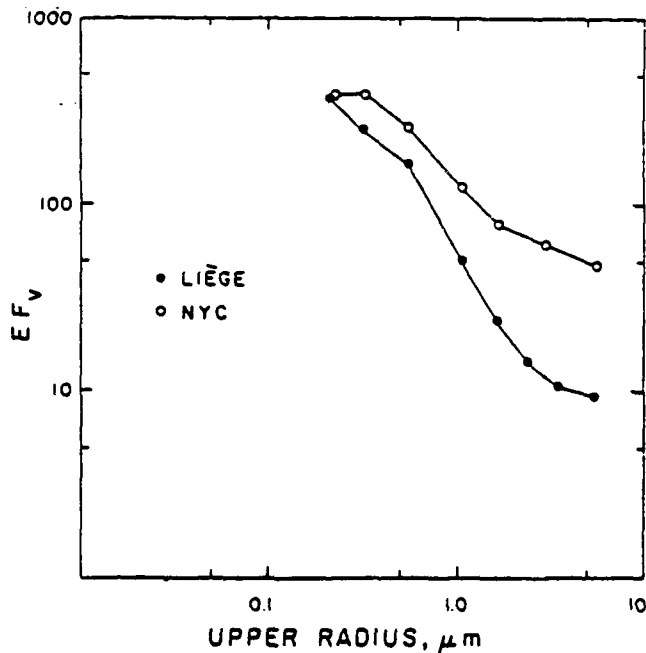
Figure 2. Simulated variation of aerosol-crust enrichment factors of vanadium during progressive removal of the largest particles from an aerosol.
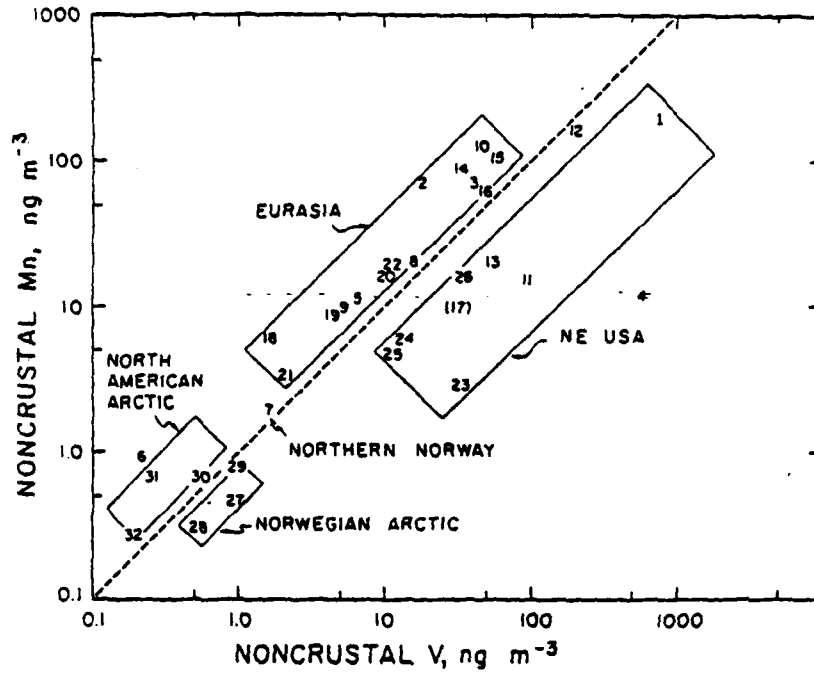
Figure 3. Scatter diagram of atmospheric concentrations of noncrustal Mn and V in Eurasia, the northeast United States, and the Arctic.
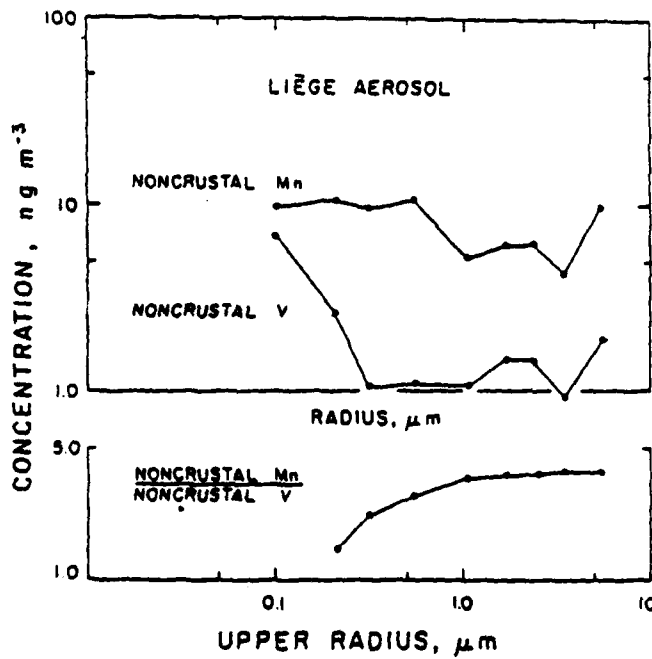
Figure 4. Mass-size functions of noncrustal Mn and V in the Liège aerosol; simulated variation of the Mn/V ratio during progressive removal of the largest particles from the Liège aerosol.

Table 1. Concentrations of Mn, V and Al in North American, Eurasian, and Arctic aerosols.

Sample Number[a]	Location	Type of sample, dates, etc.	Reference	Elemental concentrations, ng m^{-3}				
				Total Mn	Total V	Al	Noncrustal Mn	Noncrustal V
1	Buffalo, NY	Filter samples from 4 sites	Pillay and Thomas (1971)	220	800	2200	194	796
2	Liège, Belgium	Summed stages of cascade impactor, April-May 1972	Rahn et al. (1972)	92	21.3	1550	74	18.7
3	Paris	Average of 8 24-h samples at 5 locations, November & December 1970	Belot et al. (1971)	82.5	42.7	902	72	41.2
4	Boston, Mass.	Filter samples, spring 1969	Zoller & Gordon (1970)	20	600	700	12	599
5	Wraymires, UK	Average of continuous filter samples, 1972-73	Cawse (1974)	12.8	7.05	148	11.1	6.8
6	Jungfraujoch, Switzerland	Average of continuous filter samples, 10 November 1973- 10 March 1975	Dams & DeJonge (1976)	1.5	0.29	51	0.90	0.21
7	Skoganvarre, Norway	Average of continuous filter samples, November 1971 - May 1972	Rahn (1976)	2.53	1.70	43.3	2.02	1.63
8	Chilton, UK	Average of continuous filter samples, 1972-73	Cawse (1974)	24.5	17.2	325	20.7	16.7
9	Plynlimon, UK	Average of continuous filter samples, 1972-73	Cawse (1974)	11.52	5.33	150	9.77	5.1
10	Gent, Belgium	Average of several 24-h filter samples	Heindryckx & Dams (1973)	142	48	1140	129	46.1
11	Washington, DC	Average of weekly filter samples, July-November 1970	Henry & Blosser (1971)	50	100	3000	15	95
12	Philadelphia, PA	Average of 2-3 weeks' filter samples, July-August 1970	Henry & Blosser (1971)	200	200	3000	165	195
13	Bayside, NY	Average of 5 24-h filter samples, September 1970	Persiani (1971)	30	56	860	20	54.6
14	Rotterdam, Holland	Average of 1 year's filter samples during 1971-72	Evendijk (1974)	106	37	1710	86	34.2
15	Schiedam, Holland	Average of 1 year's filter samples during 1971-72	Evendijk (1974)	135	61	2260	109	57
16	Maassluis, Holland	Average of 1 year's filter samples during 1971-72	Evendijk (1974)	83	48	1480	66	46
17	Sutton, UK	Filter samples during 1969	Hamilton (1974)	14.3	32.1	370	10.0	31.5
18	Rena, Norway	Approx. 200 km N of Oslo. Average of 8 filter samples, 26 March - 6 June 1973	Larssen & Rahn (1977)	8.52	1.89	189	6.3	1.58
19	Tveiten, Norway	South coast of Norway. Average of 8 filter samples, 26 March - 7 June 1973	Larssen & Rahn (1977)	11.4	4.78	190	9.2	4.46

