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THE EURASIAN SOURCES OF ARCTIC AEROSOL

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Table of Contents

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Introduction

There is now considerable evidence that during winter the Arctic atmosphere contains surprisingly large amounts of submicron aerosol. High concentrations of sulfate, ^{210}Pb, and trace elements such as V and Mn suggest strongly that much of this aerosol is the product of aging of polluted air masses from midlatitudes, although some of it may be natural. The concentrations and compositions are similar for the aerosols of northern Norway, Bear Island, Spitsbergen, and Barrow (Alaska), suggesting a basic unity of the Arctic aerosol. An overview of the present knowledge and understanding of the Arctic aerosol can be obtained from Rahn et al. (1977), Rahn (1978), Rahn and Shaw (1978), Rahn and McCaffrey (1979a), Kerr (1979), Rahn and McCaffrey (1979b), Larssen and Hanssen (1979), and Rahn and McCaffrey (1979c).

Potential sources of the Arctic aerosol include the northeastern United States, Europe/UK, the European USSR and eastern Asia (Japan, Korea, China). Possible pathways to the Arctic are shown in Figure 1. The weight of recent evidence, as follows, points more and more to the Euroasian region as the major source of Arctic aerosol:(1) High aerosol concentrations at both Barrow and Fairbanks, Alaska are associated with cold air from the north rather than with warm air from the south. This association appears to eliminate eastern Asia via the Pacific pathway as a major source. Presumably, pollution aerosol from eastern Asia is effectively removed from the atmosphere during transport across the Pacific Ocean along the major storm tracks, where precipitation is abundant. (2) Analogous reasoning would be that aerosol from the northeastern United States would not reach the Arctic in significant amounts

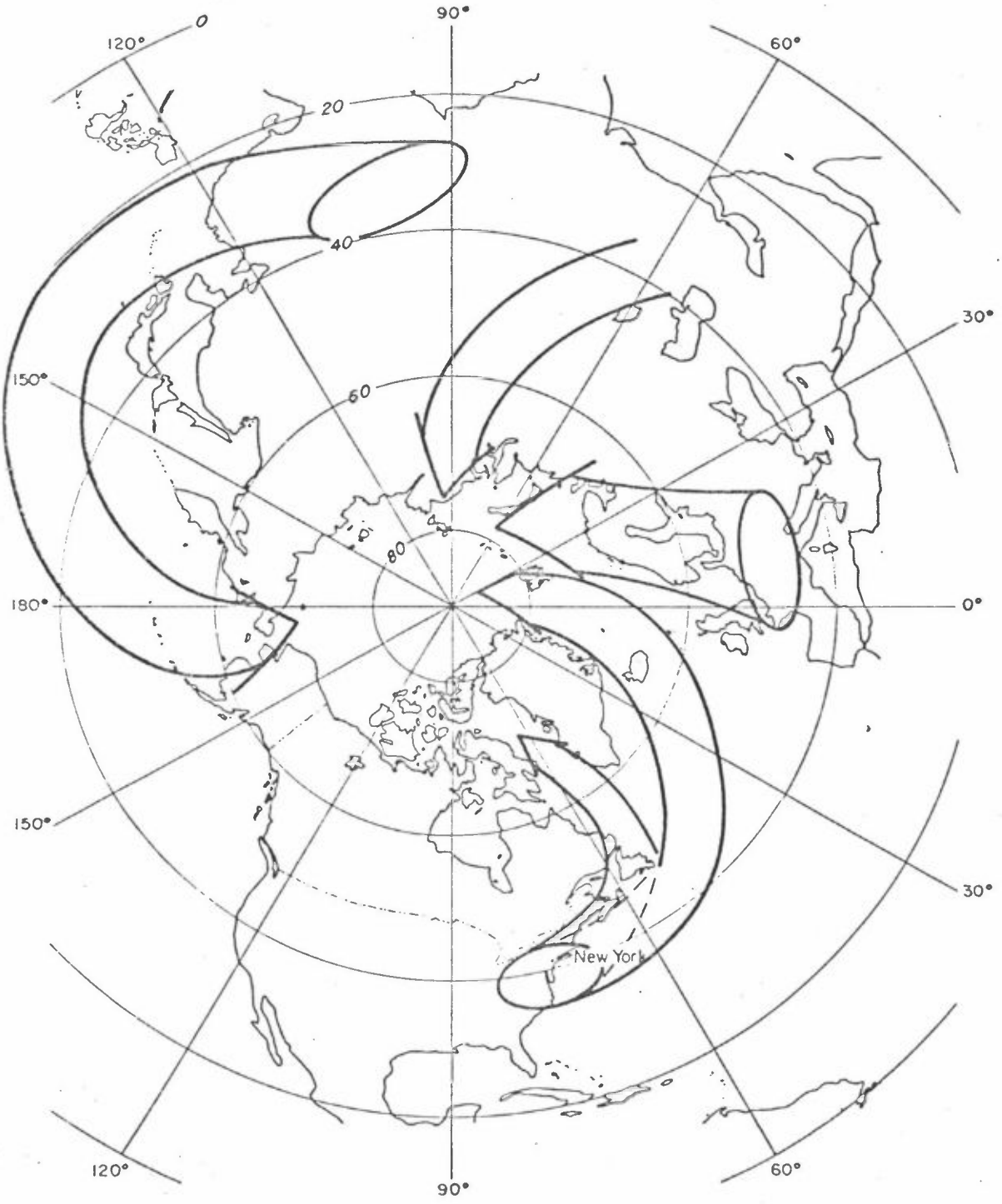

Figure 1: Possible pathways of pollution aerosol to the Arctic.