Sample Number	Location	Type of sample, date, etc.	Reference	Elemental concentrations, ng m^{-3}				
				Total Mn	Total V	Al	Noncrustal Mn	Noncrustal V
20	Tange, Denmark	Average of 10 filter samples, 26 March - 16 May 1973	Larssen & Rahn (1977)	23.2	11.0	521	17.1	10.1
21	Collafirth, Shetland Islands UK	Average of continuous filter samples, June 1972-May 1973	Cambray et al. (1975)	3.9	2.15	53.9	3.3	2.06
22	North Sea	Average of continuous filter samples, June 1972-May 1973	Cambray et al. (1975)	21.5	11.9	159	19.6	11.6
23	Prince Georges County, Maryland	Average of filter samples, August 1973	Gladney (1974)	13.5	35	910	2.87	33.5
24	Narragansett, R.I.	Average of continuous filter samples, October-November 1977	T.J. Conway & K.A. Rahn (unpublished data)	9.28	13.18	303	5.74	12.68
25	High Point, NJ	Average of continuous filter samples, 7 February - 1 March 1977	T.J. Conway & K.A. Rahn (unpublished data)	9.98	11.81	407	5.22	11.13
26	New York City	Average of continuous filter samples, October-November 1977	T.J. Conway & K.A. Rahn (unpublished data)	21.1	36.4	401	16.41	35.7
27.	Bear Island	Average of continuous filter samples, November 1977-March 1978	K.A. Rahn and B. Ottar (unpublished data)	1.15	1.03	59.2	0.46	0.93
28	Spitsbergen	Average of continuous filter samples, November 1973-March 1974	B. Ottar (unpublished data)	0.84	0.62	45.7	0.31	0.54
29	Spitsbergen	Average of continuous filter samples, February-April 1978	K.A. Rahn & B. Ottar (Unpublished data)	1.24	0.99	39.6	0.78	0.92
30	Barrow, Alaska	Average of continuous filter samples, November 1976-April 1977, November 1977-April 1978	T.J. Conway & K.A. Rahn (unpublished data)	1.12	0.59	29.9	0.71	0.54
31	Barrow, Alaska	Average of continuous filter samples, November-December 1976 and November-December 1977	T.J. Conway & K.A. Rahn (unpublished data)	1.10	0.21	29.4	0.70	0.25
32	Mould Bay, NWT	Average of continuous filter samples, November-December 1979	Barrie et al. (1981)	--	--	--	0.28	0.19

II. Renewal Proposal: 1 October 1980 - 30 September 1983

A. Introduction and rationale

We are requesting three years' more support to continue our study of the Arctic aerosol in much the same fashion as we have for the last four years. Our major goals are (still) to better understand the sources, transport, in situ physicochemical characteristics, and environmental effects of the Arctic aerosol. As before, we plan to deal in a broad, interdisciplinary manner with the entire phenomenon of the Arctic aerosol, working on different aspects of the problem simultaneously. Primary areas of interest for 1980-83 include further delineating the spatial and temporal distributions of Arctic aerosol and Arctic haze, deducing source areas from chemical composition of the aerosol, understanding the major meteorological features of how the aerosol is transported to the Arctic, modeling the large-scale chemical and physical changes of the polluted air masses during transport through numerical simulation, correlating chemical and optical measurements, investigating historical and climatological aspects of Arctic haze, and better determining the radiative, nucleational, and depositional effects of Arctic aerosol.

Work on nearly all these topics is already underway; we anticipate steady progression and evolution during the next three years, with no major departures foreseen. This approach has worked successfully for us in the past; we see no reason to alter it now. Study of the Arctic aerosol has become a recognized sub-discipline in the last years; at the same time, it has taken on broadness and depth to the point that many more than the three years' effort proposed here will be required to answer the questions satisfactorily.

During the past two years we have been able to formulate a number of scientific questions quite precisely. Many of these, with a few answers, have been treated implicitly in Section I.C. above. The following is a partial list of questions which are currently on our minds, and which we will try to answer during the coming contract period:

- How extensive are the systematic differences in chemical composition of the aerosols of the North American and the Norwegian Arctic?

- Does the composition of the aerosol of the North American Arctic really imply the existence of a previously unrecognized pollution source? If so, what is it, where is it, and how important is it? Can its effects be seen in other parts of the Arctic as well?

- How important is eddy-diffusion transport of aerosol in and around the Arctic, relative to organized transport (trajectories)?

- What are the major forces which account for the seasonal variations of Arctic aerosol, the spring maximum in particular?

- Can these forces also explain why there is a less pronounced summer minimum in the Norwegian Arctic than in the North American Arctic?

- What are the major periodicities of aerosol in the Arctic? Are they the same or different on the two sides of the Arctic?

- How important is the USSR as a source of Arctic aerosol?

- Can the Arctic be used as the first large-scale test area for the Chemical Element Balance (CEB) method of deducing sources from composition?

- We believe that Eurasia is the major source of the Arctic aerosol. Under what, if any, circumstances can North America contribute significantly?

- Where do the high concentrations of ^{222}Rn and ^{210}Pb of Barrow in winter come from? Are their sources the same as for the bulk of the aerosol?

- We have postulated long residence times for the Arctic aeros particularly during winter. Can we determine residence time perimentally? In particular, can ^{222}Rn and ^{210}Pb be used to vide quantitative estimates of transit time and/or residence time Arctic aerosols?

- To what extent might pollution-derived cloud-condensation nuclei, which now seem to be ubiquitous in the Arctic during winter, modify the radiative properties of Arctic clouds?

- Why is mean Arctic snow so different in trace-element composition from mean Arctic aerosol? What implications does this have for interpreting historical data from the Greenland Ice Sheet?

- Might deposition of pollution-derived Arctic aerosol already be affecting the ecology of the North Slope, such as growth of lichens or efficiency of nitrogen fixation by blue-green algae?

- What is the source of the abundant Br in the Arctic atmosphere or spring? Can this Br possibly be responsible for the temporary disappearances of near-surface O_3 at Barrow during spring?

- What is the primary altitude of transport of aerosol ultimately observed at the surface in the Arctic?

- What is the particle-size distribution of the Arctic aerosol? Is it a steady-state, self-preserving distribution?

In the near future, we expect to see gradual and steady shifts in the orientation of our work: (1) Our data collection will become progressively more directed with time. While we continue to accumulate samples, analysis will be more oriented toward less-understood locations, interesting times of year, potential tracer elements, etc. In other words, we will more selectively accumulate our data to fill gaps in our knowledge. (2) We will spend more of our time interpreting our data, formulating ideas, etc., and relatively less on general collection of data. We will still generate many data, however. (3) We will write considerably more articles than in the past, now that our ideas on the Arctic are settling down. We have a backlog of articles ready to be worked on.

We close this section by noting that there are a number of related projects underway on Arctic air chemistry, both in our laboratories and in others. G. Shaw and K. Rahn have a two-year NSF grant to continue

studies of the radiative effects of Arctic aerosols, as of May 1980.
R. McCaffrey and K. Rahn have an NSF grant to study aerosol-snow fraction-
ation. A renewal proposal, to continue study of deposition in the Arctic,
is now being written. We hope to get support from NOAA/ARL soon, as well.

But the cornerstone of our support is still ONR, and has been since
the beginning of our work in 1976. Thanks to ONR's steady assistance, we
have been able to conduct our studies in the thorough, long-term way that
the topic requires. Few projects are so lucky; for this we owe a great
debt to ONR.

A large change in the related programs during the last two years is
the increase in number at other institutions. Two years ago, we were
nearly the only group working on the Arctic aerosol. Now, there are
active programs at NOAA/GMCC, Lawrence Berkeley Laboratory, New York Uni-
versity, Bell Laboratories, Georgia Tech, Atmospheric Environment Service
of Canada, Carnegie-Mellon University, University of Wyoming, Danish Air
Pollution Laboratory, and Norwegian Institute for Air Research. These
programs help our work considerably, by providing a breadth that would
not otherwise be possible.

B.   Plan of the work

1.   Routine sampling of Arctic surface aerosol (URI, UA)

We plan to continue our routine sampling of Arctic surface
aerosol for at least this contract period. Such samples have obvious val-
ue as a long-term library; during this contract we will accumulate years
5, 6 and 7 at Barrow and Fairbanks. We are the only group taking regular
aerosol samples at carefully chosen locations in Alaska.

We also feel very strongly that the air quality of a given remote loca-
tion can be properly understood only after several years' data have been
collected. From the beginning, we have rejected the tendency to limit our
sampling to brief campaigns which, while admittedly intensive, miss much
crucial information such as timing of seasonal variations, periodicities,
etc. We have had several spring campaigns, but only as supplements and
complements to routine, continuous sampling.

On the other hand, we are not a monitoring agency like NOAA/GMCC,
which thinks in terms of decades of records. We study the Arctic atmosphere
scientifically; it happens to take a time somewhere between the weeks of
campaigns and the decades of true monitoring. In principle, a few annual
cycles should be enough. "A few" is not very well defined in this kind
of study. At first we thought that two years would be enough, but our
first two winters (1976-77 and 77-78) were extremely unusual. Our third
winter approached normality, but the fourth was unusual again. Maybe
the Icelanders are right, that 5-10 years is required.

In any event, some more years of operation will be necessary, because
much of the Arctic Air-Sampling Network only began in 1979-80 (the three
Canadian sites, the five Greenland sites, improved Spitsbergen sites, our
Iceland and Ireland sites). At present, we are sampling at Barrow, Poker
Flat (Fairbanks), Narragansett (RI), Iceland and Paris, with Ireland just
about to start. In spring 1981 we will start at Nord, Greenland. Ireland
will probably run for only one year; the other sites will operated indefin-
itely. Thus we have stabilized at six sites.

2. Chemical analysis of the routine samples (URI)

As noted above in Section I.B.2, nondestructive neutron activation has been the heart of our analytical program, and will remain so for the near future. The elements determined routinely by short neutron activation give an excellent overview of the Arctic aerosol (Na and Cl for sea salt, Al and Ti for the crust, V and Mn for pollution sources, etc.). In addition, we are beginning atomic absorption of the Barrow aerosol samples, with first results expected in fall 1980. Pb and Cd are of greatest interest. The technique can be easily extended to Mn, Cu, Zn, Ag, and several other elements.

We need to analyze some of the Barrow samples by long irradiations also. During the coming contract period we hope to do at least 1 or 2 years of Barrow samples in this way.

We are steadily accumulating data for ^{210}Pb from our Barrow filters. This is potentially important data, because EML stopped determining ^{210}Pb at Barrow after one year of data from 1975-76. We are thus the only source of ^{210}Pb data for Alaska. We plan to complete our counts of the first two years of filters from Barrow, then move to filters from Spitsbergen and Bear Island. This work proceeds slowly, though, because a count takes typically three days, and because we only have access to the LEPS detector half-time.

We continue to have the capacity to determine sulfate from samples of Arctic aerosol. Three techniques are now available: the ^{131}Ba technique originally used, a more sensitive colorimetric technique, and ion chromatography. We will probably only determine sulfate as the need arises, not routinely.

3. Daily aerosol samples during winter (URI)

As an experiment, we took daily aerosol samples at Barrow, in addition to the routine 4-day samples, during winter and spring of 1979-80. The major goal of this project was to determine whether there were regular periodicities in the Barrow aerosol of shorter time scale than the quasi-monthly cycle which shows up clearly in our 4-day samples. A quasi-weekly periodicity is seen at Bear Island, and seems to be related to the passage of synoptic systems through the area. Comparison of the relative intensities of the various periodicities on both sides of the Arctic may reveal much about the modes of transport of aerosol, in particular the relative contributions of standing eddies (long waves) and transient eddies (synoptic systems). Ultimately, this should lead to a much greater predictability for aerosol and haze in various parts of the Arctic.

We will take one more winter of daily samples at Barrow during 1980-81. Because of the large number of samples involved, the project will probably stop here.

4. Optical measurements and radiative effects of the Arctic aerosol (UA, URI)

Radiative studies in the Arctic are presently sponsored both by this ONR Contract and by the Shaw-Rahn NSF grant "Radiative effects of Arctic haze". As we see it, our radiative studies fall into three broad categories: "initial" direct measurements of optical depths, skylight, etc.;

"intermediate" inference of other important optical properties such as particle-size distribution, scattering phase function, and albedo of single scattering from the primary data, usually by inversion; and "final" calculation of radiative effects on the Arctic from the primary and inferred optical properties. We consider that the primary optical measurements, which can be correlated with our chemical measurements, are supported primarily by ONR, that the inference of size distribution, etc., is supported equally by ONR and NSF, and that the deduction of radiative effects in the Arctic is supported mainly by NSF.

We are presently taking routine measurements of atmospheric turbidity at Fairbanks. These measurements have been of great value to us for several reasons. They have shown clearly that Fairbanks is less hazy than Barrow in late winter and spring (there is also less pollution aerosol at Fairbanks than at Barrow), that pulses of haze do reach Fairbanks in spring (February-April), and that this haze is associated with cold air, $SO_4^=$ and V, i.e., has come from the Arctic. Because this haze is basically transported to interior Alaska from another region, its appearance depends strongly on the local large-scale circulation, hence varies greatly from year to year. During the winter of 1979-80, for example, which was the third warmest in recorded history at Fairbanks, there were no haze events at all, according to G. Shaw. We plan to continue these measurements in Fairbanks indefinitely. Understanding the Fairbanks situation helps us delimit the boundaries of Arctic haze, as well as the circumstances under which it is transported out of the Arctic.

Thanks to our interest in haze at Barrow, the NOAA/GMCC program has restarted its routine measurements of atmospheric turbidity there, and supplies us the data upon request. Their instruments are not state-of-the-art, however. We will supply them with a more sensitive, better-calibrated sun photometer as part of the Shaw-Rahn NSF radiative effects grant. This grant will also make available 9 other precision photometers which can be dispersed around the Arctic Air-Sampling Network. Within 2-3 years, we should have a much better picture of the horizontal distribution of Arctic haze.

In addition to the routine optical measurements, we will collect data from a variety of Arctic locations on field campaigns (see next section). These trips are major sources of optical data for us.

During the next three years we will continue to infer properties of the Arctic aerosol such as size distribution, phase function, and single-scattering albedo, from optical (or chemical) measurements. Whenever possible, results from two or more methods will be compared. For example, the single-scattering albedo can be derived from diffuse and direct optical measurement, from reflectivity of filters compared with scattering in nephelometers, and from chemical measurements of elemental carbon, vanadium, sulfate, sea salt, and soil. Particle-size distributions can be obtained from inversion of optical measurements, direct counting of particles on Nuclepore filters, inversion of data from Nuclepore diffusion chambers, or chemical measurements on stages of cascade impactors. Combining the results of different techniques should provide much more reliable estimates of the property in question.

Once these inferred optical properties of the Arctic aerosol are known to our satisfaction, they will be used in the NSF project to estimate direct radiative effects of Arctic haze.

In connection with his optical studies of Arctic haze, Dr. Shaw will be attending the First International Workshop on Light Absorption by Aerosol Particles at Colorado State University, 28 July - 8 August 1980. At this workshop, various groups will be measuring various properties of atmospheric radiation, inverting these data to infer properties of the aerosol, then checking the results against the same properties measured directly. This intercomparison should provide a most useful test of the techniques used in our studies of Arctic radiation.

5. Field campaigns (URI, UA)

In the past, we have had one major field campaign per year, usually during spring, in which a variety of optical and chemical measurements was made simultaneously. (The July-September 1980 YMER expedition is our first summer campaign). We intend to continue this practice in modified form, throughout the next contract period.

Our winter-spring campaign for 1980-81 will be a single-engine aircraft experiment. An opportunity that we cannot refuse has been presented to us by Mr. Donald W. Heckert of Fairbanks, an extremely experienced pilot who has recently developed a strong interest in the Arctic aerosol. For a total of $12,000 he will construct an air-sampling system to our specifications, install it in a local single-engine aircraft, modify the aircraft for longer range, and conduct six 10-hour missions between Fairbanks and the ice pack north of Barrow. The flights will take place at monthly intervals from November 1980 through April 1981, at times to be decided by Mr. Heckert according to a set of requirements worked out with K. Rahn and G. Shaw. The experiment will consist of hourly 4-inch high-colume filter samples, shorter-period micro-impactor samples, and sun-photometer measurements as light permits. The filters will be analyzed chemically by neutron activation, atomic absorption, and other techniques as needed. The impactor samples will be examined by scanning electron microscopy for morphology, particle-size distribution, and some indication of elemental composition. Mr. Heckert will be accompanied by Mr. B. Miller, also of Fairbanks. Both Mr. Heckert and Mr. Miller have a great deal of experience in aircraft sampling. It should be noted that Mr. Heckert will donate his time to this project.

A mission will consist of $3\frac{1}{2}$ hours flying from Fairbanks to Prudhoe Bay or Barrow, 3 hours over the ice, then $3\frac{1}{2}$ hours back to Fairbanks. The samples en route will provide a N-S transect over the Brooks Range, which seems to be the mean dividing line between haze and nonhaze during winter. Once over the ice, a three-level vertical profile will be taken. If the destination is Barrow, the current ground-level sample at the GMCC observatory will serve as the fourth level.