via an Atlantic pathway, because most of it would be removed in the storm belts there. Available data, although scant, appear to confirm this idea. Concentrations of $SO_4^=$ and V in both northern and southern Greenland during winter 1977-78 were several times lower than in the Norwegian and Alaskan Arctic. In studies of sulfate over the northern Atlantic between Europe and North America, Gravenhorst (1978) has also found relatively low concentrations (mean of 0.9 $\mu g\ m^{-3}$). Although these measurements were on ships, they can be considered representative of more than just the marine boundary layer, for the following reasons:(a) Concentrations of sulfate-containing particles over the Bay of Biscay and the Canary Islands show highest concentrations near the surface and monotonic, moderate decreases with altitude (Gravenhorst, 1978).(b) A similar vertical distribution is seen for sulfate concentations over Europe, as summarized by Georgii (1978). (Concentrations of SO_2 over Europe and the North Atlantic also seem to decrease or remain constant with height up to 4 to 5 km (Georgii, 1978)). Thus, there exists a broad minimum in concentration of sulfate over the North Atlantic in winter, relative to both the northeastern United States and the Arctic. Aerosol transport between these two locations would therefore have to occur via a pathway quite different than either of those shown in Figure 1 (via eastern and western Greenland), which is unlikely for meteorological reasons. (3) Numerical simulations of aerosol transport to the Arctic (Rahn and McCaffrey, 1979a) have shown that the winter Arctic aerosol is compatible with a Eurasian precursor but apparently not with a North American precursor and a North Atlantic pathway. (4) Winter maps of $SO_4^=$ and V concentrations (Rahn and McCaffrey, 1979c) reveal a broad tongue of high concentrations extending northward from Eurasia into the Norwegian Arctic and apparently into the Alaskan Arctic as well. The most reasonable conclusion from this pattern is that Eurasia is in fact the main source of the Arctic aerosol.

A Eurasian source makes good physical sense. The zones of major pollution emissions in Europe lie roughly 15^o farther

north than the corresponding zones in the northeastern United States. A pathway from Europe to the Arctic will be 2000 to 5000 km long, whereas one from North America via Iceland will be at least 8000 km long. Lastly, paths from Europe to the Arctic are mostly over continents, where relatively low rainfall increases aerosol residence times, whereas paths over the North Atlantic should have more precipitation and, hence, enhanced removal of aerosol.

Within the Eurasian sphere, there are a number of pieces of recent evidence that a pathway via the western or European USSR is important: (1) In the numerical simulations of Rahn and McCaffrey (1979a) a delayed pulse of ^{222}Rn was needed to provide the high ^{210}Pb and $^{210}Pb/SO_4^{=}$ at Barrow. This pulse was interpreted as coming from the western USSR as European air passed ENE-ward, then northward into the Arctic. The entrance-zone to the Arctic was estimated to be centered on Novaya Zemlya. (2) According to Bolin and Persson (1975), the mean flow of polluted UK air during winter is in fact toward the ENE. From London, for example, the mean trajectory reaches the vicinity of Stockholm after 4 days. An extension of this trajectory would pass over the western USSR and slowly curve northward. (3) Maps of mean surface pressure, such as that for January reproduced in Vowinckel and Orvig (1970), show a broad zone where air flows northward from the USSR into the Arctic. For January this zone is between Novaya Zemlya and the New Siberian Islands. Much of this air has previously come from Europe. (4) Streamlines of mean January air flow yield the same conclusion (Trewartha, 1968). (5) Recent examination of the meteorological conditions during periods of high aerosol concentration at Bear Island in winter has shown that the most common surface air flow was from the NE, at least for episodes between mid-December 1977 and mid-March 1978. This flow was caused by one or more low-pressure areas located along the Norwegian, Barents, and Kara seas, the mean result of which is clearly shown in the January pressure map in Vowinckel and Orvig (1970). Air flow into these lows seemed to be generally from the south, and appeared to take

place between the Kola Peninsula and the Taymyr Peninsula. In broad terms, the air flowing from the NE to Bear Island appeared to have come most immediately from the western USSR (between roughly Leningrad - Moscow and the Urals), and before that probably from Europe. We have thus dubbed these "return-flow" episodes, to emphasize the difference between "direct-flow" episodes of the more expected type which are the rule in earlier fall and later spring. The apparent path of air in a return-flow episode coincides very nearly with the path proposed by Rahn and McCaffrey (1979a): "(From Europe) The air mass moves to the northeast, into European Russia....., then at some point turns northward to the Arctic ..... passing over Novaya Zemlya, then splitting (in the mean) into two brances, one which flows southward along the east coast of Greenland and the other which flows westward along northern Greenland and the Canadian Arctic Islands, eventually reaching Barrow". This path is shown in Figure 2 (after Kerr, 1979).

Goal of the present work

This evidence for a path over the USSR into the Arctic raises the question of the relative contributions of Europe and the USSR to the pollution component of the Arctic aerosol. The previous numerical simulations of Rahn and McCaffrey (1979a) considered pollution contributions from only Western Europe; the only role attributed to the USSR was as a source of natural ^{210}Pb. But Eastern Europe and the USSR obviously contribute to Arctic sulfate as well, as can be seen either from the older OECD emissions survey (Semb, 1978) or from the newer EMEP survey (Dovland and Saltbones, 1979). We therefore decided to use the calculation method of Rahn and McCaffrey (1979a) to estimate the relative contribuions of Western Europe, Eastern Europe, and the western USSR to Arctic sulfate, with particular attention given to the USSR sources. The methods, results, and implications of these calculations form the rest of this document.