Both the transects and vertical profiles will provide valuable information about the Arctic aerosol. The transects should give an idea of just how large the instantaneous contrast in aerosol aloft between the North Slope and interior Alaska can be. Until now, we have had data on only longer-term contrast, at ground level. They should also elucidate the role of the Brooks Range as a climatic divider, hence a divider of aerosol. These atmospheric transects can be compared with transects of

deposition in snow and in lichens. We intend to generate the former under
the NSF aerosol-snow continuation grant with R. McCaffrey; we will aim
for the latter through some sort of cooperative project which we will try
to establish with Wayne C. Hanson and Marianne Krasny (see Section I.C.7
above). This cooperative project will compare two measures of atmospheric
contrast (surface and aloft) with two measures of depositional contrast
(snow and lichens), and should considerably enlighten our understanding
of the contrast between the aerosol and its deposition in and around the
Arctic.

The vertical profiles of aerosol in the Arctic are badly needed, as
well. In the most general terms, they will indicate how well our measure-
ments of the surface aerosol represent the totality of the Arctic aerosol.
Until now, we have had vertical profiles of only turbidity, which is a
function of both aerosol and relative humidity. These flights will pro-
vide information on the aerosol alone, which can then be used to infer the
effect of relative humidity on the intensity of Arctic haze. Among other
things, this should help decide whether the surface clear layer in the
Arctic is caused by a deficit of aerosol or by low relative humidities,
or by both. This question has plagued us for the last two years. Verti-
cal profiles may also shed some light on mixing from aloft into the surface
layer, and on removal rates of aerosol from the surface layer, both of
which are needed to properly understand long-range transport of aerosol
in the Arctic. If the chemical composition of aerosol above the boundary
layer is markedly different from that within it, different sources for the
two aerosols and little local mixing would be implied. It would also im-
ply that the aerosol measured at the surface has been transported long dis-
tances within the surface layer. Small differences between the surface
layer and aloft would imply some combination of slow removal and fast
mixing.

All in all, then, we consider this aircraft experiment to be a poten-
tially valuable one.

Our other field work for 1980-81 will be to establish a high-volume
sampling site at Nord, Greenland in spring 1981. We do not foresee any
unusual problems with this project; it should take only 2 days to install.
It is possible that the person traveling to Nord will stay longer and make
a mini-campaign out of it, including several different types of measurements.

Field work for the second and third years of this contract is harder
to anticipate. There will be an optically oriented field program each
year under the Shaw-Rahn NSF grant; we may wish to link subsequent ONR
field studies with these NSF trips. Based on the results of the 1980-81
aircraft experiment, we may decide that a more detailed program should be
conducted, for example with the NCAR Electra. This was mentioned in the
last proposal, but later proved to be premature. We will try for some
degree of field work each year, though, and will be flexible enough to
adapt to whatever opportunity should arise, such as the YMER study in 1980.

6.   Particle dynamics of the Arctic aerosol (UA)

An intriguing aspect of the Arctic aerosol which we would
like to pursue during the next three years is its particle dynamics.

Questions of interest include:

· What is the rate of gas-to-particle conversion in the Arctic as a
function of time of year, and how does it depend on concentration
of precursor gases, sulight, etc.?

· Is the spring maximum of secondary substances like $SO_4^=$ caused by
rapid conversion of precursor gases already in the Arctic by UV
light as the sun rises, or by transport after oxidation farther
south?

· How great is the size-dependent flux of material through the aero-
sol particle spectrum?

· Is the Arctic particle-size distribution at equilibrium, i.e. self-
preserving?

· What are the relative efficiencies of in-cloud processes, below-
cloud processes, ice-crystal scavenging, sedimentation, surface
impaction, chemical reactions, etc., in removing particles from
the Arctic atmosphere?

Answers to these and similar questions may help us to estimate the
rate of deposition of Arctic aerosol from the atmosphere, its residence
time in the Arctic, the ratio of secondary and primary aerosol, as well
as the underlying reasons for the observed particle-size distribution
of the Arctic. These answers should lead us to a deeper understanding
of the causes of Arctic haze.

7. Cloud-active aerosol in the Arctic (Borys Ph.D. thesis, CSU)

During the coming contract period, Mr. Borys will finish
his Ph.D. thesis on cloud-active aerosol. By September 1980 all his field
work will be finished; work for the contract period will consist of finish-
ing analysis of the samples, reducing and compiling the data, interpreting
the data, and writing it up, first as a thesis and then in a series of
articles.

In addition to the thesis, Mr. Borys has additional courses and depart-
mental examinations to complete, as well as a thesis defense. Ideally,
this would all be finished in the first two years of the contract, but
this timetable is not certain.

8. Climatology of Arctic haze (Raatz Ph.D. thesis, UA)

The Ph.D. thesis of Mr. W.E. Raatz of the Geophysical
Institute, University of Alaska, will continue throughout the coming con-
tract period. This work will have two main components, historical (ana-
lysis of past Ptarmigan records of haze) and current (analysis of data
from Arctic Air-Sampling Network; correlation of these data with synoptic
conditions in the Arctic). Analysis of the Ptarmigan data will consist
of intensive examination of the 114 flights during which haze was reported,
to determine the meteorological conditions associated with the haze, and
hopefully also to determine something about the horizontal and vertical
distributions of the haze. Unless something unexpected turns up, however,
the Ptarmigan data will be less helpful than we had anticipated.

The course of the current climatological study of Arctic haze is not easy to predict. Neither the methods nor the goals are precisely defined at this point. The general goal is to understand the major forces that control Arctic haze by using climatological/statistical methods. The seasonal trends of the different elements will be compared with such meteorological variables as temperature, precipitation, cloudiness, placement of synoptic systems, etc., and relations between them will be sought by a variety of statistical techniques. The same can be done for the quasi-monthly cycles at Barrow. Data from the Norwegian Arctic may be used if time permits. An intensive meteorological analysis of a single pollution episode at Barrow will be attempted.

It is not yet known which of these approaches will work. We hope, though, that the sum total of Mr. Raatz's efforts will yield a climatology of Arctic haze which can help us better understand its occurrence, and lead to a reasonable predictability.

9. Interviews with former Ptarmigan observers (URI, UA)

Two years ago we proposed to interview two retired Ptarmigan observers now living in California in order to try to glean whatever further information we could about the occurrence of Arctic haze. Unfortunately, none of KAR's trips during the last two years came close enough to California to make these side trips worthwhile. We will hold over the modest funding for this effort and hope to use it during this contract period. Perhaps it is just as well that these interviews were postponed, because in the meantime we have begun to work with the raw Ptarmigan data ourselves, and can ask much more specific questions now than we could have earlier. W.E. Raatz may also participate in these interviews.

10. Arctic data bank (URI, UA)

Beginning with this contract period, URI and UA will be organizing their chemical and optical data on the Alaskan aerosol more formally so that it will be readily available to interested users outside our project. The exact mechanism for doing this has not yet been determined, but it will almost certainly involve generating a hard copy for each request, with the data stored in a computer. We do not have enough data to warrant distributing it on magnetic tape.

During the meeting of the Arctic Air-Sampling Network on 8 May 1980, organization and dissemination of data within the Network itself was discussed. There was a general consensus that there was still insufficient data to warrant having it stored in some central facility, such as NILU. It seems that, at least for the next 2-3 years, each laboratory will take care of its own data. The Arctic Newsletter may advise the Network about times, places, and elements for which each laboratory has data, however.

11. Meteorological analysis (URI, UA, CSU)

We will continue to improve our meteorological understanding of transport of pollutants to the Arctic as best we can during the coming contract period, both by our own efforts and through cooperative projects with other laboratories. In the past, we have wondered just how much time we should expend on meteorological analysis. Because neither URI nor UA is a center of dynamic meteorology, we are restricted to using

rather rudimentary methods. Experience during the last 3-4 years has shown, however, contrary to general expectation, that very much can be done with these elementary methods. This is due in part to the early stages of study of Arctic meteorology and in part to the sparsity of meteorological data from the Arctic. In the paragraphs below, we discuss the various ways in which we will proceed.

One of the major ways that we infer the nature of circulation in the Arctic atmosphere is through chemistry. The chemical composition of the aerosol at a given location and time can be used to deduce its general source area, which in turn places major constraints on the path of the air mass. The most recent example of this is the Mn/V ratio, which seems to show that much or most of the air in the North American Arctic has come from sources eastward of Europe (see Sections I.C.1, I.C.3, and I.E.7 above). This is but one example--more will surely appear as data on the Arctic aerosol increase. The power of such pure "chemometeorology" is largely unrecognized by the atmospheric community today; we intend to press forward with its development, application, and promulgation.