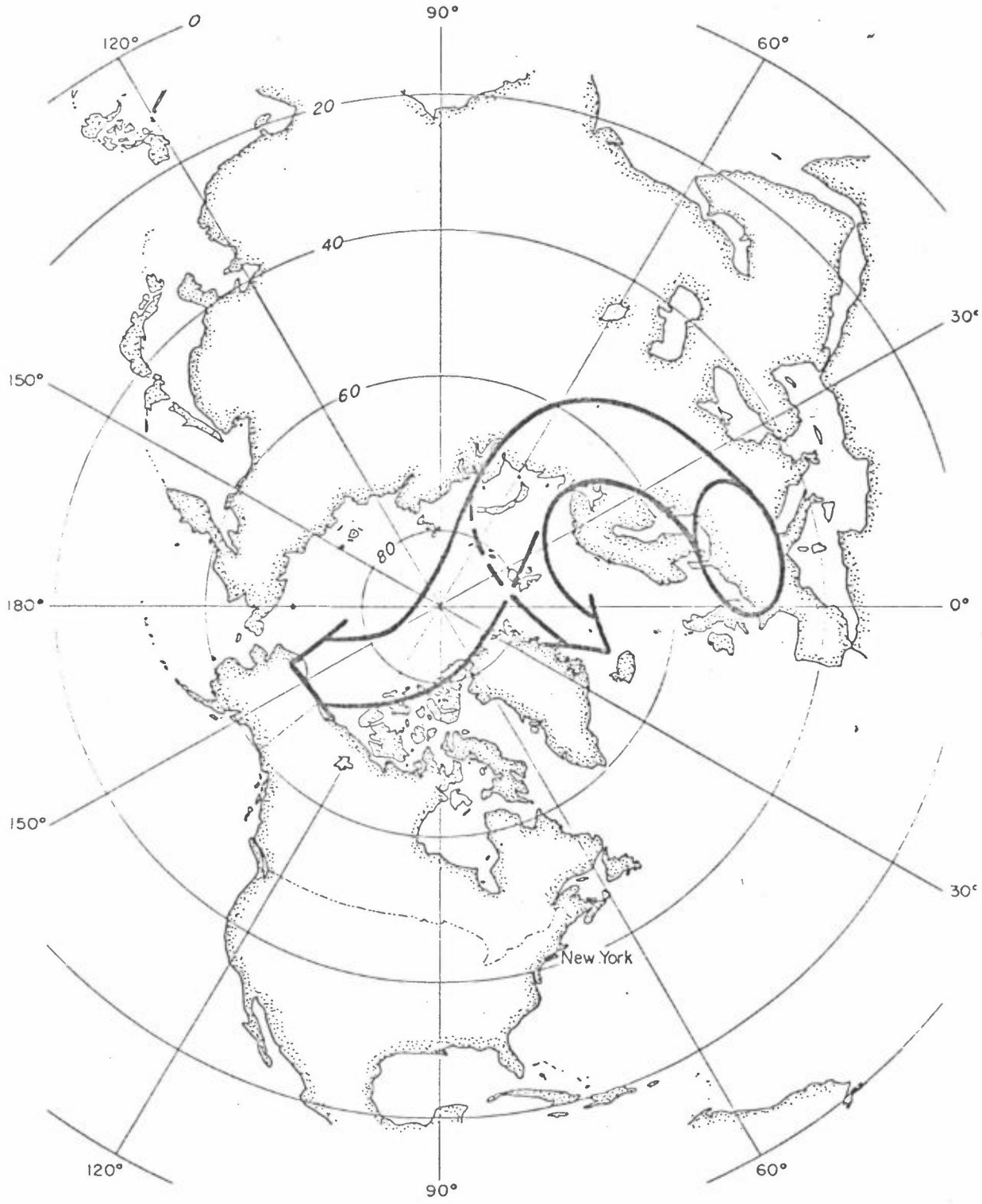

Figure 2: The proposed "return-flow" pathway from Europe via the western USSR to the Arctic.

Pathways considered

In order to perform the most realistic simulations possible, more objective information on entrance-zones of Eurasian air into the Arctic was needed than could be provided by the above-mentioned survey of synoptic conditions associated with high aerosol concentrations at Bear Island. A new survey was thus carried out, admittedly semiquantitative, of the longitudional zones near latitude 70°N where air flowed northward into the Arctic. All daily hemispheric surface maps for 00Z from October 1977 through April 1978 were examined; zones of northward flow (based on both isobaric patterns and observed winds) which could penetrate significantly into the Arctic (admittedly somewhat subjective - the criterion was that the air had to have the possibility of traveling long distances within the Arctic; flow which halted just inside the Arctic or which was nearly zonal was rejected) were recorded, usually to the nearest 10° of longitude on either end. The region from Greenland eastward to the Bering Strait was examined, but flow of Pacific air northward over the Bering Strait was not counted because it does not contribute to Arctic pollution. Flow near Greenland was considered, however, because European pollution can often enter the Arctic via retrograde flow (over Iceland) through this area. It must be emphasized here that this was considered a feasibility study only. This type of analysis ought to be done by computer for different levels in the atmosphere, using sophisticated programs such as those of E.R. Reiter of Colorado State University. Reiter's programs analyze three-dimensional meteorological data objectively and print out vertical cross-sections along a latitudinal circle for, say, the meridional component of the wind at a given instant in time. A survey of such maps daily for a period of 1 to 2 years could give a broadly based picture of where and when air enters the Arctic, as well as an idea of its recent history. In the absence of such programs, however, a relatively quick effort like this one offered the opportunity to develop a feeling for atmospheric motions that could guide the subsequent numerical simulations.

Large seasonal variations in the frequency of northward transport into the Arctic were indeed seen, with a pattern which agrees semiquantitatively with aerosol concentrations there. (Here it should be noted that this analysis is primarly of frequency of northward transport and not of intensity, so that results with better-than-semi-quantitative validity should not be expected. Intensity of transport is indirectly included, however, by the criterion of penetration into the Arctic.) The results are shown in Table 1, as the number of "degree-days" for each of the seven months, degree-days being defined as the widths in degrees longitude of northward transport across the 70th parallel, between Greenland and the Bering Strait, summed for each month.

Table 1: Relative monthly transport of air to the Arctic.

Month	Degree-days across 70°N, Greenland-Bering Strait
October 1977	990
November 1977	1700
December 1977	2760
January 1978	1925
February 1978	2160
March 1978	2690
April 1978	740

Several interesting features of this table can be noted: (1) A seasonal variation of a factor of 3-4x is seen even without high summer being included. Actual seasonal variations of Arctic aerosol, after correction for variations at the source (about a factor of three), are roughly a factor of 7 in the Alaskan Arctic and somewhat less in the Norwegian Arctic, more like a factor of 4 to 8. Thus, our factor of 3-4 agrees surprisingly well with actual concentration data in the Arctic. (2) The winter period of transport of Table 1 (November through March) coincides closely with the actual time of most concentrated aerosol in the Arctic. (3) The January-February minimum seen here resembles a January minimum often seen at Barrow,

but this may be fortuitous, because the Barrow minimum seems to be caused by northward transport of Pacific air into the Alaskan Arctic. There may, however, be a meteorological link between stron northward transport over the Bering Strait and diminished northward transport on the other side of the Arctic. No pronounced January-February minimum in $SO_4^=$ concentration was seen at Bear Island, in any event.

An idea of the principal zones of transport into the Arctic can be seen from monthly frequency distributions as a function of longitude, as shown in Figure 3. (The resolution into components shown in this figure are one interpretation only, and subject to discussion and revision). There seem to be two recurring broad zones of transport, centered respectively on northern Scandinavia-Kola Peninsula and the Taymyr Peninsula. There is evidence, particularly during February, for a third intermediate zone, centered on Novaya Zemlya. We propose that these two basic zones of transport are so widely separated because they are caused by different meteorological mechanisms: transport over Scandinavia takes place in the confluence of a low-pressure system near Iceland and a high-pressure system centered over Europe or the western USSR, whereas transport over the Taymyr region results from the interaction of the Asiatic high in its normal winter position in southern Siberia and eastern Kazakhstan with the easternmost extension of the Icelandic low over the Barents and Kara Seas. In the former case aerosol approaches Bear Island from the east along a track with anticyclonic curvature; in the latter case it approaches Bear Island from the northwest along a cyclonic path.

Figure 3 shows the relatively large monthly variations in transport path that can take place - at present it is not known whether these patterns are reproducible or random. But for this period, at least, transport over Scandinavia was common during October, November and December, decreased greatly during January and February, was strong again during March, and weak again during April when the summer situation of decreased transport in all sectors began to be established. Transport via the Taymyr sector was weak in October, increased

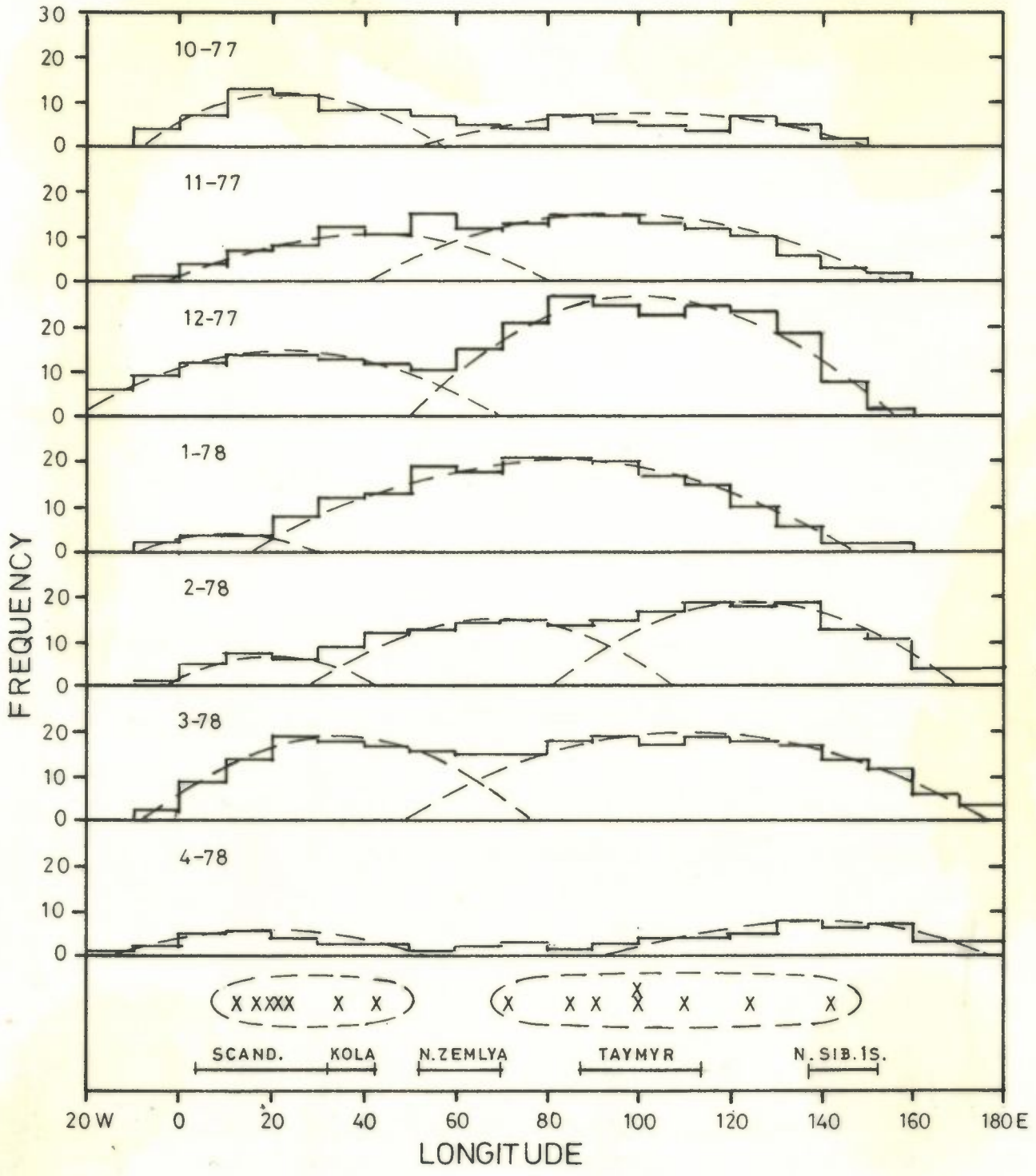

Figure 3: Monthly frequency distribution of entrance-zones of northward air flow to the Arctic, October 1977 through April 1978. Crosses represent centers of the components of each monthly distribution and tend to cluster around two entrance-zones.

in November, was strong in December through March, and decreased in April.