Once the sources are known, the transit time can be deduced from trajectories or possibly through radionuclides (both discussed in more detail below). We then use numerical simulation of aging of the air mass to derive information on the various physical and chemical changes which occur during aging, such as the rates of gas-to-particle conversion, rates of dry and wet deposition, and the degree of large-scale dilution. Of course, atmospheric conditions at the source must be known in order to simulate aging. Knowledge of conditions at points between the sources and the Arctic also constrains the system, and can be used to deduce changes in rates during transport. Our first simulations appeared in Rahn and McCaffrey (1980; reproduced here as Section I.E. above. Because data from only the sources and end points were used there, conclusions were general (but valuable). We are now working on a considerably refined version of these simulations (Rahn-Brosset-Ottar) to be presented at the General Motors Research Symposium on particulate carbon in the atmosphere, in October 1980. Data from four additional degrees of aging in and around Eurasia will be included there, and used to check the original simulations.

The potential application of such simulations is nearly unlimited. We intend to use them to check the reasons for the high $SO_4^=/V$ ratio in the Arctic, the spring maximum of aerosol in the Arctic, the fall maximum of certain constituents, and the magnitudes of the annual cycle in various locations in the Arctic. In addition, they can be used to check the relative importance of competing variables, such as cloudiness and precipitation for removing aerosol.

We would also like to try to use continental radionuclides (^{222}Rn, ^{210}Pb, ^{210}Bi, ^{210}Po) to deduce information about residence times and transit times for aerosol in and around the Arctic. For some time now, R.J. McCaffrey has been studying these possibilities, with an eye toward an eventual project. The results have been generally discouraging, although a glimmer of hope has come recently. Close examination of the radionuclide literature reveals that nearly all the data have been interpreted in terms of steady-state conditions. This now appears to be a serious oversimplification under normal circumstances and a fatal one in the Arctic. Non-steady-state conditions in the Arctic can be dealt with, however, when measurements near the continental source are available, in

addition to end-point data from the Arctic. If the source of ^{222}Rn and
^{210}Pb is, as we suspect increasingly, the USSR, near-source data will
be extremely hard to obtain, either through literature or via coopera-
tive experiment. Dr. McCaffrey has been working on ways to circumvent
this problem; he has very recently found one, but it is so risky and
involves such assumptions and simplifications that it is impossible to
say without testing whether it will work. During the first year of this
contract, we hope to collect some data that can be used to very prelimi-
narily evaluate this technique; if it looks sufficiently promising, a
proposal will be submitted to NSF or some other agency.

We are very interested in the question of eddy diffusion vs. orga-
nized transport in the Arctic, as raised by G. Shaw. During the next
three years, this model will be refined, extended, and tested in as many
ways as possible. Critical tests should be available from comparison with
John Miller's trajectories, patterns of chemical composition in and around
the Arctic, periodicities of aerosol on both sides of the Arctic, and
patterns of correlation of haze with Arctic synoptic systems.

One of the most effective ways in which our meteorological analysis
proceeds is via cooperative projects. There are very few people who call
themselves Arctic meteorologists, and those who do are usually concerned
more with micro- or meso-scale phenomena than with the synoptic- or larger-
scale processes which we need to understand. Thus, the Arctic meteorology
of concern to us is all done by non-Arctic meteorologists.

At present there are three main Arctic meteorological projects of
interest to us. The first, which is not really a cooperative project,
is calculation of 5-day, 850-mb trajectories to Barrow by J. Miller of
NOAA/ARL in Silver Spring, Maryland. We have examined all these trajec-
tories for the first four years and analyzed them statistically. They
have been of great use to us, and we will continue to discuss them with
Dr. Miller.

As a result of the May 1980 symposium at URI, Dr. Toby Carlson of
Penn State University has taken an interest in large-scale Arctic meteor-
ology. During summer 1980 he will be looking into possible thermodynamic
constraints on the introduction of pollutants into air masses later ob-
served in the Arctic. This type of analysis may be very useful to us
in deducing what kind of air mass the aerosol observed at Barrow was ori-
ginally introduced into.

During the past year we have been in contact with Dr. Harry van Loon
of NCAR. Dr. van Loon is a dynamic meteorologist who has devoted a great
deal of effort to understanding large-scale features of the circulation
in the Northern Hemisphere, up to and including the Arctic. He has of-
fered to help us however he can; we will take him up on it this year.

12. Sources of atmospheric Mn (URI)

Now that the Mn/V ratio holds promise of discriminating
between broad source regions for Arctic aerosol, it becomes imperative
to better understand the sources of atmospheric Mn. The sources of
atmospheric V are quite well understood, thanks to a comprehensive survey
by Zoller et al. (1973). No such survey exists for Mn, however, to the
best of our knowledge. To remedy this, we will study the sources of Mn

during an early stage of the next contract period. This will be a more involved task than it was for V, though, for Mn has a greater variety of sources, including heavy industry. Once we develop a feeling for the principal sources of Mn to the atmosphere, we will be able to predict what kind of area should generate the high Mn/V ratios found at Barrow and Mould Bay.

13. Br in the Arctic atmosphere (URI)

Our discovery of high concentrations of Br in the Arctic aerosol, with the puzzles it has created, is described in Section I.C.8 above. We would very much like to follow up this potentially exciting topic during the next contract period, but proper study of it demands more time than we can afford with our present manpower. If time permits, we may try a small, carefully planned study, probably involving both particulate and gaseous Br (plus Cl and I for comparison).

14. Trace elements in size-fractionated desert soils (Mainz, URI)

As noted in Sections I.B.10 and I.C.9 above, the analysis and tabulation of data from these soil samples is now complete. During the coming contract period, various articles and a large technical report will be written by L. Schütz and K. Rahn. This should be completed within the contract period. Most of this work will be done by Dr. Schütz.

The success of this project has given birth to others. The Max-Planck-Institut für Chemie and the Institut für Meteorologie der Universität, the past and present employers of Dr. Schütz, respectively, are sponsoring a program under which desert soils from all over the world are being systematically collected for eventual analysis. The goal is to build up a trace-element inventory for the aerosol-size fraction of all major desert soils. This program is expected to continue indefinitely; URI will play a small but continuing role in generating and interpreting the data.

15. Cooperative programs (URI, UA)

The importance of cooperative programs with other institutions to our overall Arctic effort has been noted several times in this document. Cooperative programs in force during the 1978-80 contract period were reviewed in Section I.B.6; here we discuss the prospective cooperative programs for the next three years.

Our greatest cooperation will continue to be with the GMCC program. In spite of the withdrawal of support by the Naval Arctic Research Laboratory, GMCC has committed itself to keep the Barrow observatory open at all costs and to keep our program running there. At present, Barrow is in the news probably as much as any of the four GMCC sites (the others are at Mauna Loa, American Samoa, and the South Pole), and has a program of research second in importance only to Mauna Loa.

Similarly, cooperation with Dr. Neal Brown of Poker Flat Research Range will continue. We expect our program at Poker Flat to last many years, and to grow in importance as the atmospheric contrast between interior Alaska and the North Slope becomes more widely recognized. Both

the Poker Flat and Barrow authorities will provide meteorological and/or depositional data to complement our aerosol studies.

Concerning the other sampling sites: the program with the Icelandic Meteorological Office will probably last another 5-10 years (at their urging), with practically no cost to us; the Ireland program with University College Galway (under subcontract) will probably last for only one year; the Paris aerosol sampling, with the Service des Pollutions de la Laboratorie Central, Préfecture de Police, will probably continue through this contract period, again at no cost to us.

Studies with Dr. Joan M. Daisey of New York University, on the organic fraction of the Arctic aerosol, are continuing. We are taking a suite of samples for her on the YMER this summer. Dr. C. Weschler of Bell Laboratories will continue to work with us in trying to track down the source of the high concentrations of silicones in the Barrow atmosphere. Dr. Edward M. Patterson of the Georgia Institute of Technology will continue to measure the absorptive properties of aerosol collected on our Arctic filters. So far, his efforts have been unfunded, but he hopes to acquire some support during the next contract period.

Cooperation between URI and the Norwegian Institute for Air Research will continue indefinitely. URI will analyze more filters from NILU's Bear Island and Spitsbergen sampling sites, the projected field study at Spitsbergen will be arranged through NILU, and a series of joint papers will be written on the research already completed. One of these (Nature, SO_2 at Bear Island) is in its final stages, and another (for the GM conference) is being prepared. Cooperation between URI and NILU has benefited both institutions greatly so far; we see no reason that the future should be different.

Cooperative projects presently being developed with Oregon Graduate Center and General Motors were already discussed in Section I.B.6 above. We expect to be interacting with Dr. T.J. Pepin of the University of Wyoming, as well. Dr. Pepin measures the vertical distribution of aerosol in the Arctic and Antarctic atmospheres via satellite. A genuine cooperation will surely be built up here: he provides us with badly needed vertical information for the Arctic, as well as data from regions otherwise inaccessible to us; we in turn provide him with a broad framework in which to interpret his data.

One of our most-awaited cooperative programs for the coming contract period will be with Environmental Measurements Laboratory. As noted above in Section I.B.6, they are lending us their prototype ERM-3 radon counter for use at Barrow. This unit is highly sensitive, and should easily detect even the lowest concentrations there. It comes intact and completely calibrated, with a computer program for processing the data. Its great strength is that it works completely automatically, with no moving parts, and gives a reading each 2000 seconds. It uses the two-filter system to measure freshly formed ^{218}Po (3.05-min half-life), the first daughter of ^{222}Rn, formed in its own chamber after the first filter, hence involves no assumption of equilibrium between Rn and its daughters. We will pick up the ERM-3 in July 1980, use it at URI during summer and fall to become familiar with it, then hopefully install it at Barrow. We are extremely pleased about acquiring this instrument. The half-hourly data it provides can be compared with the hourly means for a variety of other atmospheric species measured at Barrow, to give an excellent idea of the origin of

the Rn observed there, as well as its dependence on meteorological conditions. The ERM-3 will provide a gold mine of information to us. Together with the ^{210}Pb data we are already generating, it will allow us to better assess the potential for deriving residence times and transit times for pollution aerosol in and around the Arctic from continental radionuclide data (see Section II.B.11 above).

Three cooperative meteorological programs have been already discussed in Section II.B.11 above.

16.   Trace elements in size-fractionated Barrow aerosol
        (Lewis M.S. thesis, URI)

In September 1980, K. Rahn will acquire an M.S. student, Miss Noëlle F. Lewis. Her thesis topic will be determination of trace-element concentrations in the Barrow aerosol as a function of particle size. She will use high-volume cascade impactors and the available URI sampling facilities at Barrow. This work should be very valuable to our Arctic studies because it will: (a) provide an overall size distribution of the Arctic aerosol (not yet available); (b) allow us to estimate the rate of dry and wet deposition of trace elements in the Arctic; (c) provide a record of the way that trace elements have been removed during long-range transport to Barrow (by comparing with original size distributions); (d) allow us to better interpret bulk aerosol-crust enrichment factors derived from filter samples; and (e) provide a test of our recent ideas on the reason for the high Mn/V ratio found at Barrow.

In the past we have refrained from undertaking this project because the impaction surface must be coated with an oil. Selection of the proper oil involves testing many candidates for their trace-element contents, which is no small job. But the addition of Miss Lewis to our roster makes such a study possible.

Although this work will be supported mostly by the Shaw-Rahn NSF grant "Radiative effects of Arctic haze", we mention it here because its results will find broad application within our ONR Contract. We anticipate completion of this thesis two years after the starting date.

17.   Activities of the Arctic Air-Sampling Network

We will continue to work toward the advancement of the Arctic Air-Sampling Network. For at least the next three years, K. Rahn will continue to edit the Arctic Newsletter. It will appear as needed, at irregular intervals, but at a minimum every six months.

The Arctic Network will have small technical meetings in May 1981 and May 1982, then a larger Third Symposium on Arctic Air Chemistry in May 1983. The technical meeting in May 1981 will be held at the Atmospheric Environment Service, Downsview, Ontario, and will be organized by Dr. L.A. Barrie. No locations have been determined for the other two meetings, however. We suspect that URI will organize the Third Symposium.

18. Workshop on aerosol-snow relations (URI)

There is presently an initiative to hold a French-American workshop on the relations between the trace-element content of polar aerosols and snows. The impetus comes from the French side, the group of C. Lorius and C. Boutron in Grenoble, which measures trace elements in Arctic and Antarctic snows. The workshop is to be held in May 1981 in Grenoble. It is to be sponsored by simultaneous proposals to CNRS and NSF. Because of URI's work on trace elements in the Arctic aerosol, K. Rahn has been asked to organize the American portion, and has accepted. We are looking forward to this workship, for we expect it to establish once and for all that the trace-element content of polar snow is not a simple reflection of that of polar aerosol.

19. Statistical studies (URI, UA)

During the Second Symposium on Arctic Air Chemistry, in May 1980, a general consensus developed that statistical techniques would be used with increasing frequency to assist in interpreting data from the Arctic Air-Sampling Network. Several attractive techniques are now available, including cluster analysis, factor analysis, and the presently developing Chemical Element Balance techniques. Dr. N.Z. Heidam of Denmark presented a paper on factor analysis of data from Greenland; his approach is sure to be taken up by others, for example W.E. Raatz of UA.

During the coming contract period we wish to explore the possible application of Chemical Element Balance (CEB) techniques to the Arctic aerosol, in cooperation with persons already using it for other aerosols. The CEB approach has been developed primarily for urban areas. It attempts to account for the mass of aerosol in terms of several sources of aerosol of known composition and the amounts of a marker element for each in the ambient aerosol. We share the same goal for the Arctic aerosol, namely to determine its major sources, but on a different scale: we deal with pollution from various continents instead of from various stacks in a city, for example, and with much longer transport from source to receptor, so that chemical alterations in the atmosphere should be much greater than within an urban area. But the CEB techniques are so close to what we need that we should pursue them. It may very well be that the Arctic will provide the first remote-area, large-scale test of CEB methods.

At this time it is impossible to predict how successful this work will be or how far it will proceed during the next three years. We can only say that we consider it a very promising approach, and will look into it seriously.

C.   Facilities available

URI - Graduate School of Oceanography

Two well-equipped chemistry laboratories, one in the Horn Building
at the Graduate School of Oceanography and the other in the Rhode Island
Nuclear Science Center (RINSC) adjacent to the School of Oceanography,
will be used in this work. A new $30,000 Class 100 clean laboratory has
just been constructed in the RINSC and is available for use. Also availa-
ble for this project are two laminar-flow clean benches. A machine shop
is located on the main campus.

All neutron activation analysis will be performed at the RINSC. The
RINSC swimming-pool reactor operates at two megawatts and delivers a ther-
mal neutron flux of approximately 3×10^{12} n cm^{-2} sec^{-1} at the irradiation
sites. Counting equipment available for this work includes several large
Ge(Li) solid-state detectors and associated electronics, which may be
used with Nuclear Data Model 2200 or Canberra Industries Model 8100 4096-
channel analyzers or a Digital Equipment Corporation PHA-11 pulse-height
analysis system. Data and spectrum reduction on these systems can either
be done via magnetic-tape output with subsequent use of the IBM 370-65
computer on the main campus or directly on the PDP-11/40 computer which
is part of the PHA-11 system.

Also located at the RINSC is a complete atomic absorption analysis
laboratory, including three Perkin-Elmer instruments (Models 360, 503 and
603), HGA-2000 and HGA-2100 heated graphite atomizer attachments and other
accessories.

For possible analysis of sulfate, nitrate, etc., the RINSC has a
Bausch and Lomb Spectronic 21 colorimeter which is freely available for
our use; the atmospheric chemistry group of the GSO also has a Dionex ion
chromatograph for similar analyses. The Graduate School of Oceanogrpahy
also has a Stereoscan S4 scanning electron microscope (Cambridge Scienti-
fic Instruments, Lts.) which is available for general use. It features
two CRT displays, a magnification range of 20-200,000 X and a resolution
in the secondary electron imaging mode of 150A at 30 kV. In addition, the
GSO has a JEOL 50-JXA electron microprobe with 3-wavelength spectrometers
and the KRISEL automation package, including a PDP-11/05 15K computer.
This unit has 70-A resolution and a 1-μm beam.