This figure also offers evidence for the westward migration of the USSR zone of transport in high winter proposed from surface pressure maps and chemical data by Rahn and McCaffrey (1979c). The USSR zone is seen to migrate about 20° to 30° westward from October to January-February, and then about 70° eastward in April. This progression is in qualitative agreement with the intensity and therefore westward extension of influence of the Asiatic high while its center remains in the normal winter location.

Also reflected in Figure 3 is the general dominance of return-flow transport to the Arctic between December and March already noted and commented on above.

The total frequency distribution for October 1977 through April 1978 is shown in Figure 4, resolved into its two major components with a break point placed at 60°E (the Urals), the most common divider for the individual months shown in Figure 3. (This does not necessarily imply that the Ural Mountains actually cause part of the separation of pathways. This idea is quite plausible, however, and deserves to be examined further). The ratio of transport frequency through the USSR compared to Scandinavia is then 2/1.

Aerosol sources and specific pathways

As mentioned above, only the SO_2 - $SO_4^{=}$ system is considered here, with the new data on SO_2 emissions of Dovland and Saltbones (1979). The principal changes in this 150-km EMEP grid relative to the data in the 127-km OECD grid (Semb, 1978) are reduced emissions from the Moscow and Leningrad regions, with compensating increases in the Don Basin, and the appearance of a narrow, N-S elongated region of strong emissions in the southern Urals, centered near Sverdlovsk. These latter emissions seem high relative to the local population density, and so would seem to be primarily associated with heavy industry in that region (a great deal of mining and smelting, particul-

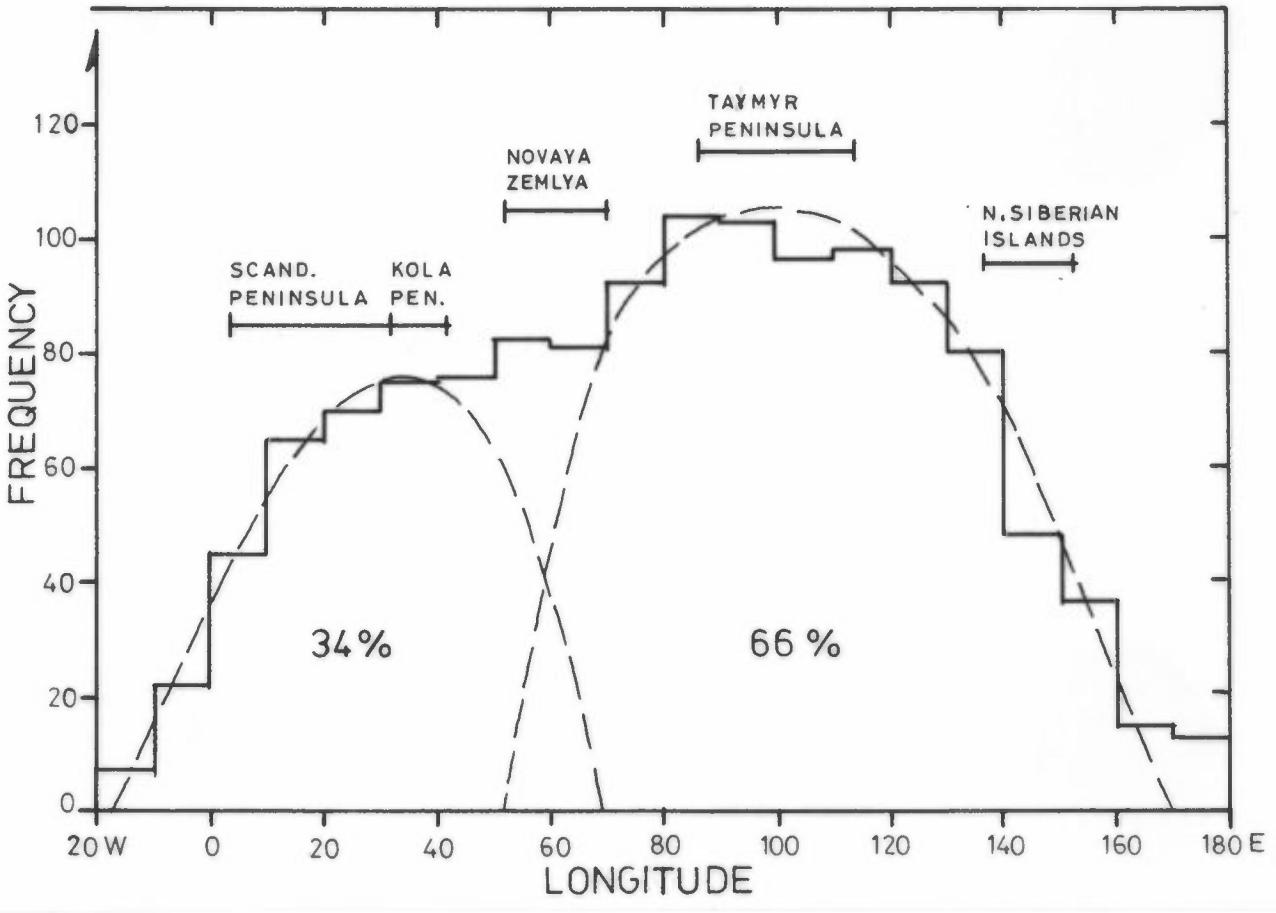

Figure 4: Total frequency distribution of entrance-zones of northward air flow to the Arctic, October 1977 through April 1978.

arly of copper and tin). The new grid also shows the emissions from Nikel on the northern border of Norway and Russia, whose strength is estimated at 100 000T SO_2 per year, about 10% of Sudbury, Ontario's strength.