UA - Geophysical Institute

The Geophysical Institute of the University of Alaska is housed in
the eight-story C.T. Elvey Building which was completed in 1970 on the
West Ridge of the Fairbanks campus. Besides staff offices and laboratories,
the building accomodates supporting services such as machine and carpentry
work, electronics design and fabrication, photography, drafting, stenography,
computing data processing, the library, archives, conference facilities,
etc. The Electronics Shop is experienced in designing, building, testing
and repairing a wide range of scientific instruments. Its capability
ranges from building small metering devices to design and fabrication of
data acquisition systems using the latest in microprocessor control. The
Electronics and Photography Stockrooms maintain a parts inventory of over
6,000 line items. This is a vital source of the latest electronic compo-
nents for new designs, spare parts, supplies, and bulk quantities of film,

paper and chemicals for the Institute's photo needs. The Machine and Carpentry Shop is probably unique in Alaska. Well-equipped with modern tools ranging from a precision jig borer to shapers, planers, milling machines, grinders, lathes and specialized facilities such as anodizing equipment, it includes also the welding and woodworking shops. The Photography Section offers a variety of technical and graphic-arts services, including printing in color and black and white, routine processing of film from geophysical recorders, motion picture processing, reproduction of records, photo-fabrication, and slides for visual presentations. The Drafting Section produces illustrations for articles, reports and slides as required by the scientific staff. The Electron Microscope Laboratory is well equipped for biological and physical applications of electron microscopy. The JEM-6AS transmission electron microscope was acquired in 1964. A new addition to the laboratory is the JSM-35U scanning electron microscope, installed in the fall of 1977. Equipment such as a JEOL vacuum evaporator, a Zeiss compound research light microscope, and three Porter-Blum ultramicrotomes are available for use in specimen preparation.

Computer programming and data processing: The programming group is familiar with a variety of computers and a number of different programming languages and operating systems. The staff is available to write applications programs on the University's new VAX computer system or to provide consulting services for investigators. Time-share terminals to the computer are available throughout the building. A batch-entry station is available in the adjacent O'Neill Building. The Institute's data-processing system is built around a NOVA mini-computer which controls analog/digital concersion, plotting, two magnetic tape drives, and more specialized equipment. Personnel are available for key punching, data entry, scaling and other data-processing services. The Geophysical Institute Library offers a comprehensive coverage of solar-terrestial physics, aeronomy, meteorology, climatology, glaciology, oceanography, the solid-earth sciences, and environmental issues. The present collection includes more than 15,000 volumes, supplemented by reports and data compilations. The library subscribes to some 430 journals, magazines and newsletters from all over the world. The Geophysical Archives have been recently combined with the remote-sensing data library to provide shared facilities and a convenient broad data base. This combined facility is located adjacent to the library, which provides easy accesss to scientific books, journals and reports as well. The Geophysical Archives contain a worldwide collection of auroral and magnetic records, together with a more selective sampling of records in the other geophysical disciplines, and the most complete collection of Alaskan remote-sensing data in the State. This includes Landsat and NOAA radar imagery and microform listings of current imagery acquisitions. Analytical equipment is also available, such as a roll-film viewer, microfilm reader, microfiche reader/printer light tables, and stereoscopes.

D. <u>Current support and pending applications</u>

<u>Dr. Kenneth A. Rahn - Associate Research Professor, University of Rhode Island</u>

<u>Title</u>	<u>Agency</u>	<u>Duration</u>	<u>Amount</u>	<u>P.I. Time</u>
<u>Current support</u>				
Arctic Haze: Natural or Pollution? (This contract, N00014-76-C-0435; with Dr. G.E. Shaw of University of Alaska)	ONR	1 Oct. 1978 - 30 Sept. 1980 (Renewal expected)	$149,761 FY80 (URI portion $114,058)	70%
Aerosol-snow fraction- ation of trace elements (with Dr. R.J. McCaffrey) (ATM78-17328)	NSF	1 June 1979 - 30 Nov. 1980	$76,700	8%
Methyl iodide and the atmospheric iodine cycle (with Dr. R.A. Duce and Mr. E.L. Butler of URI) (ATM78-16516)		15 Oct. 1978 - 30 Apr. 1982	$36,000 FY80	7%
Radiative effects of Arctic haze (with Dr. G.E. Shaw of the Univer- sity of Alaska) (DPP79- 19816)	NSF	15 May 1980 - 14 May 1982	$23,000 First year	15%
<u>Pending applications</u>				
This proposal	ONR	1 Oct. 1980 - 30 Sept. 1983	$149,000 FY81 (URI share $114,000)	75%

<u>Dr. Glenn E. Shaw - Associate Professor, University of Alaska</u>

<u>Current support</u>				
Arctic Haze: Natural or Pollution? (This contract, N00014-76-C-0435; with Dr. K.A. Rahn of the Univer- sity of Rhode Island)	ONR	1 Oct. 1978 - 30 Sept. 1980 (Renewal expected)	$35,703 (UA portion)	4 mo. (FY79,80)
Magnitude and Variability of Solar Spectral Irradiance (ATM78-12185)	NSF	1 June 1979 - 30 Nov. 1980	$107,870	3.5 mo.
Submicron Particles in Antarctica (DPP79-20804)	NSF	1 Aug. 1980 - 31 Jan. 1982	$53,380	3 mo.

D.   <u>Current support and pending applications</u> (cont.)

Title	Agency	Duration	Amount	P.I. Time
Radiative Effects of Arctic Haze (with Dr. K.A. Rahn of University of Rhode Island) (DPP79-19816)	NSF	15 May 1980 - 30 Oct. 1981	$82,881	4 mo.

<u>Pending applications</u>

Title	Agency	Duration	Amount	P.I. Time
This proposal	ONR	1 Oct. 1980 - 30 Sept. 1983	$34,892 (UA portion, FY81)	1 mo. (FY81)
A Twilight Radiometry Study of the Temporal Evolution of the Mt. Saint Helens Stratospheric Dust Veil	NSF	1 year	$26,393	
The Magnitude and Variability of Solar Spectral Irradiance (Continuation of ATM78-12185)	NSF	1 year	$69,261	
Final Design Development of a Multiwavelength Solar Radiometer Intended for Solar Energy Application	DOE	1 year	$43,181	

E. Budget - Fiscal Year 1981

		URI	UA
(1)	Salaries		
	Co-Principal Investigator Dr. Kenneth A. Rahn, Associate Research Professor - 9 months	24,000	
	Mr. Thomas J. Conway Research Specialist - 12 months	17,000	
	Ms. Carla N. Pickering Administrative Secretary - 7 months	6,400	
	Co-Principal Investigator Dr. Glenn E. Shaw Associate Professor - 1 month		4,312
	Mr. Wolfgang E. Raatz Research Assistant - (70% of 7.5 mo. @ 1565) (30% cost-shared by Geophysical Institute)		8,216
	Electron Microscope Technician - ½ month @ 3030		1,515
	SUBTOTAL		14,043
	7.6% salary increment		1,067
	SUBTOTAL		15,110
	Reserve for annual leave 11%*		641
	Holiday and sick leave 9.5%*		554
	TOTAL SALARIES	47,400	16,305
(2)	Staff benefits		
	19% of 41,000	7,790	
	23% of 6,400	1,472	
	15% of 16,305		2,446
	TOTAL STAFF BENEFITS	9,262	2,446
(3)	Permanent equipment		
	High-volume aerosol-sampling station for Nord, Greenland	1,500	
	Computer/word processor (partial)	2,000	
	TOTAL PERMANENT EQUIPMENT	3,500	
(4)	Expendable equipment and supplies	4,000	2,500

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*Annual leave, holiday and sick leave not calculated
on Research Assistant.

E.   Budget - Fiscal Year 1981 (cont.)

		URI	UA
(5)	Travel		
	Domestic:		
	Miscellaneous meetings	1,000	
	TOTAL DOMESTIC TRAVEL	1,000	
	Foreign:		
	Arctic Network Technical Meeting, Toronto, May 1981 (3-5 persons)	1,000	
	GAF Meeting, West Germany, October 1980 (KAR)	1,000	
	Nord, Greenland, spring 1981 (TJC)	1,000	
	TOTAL FOREIGN TRAVEL	3,000	
	TOTAL TRAVEL	4,000	
(6)	Publications	1,250	200
(7)	Other		
	Computer	500	
	Maintenance of sampling & analytical equipment	500	
	Freight	2,000	
	Telephone	1,000	
	Meteorological maps	500	
	R. Borys, Colorado State University, measurements of cloud-active aerosol	5,000	
	Illustration services	500	
	Aircraft charges for winter 1980-81 aerosol-sampling experiment (60 hours @ $200)	8,000	4,000
	TOTAL OTHER	18,000	4,000
(8)	Indirect costs		
	Overhead @ 56.3% of total salaries	26,686	
	Overhead @ 57.9% of total salaries		9,441
	TOTAL COSTS	114,098	34,892
	GRAND TOTAL	148,990	

F. Comments on the budget - Fiscal Year 1981

We are submitting a budget for only the first year of this three-year contract, in accord with ONR practice. We anticipate similar levels of real effort for years 2 and 3, with total costs of approximately $162,800 and $178,000, respectively.

At the request of the ONR Arctic Program, our budget for FY 1981 has been kept to just under $149,000, slightly less than the FY 1980 level of $149,761. In real terms this means sizeable cuts in nonsalary items, for major salary increases have been received or are expected for K. Rahn, T. Conway, C. Pickering and G. Shaw, and W. Raatz has been taken on by the University of Alaska. In addition, the University of Rhode Island is charging an additional 19-23% of salaries for fringe benefits. In FY 1979, 56% of our Contract funds went to salaries and overhead; in FY 1980 it was 61%; now, in FY 1981, it is 75%.

Degree of effort

At the University of Rhode Island, the scientific time devoted to this Contract remains the same as last year, 9 months for K. Rahn and 12 months for T. Conway. Our secretary, C. Pickering, is reduced slightly, from 9 months to 7 months. In actuality, she will spend the same amount of time on this Contract, because 25% of her salary will now come from the Graduate School of Oceanography.

At Colorado State University, R.D. Borys continues to receive a one-half graduate stipend.

At the University of Alaska, G. Shaw decreases from 2 months in FY 1980 to 1 month this year (for budgetary reasons only—he desires to spend much more time than this on Arctic haze), but W. Raatz, a second-year Ph.D. student, will contribute 5.25 months to the Contract. He will actually spend 7.5 months on the Contract, for 30% of his salary will come from the Geophysical Institute; his thesis is exclusively on Arctic haze. Overall, then, the scientific effort by the Geophysical Institute will be more than doubled.

Permanent equipment

The only permanent equipment we are requesting this year is an aerosol-sampling station for Nord, Greenland ($1,500) and partial funds ($2,000) for a computer/word processor, both for URI. The computer/word processor will have a variety of uses within this Contract: correspondence within the Arctic Network, mailing of the Arctic Newsletter, data storage and retrieval for our Arctic chemical data bank, the increasingly complex numerical simulations of long-range transport of aerosols and gases to the Arctic, and assisting with the large volume of papers that we expect to publish during the next 2-3 years. The rest of the funds will come from a variety of sources.

Travel

Our travel budget of $4,000 for FY 81 is considerably smaller than last year's budget of $24,500. This is because Mr. Borys' extensive field studies will finish in summer 1980, and because almost all of our field work for FY 1981 will be associated with Mr. Heckert's aircraft flights.

It is not yet certain that K. Rahn will attend the GAF meeting in W. Germany in October, 1980. If not, this money will be used for other foreign travel that is sure to arise.

We are deliberately vague about details of domestic travel because experience has shown that it can no longer be laid out neatly a year in advance. The level of $1,000 is reasonable, based on past years.

Publications

We are requesting a total of $1,450 for publications this year. We expect to spend all this, for there will be many publications, as mentioned in Section II.A above.

Other costs

The principal items in this category are Mr. Borys' graduate stipend at CSU and Mr. Heckert's aircraft costs. A new wrinkle at URI is charges for illustration services, at $9.50 per hour. Because of the number of drawings we expect to have made, we have budgeted $500 for this.

G.   Biographical Information

Co-Principal Investigator

NAME: Kenneth A. Rahn

EDUCATION: B.S. Massachusetts Institute of Technology, 1962
             (Chemistry)

             Ph.D. University of Michigan, 1971
             (Meteorology)

PROFESSIONAL
EXPERIENCE:

1980 - Present	Associate Research Professor, Graduate School of Oceanography, University of Rhode Island
1979 - Summer	Visiting Scientist, Norwegian Institute for Air Research, Lillestrøm, Norway
1976 - 1979	Research Associate, Graduate School of Oceanography, University of Rhode Island
1975 - 1976	Invited Visiting Scientist Max-Planck-Institut für Chemie, Mainz, W. Germany
1973 - 1975	Research Associate, Graduate School of Oceanography, University of Rhode Island
1971 - 1973	Research Associate, Institute for Nuclear Sciences, University of Ghent, Belgium
1968 - 1971	Graduate Assistant, University of Michigan Ann Arbor, Michigan
1970 - Summer	Lawrence Radiation Laboratory, Livermore, California
1963 - 1968	Science and Mathematics teacher Classical High School and Barrington College Providence, Rhode Island

PROFESSIONAL
SOCIETY
MEMBERSHIPS :   American Chemical Society
                  American Association for the Advancement of Science
                  American Meteorological Society
                  Gesellschaft für Aerosolforschung

RECENT
PUBLICATIONS:

Dams R., Robbins J.A., Rahn K.A. and Winchester J.W. (1970) Non destructive
     neutron activation analysis of air pollution particulates, Anal. Chem.
     42, 861.

Harrison P.R., Rahn K.A., Dams R., Robbins J.A., Winchester J.W., Brar S.S. and Nelson D.M. (1971) Areawide trace metal concentrations in Northwest Indiana as determined by multielement neutron activation analysis: a one-day study, J. Air Poll. Cont. Assoc. 21, 563.

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John W., Kaifer R., Rahn K. and Wesolowski J.J. (1973) Trace element concentrations in aerosols from the San Francisco Bay area, Atmos. Environ. 7, 107.

Rahn K., Demuynck M., Dams R. and DeGraeve J. (1973) The chemical composition of the aerosol over Belgium, Proceedings of the Third International Clean Air Congress, Düsseldorf, Germany, C81-C84, VDI-Verlag GmbH, Düsseldorf, West Germany.

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Rahn K.A. (1975) Chemical composition of the atmospheric aerosol: A compilation I. Extern 4. 286-313.

Rahn K.A. (1975) Chemical composition of the atmospheric aerosol: A compilation II, Extern 4, 639-667.

Rahn K.A. and Harrison P.R. (1976) The chemical composition of Chicago street dust, In: Atmosphere-Surface Exchange in Particulate and Gaseous Pollutants (1974), Symposium Proceedings, CONF-740921, NTIS, U.S. Dept. of Commerce, Springfield, VA 22161, pp. 557-570.

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G.   Biographical Information (cont.)

Co-Principal Investigator

NAME: Glenn E. Shaw

EDUCATION:

B.S. Montana State University, 1963
M.S. University of Southern California, 1965
Ph.D. University of Arizona, 1971

POSITIONS HELD & EXPERIENCE:

Engineering Associate, Engineering Research Laboratory, Bozeman,
      Montana, 1962-1963. Development of Electronics.
Research Associate, Argonne Laboratories, EBR II, Idaho Falls,
      Idaho, Summer 1962. Nuclear reactor technology.
Hughes Fellow, Hughes Aircraft Company, Electron Dynamics Division,
      Inglewood, California, 1965-1967. Electron dynamics in
      microwave devices.
Research Assistant, University of Arizona, 1965-1968. Physics
      of lightning and atmospheric electricity research.
Research Associate, University of Arizona, 1968-1971. Studies
      involving radiative transfer through the atmosphere.
Assistant Professor of Geophysics, Geophysical Institute, University
      of Alaska, May 1971-July 1974.
Associate Professor of Geophysics, Geophysical Institute, University
      of Alaska, July 1974-present.
Sabbatical leave at World Radiation Center, Davos, Switzerland
      March to September, 1977.

HONORS:

Tau Beta Pi Honorary Fraternity, March 1963.
Howard Hughes Fellowship, September 1963.
Meritorious teaching award, University of Alaska Alumnus Association,
      May 1980.

PROFESSIONAL ORGANIZATIONS:

American Association for the Advancement of Science
American Geophysical Union
American Meteorological Society
Royal Meteorological Society

PRESENT RESEARCH INTERESTS:

Atmospheric physics: application
and interpretation of optical methods to derive physical pro-
perties of the atmosphere; physics of aerosols. Studies of effects
on climate due to changes in trace gases or aerosols.

PH.D. DISSERTATION:

An Experimental Study of Atmospheric Turbidity Using Radiometric
Techniques, May, 1971, The University of Arizona.

Published Articles

Shaw, G. E., Cosmic count increase associated with thunderstorms,
J. Geophys. Res., 72, 4623, 1967.

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Telescope, 33, No. 6, 394-395, June 1967.

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Lemmon, Arizona, J. Geophys. Res., 75, No. 12, 2159-2164,
April 1970.

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McCall Glacier in northern Alaska, Proceedings of the Atmo-
spheric Radiation Conference, Fort Collins, Colorado, August
7-9, 1972.

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atmospheric extinction using direct solar radiation measure-
ments made with a multiple wavelength radiometer, J. of
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green flash, Pure and Applied Geophysics, 102, 223, 1973.

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of Climate of the Arctic, 24th Alaska Science Conference,
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        on climate, Science, Vol. 192, pp. 1334-1336, June, 1976.

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        multispectral measurements of atmospheric transparency, Antarctic
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        St. Augustine 1976 Eruption ash cloud, Science, Vol. 193,
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