There are then 5 major source regions of SO_2 to consider: Europe/UK, Moscow, Leningrad, Sverdlovsk, and Nikel. (For the moment we assume Donetz to be too far south to be along a major pathway to the Arctic). Of these, Europe/UK is clearly the strongest. For the present we consider both Leningrad and Nikel to be NW of the main transport paths through the USSR, and neglect them, although they are surely of influence from time to time.

We are left then with the Moscow and Sverdlovsk regions as the principal USSR sources to consider. Although estimates of emissions from Moscow are now reduced, they remain at roughly 75% of their former values. Thus, Moscow could well have a sizeable influence on the Arctic aerosol because of its proximity to the Arctic. A similar conclusion can be reached for the Sverdlovsk region, whose emissions of SO_2 are about equal to Moscow's.

Because Figures 3 and 4 show that USSR air can enter the Arctic anywhere between Novaya Zemlya and the New Siberian Islands during winter, pathways from Europe through the USSR to the Arctic must consider emissions from both the Moscow and Sverdlovsk regions. We assume that all major pathways pass over Moscow, but because Figure 3 shows that pathways over Novaya Zemlya (which would miss Sverdlovsk) also occur during winter, we have performed our calculations for two pathways, as shown in Figure 5. One, called the "short path", extends from Europe to Moscow, then curves northward and enters the Arctic over Novaya Zemlya. The other, called the "long path", originates in Europe and passes over both Moscow and Sverdlovsk before turning northward and entering the Arctic via the Taymyr Peninsula.

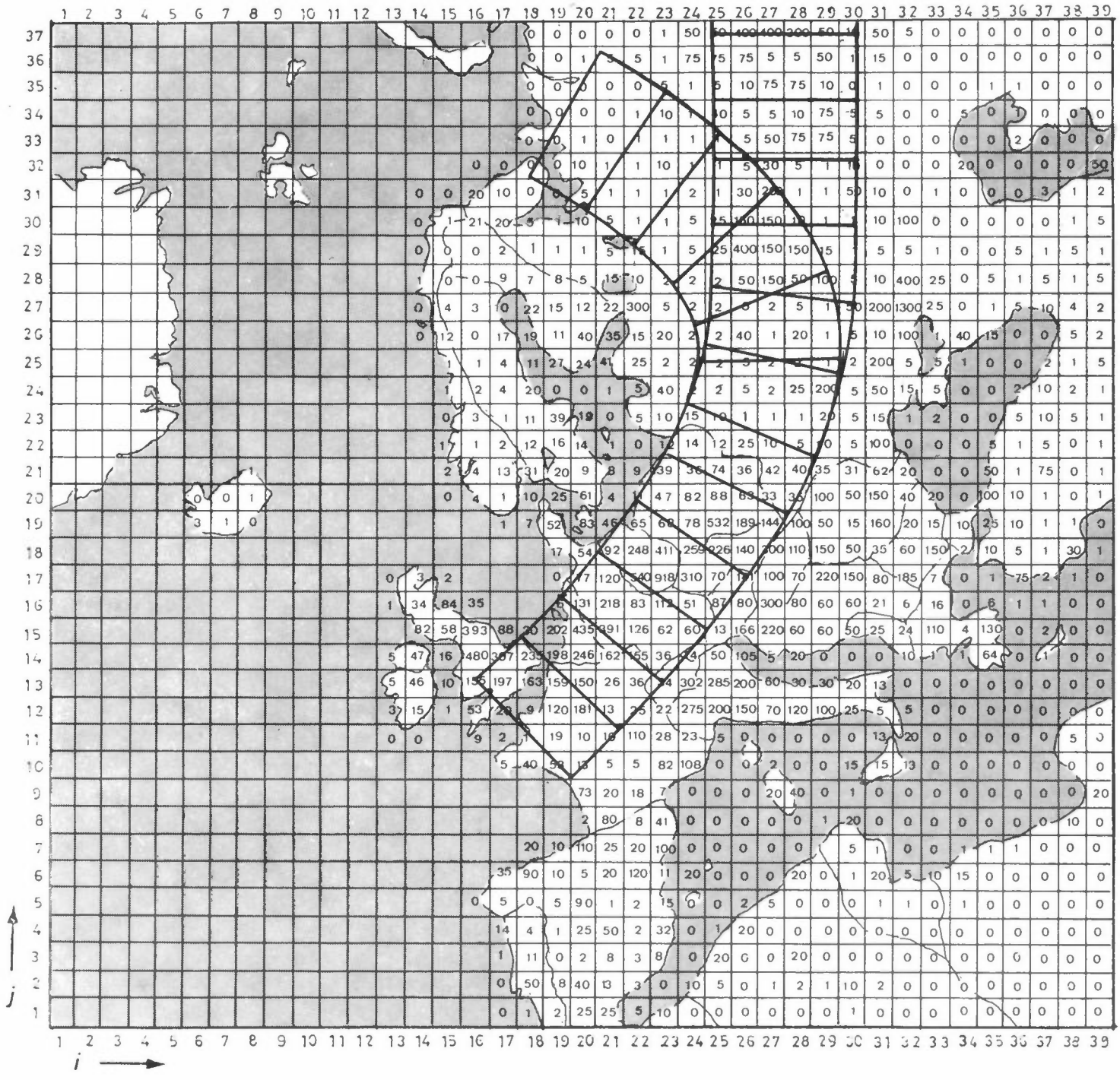

Figure 5: "Short" and "long" pathways to the Arctic via the USSR, superimposed on EMEP emission grid for SO_2.

Calculations and results

Calculations were carried out in nearly the same manner as those described by Rahn and McCaffrey (1979a), using a simple model of horizontal near-surface transport and transformation of an air mass exposed to progressive injection, conversion, dilution, and removal of sulfur compounds as it passed from Europe to the Arctic. The air parcel was somewhat arbitrarily given a N-S dimension of 800 km (both to be of synoptic dimension and to include most of the European emissions) and an E-W dimension equal to the distance it is transported in one day. It was placed near the western limits of Europe/UK and moved eastward, then northward, in discrete 1-day time steps. It was made initially free of SO_2 and $SO_4^{=}$.

During each time step it received an amount of SO_2 numerically equal to the sum of the annual emissions from the grid squares included within it (in units of 10^3 T S y^{-1}; all subsequent calculations were relative - i.e., a constant mixing height was assumed). Emissions were assumed to be exclusively SO_2. The air parcel was then transported for one day and allowed to age. New concentrations of SO_2 and $SO_4^{=}$ were calculated from the following equations:

$$(SO_2)_{n+1} = (SO_2)_n e^{-(k_{wd} + k_{ox})_n \Delta t} (D_{n+1}) + (SO_2)'_{n+1}$$

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

where

$$A_n = (k_{wd})_n + (k_{ox})_n - (k_p)_n$$

and

k_{wd} = rate constant for combined wet and dry removal of SO_2

k_{ox} = rate constant for oxidation of SO_2

k_p = rate constant for removal of sulfate

$(SO_2)'$ = emission of SO_2 into the air parcel
Δt = time step of 1 day
D = dilution factor ($D \leq 1$)
n = number of days of transport.

These formulas were derived from simple first-order equations of transformation for SO_2 and $SO_4^{=}$.

All calculations were executed in 1-day time steps, during each of which the k's remained constant. If the k's were set to vary (linearly or exponentially) during the series of calculations, they were updated daily to give a quasi-smooth change.

To take account of unequal sizes of the various boxes caused by curvature of the path, all air-parcel areas and corresponding emissions were normalized to a standard area of 800 x 346 km.

A constant transport speed of 4 m s^{-1} was used over the length of both paths. This figure was derived from calculations of Bolin and Persson (1975), which showed that the geometrical center of the endpoints of a series of 850-mb trajectories, calculated every third day from 1 October 1972 through 31 March 1973, moved ENE-ward at this rate. This velocity thus represents a mean for winter over Europe, and corresponds to 4 days travel time between London and Stockholm. Because it incorporates speeds in a variety of directions, its magnitude is lower than mean transport speeds in any given direction, which are more like 6-7 m s^{-1} at 850 mb over Europe. Calculation using 4 m s^{-1} will tend to weight sources nearer the Arctic more than those done at 6-7 m s^{-1}, and this extra weighting will be greater for species with short atmospheric residence times than for longer-lived species. For this reason, and recognizing that the case of more rapid transport of a specific air mass to the Arctic may be as important as mean transport, all calculations were repeated for 8 m s^{-1} transport speed.

The value for daily dilution was set at a constant 0.94 for the 4 m s^{-1} calculations and a constant 0.88 for the 8 m s^{-1} calculations. This parameter is surely the most poorly known of all those used in the calculations, but in spite of this we felt that some mild form of dilution had to be included both to compensate for the lack of vertical motion in the model and to recognize that some dilution, however small, must be occurring even for synoptic-scale air masses over these long distances of transport. Without dilution, distant (European) sources would be unfairly positively weighted. The dilution value of 0.94 per day was derived empirically from the factor of 1/6 after 20 days needed in the numerical simulations of Rahn and McCaffrey (1979a) to account for actual concentrations of $SO_4^=$ and V at Barrow, Alaska starting from SO_2 and V in European air masses. For a path length of 10 000 km between Europe and Barrow, a transport speed of 4 m s^{-1} would require 29 days. Assuming the same final dilution of 1/6, the dilution factor per day would then be $\sqrt[29]{1/6}$, or 0.94. For transport at 8 m s^{-1} the dilution factor per day would be $(0.94)^2$, or 0.88.

Calculations were carried out using a 3-part model of transport of polluted air masses between Europe and the Arctic, which we have been developing since May 1979 (Rahn and McCaffrey, 1979c). This model attempts to refine the calculation scheme of Rahn and McCaffrey (1979a), in which the rate constants k_p, k_{ox} and k_{wd} decreased monotonically between the sources and Barrow, Alaska. The new model is based on the variation of the aerosol removal rate constant k_p along the path Europe - European USSR - European Arctic (roughly Franz Josef Land) - Barrow (via a path between Greenland and the North Pole) which in turn is assumed to be determined mainly by the mean monthly precipitation. The rate of precipitation during summer and winter along this path defines 3 fairly distinct regions: a midlatitude zone between Europe and roughly Moscow, where the amounts are nearly the same and high, an Arctic zone north of 70-80°N (beginning at Franz Josef Land on the short path)

where the amounts are nearly the same, but several times lower than in the midlatitude zone, and a transition zone where precipitation changes rapidly between midlatitude and Arctic values (Moscow to Franz Josef Land on the short path). In this model the mean January precipitation is about 35 and 5 mm mo^{-1} for midlatitudes and Arctic, respectively, with a linear decrease with distance in the transition zone. Actual values of k_p are calculated assuming (somewhat arbitrarily) that 76 mm precipitation per month produces a residence time of 5 days (typical midlatitude values for each parameter) and that aerosol residence times ($1/k_p$) are inversely proportional to rate of precipitation and nothing else. They correspond to residence times of 11 and 76 days for midlatitudes and the Arctic, respectively. The considerable evidence for a several-fold longer residence time in the Arctic is reviewed in Rahn and McCaffrey (1979a). Rahn and McCaffrey (1979c) used this model to carry out calculations for this pathway with transit times of 5, 5 and 10 days for midlatitudes, transition zone, and Arctic, respectively; here the times used are roughly twice as long. The aerosol residence time of 5 days for midlatitudes may be too long; more attention will have to be paid to this starting point in the future.

Much less can presently be said about how the rates of oxidation and removal of SO_2 ought to vary along the path to the Arctic. Following the lead of k_p, however, k_{ox} and k_{wd} are usually given constant midlatitude values and constant but order-of-magnitude lower Arctic values, with linear decreases in the transition region. Fortunately, experience with this model has shown that results for the Arctic aerosol depend much more on the midlatitude values of rate constants than on the more poorly known Arctic rate constants.

A simplified version of this model, for the Arctic only, has been used successfully by Rahn and McCaffrey (1979c) to explain the seasonal variation of ^{210}Pb at Barrow from the seasonal variation of ^{222}Rn at 3 Arctic sites and a 7-fold seasonal variation of Arctic residence times. The complete

model failed to adequately predict the seasonal variation of V at Barrow, however. The likely reasons for this were twofold: (1) The seasonal source function for V had to be taken from the northeastern United States, where there is a January maximum. Recent evidence suggests that the European source function has a February maximum and is markedly broader then that of the northeastern United States (occupying 5 months compared to 3 for the US), but has nearly the same seasonal amplitude (factor of 3) as does the US function. (2) Lack of any time delay in the calculations to allow for the travel time between sources and the Arctic (probably close to a month for transport from Europe to Barrow). The European source function with a time delay of one month comes very close to reproducing the seasonal pattern of V at Barrow, which has a March maximum and an annual amplitude of a factor of 20 to 30.

In the calculations along the two transport paths, mid-latitude conditions were taken for the first 10 boxes, until just beyond Moscow. After that, transitional conditions to Arctic values of the rate constants were used, for 6 more boxes for the short path (until about Franz Josef Land) and for 10 more boxes for the long path (until the Taymyr Peninsula). Each path thus terminated near the local border of the Arctic, and with the same values of rate constants.

The results of the cumulative calculations are shown for the short and long paths in Figures 6 and 7, respectively, together with the course of the rate constants used. SO_2 responds rather rapidly to emissions, with peaks and valleys displaced only slightly eastward relative to the source patterns. A fairly sharp maximum is predicted to occur roughly at the border between western and eastern Europe, declining rapidly to values 2 to 2.5 times lower in the western USSR, where the emission density is correspondingly lower. OECD observations of SO_2 from western Europe confirm this picture as far as possible; the annual mean concentration field modeled by OECD gives similar predictions as well (Ottar, 1978). The effect of the USSR emissions on the SO_2 in the moving air parcel can be seen

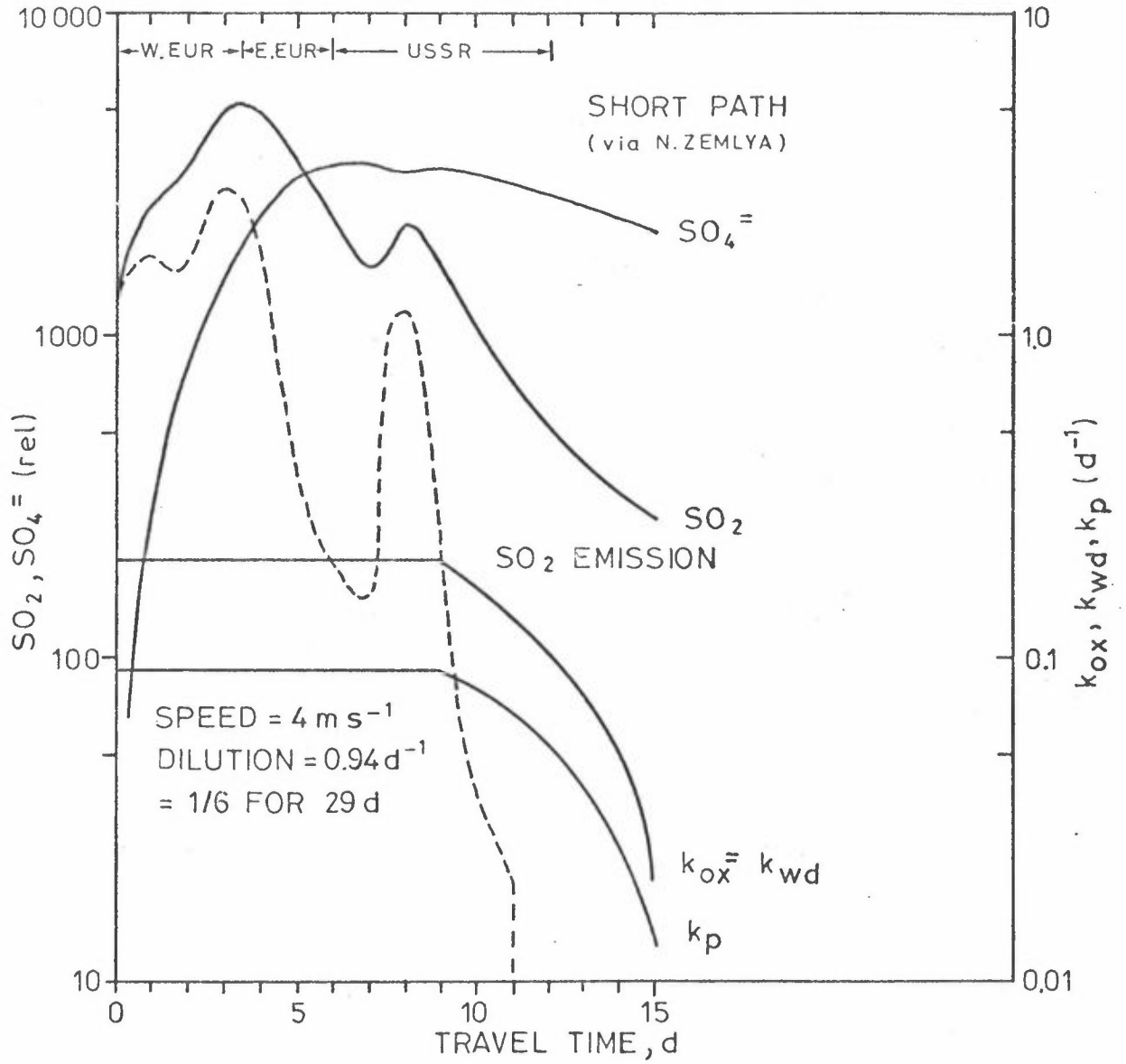

Figure 6: Simulated transformation of a polluted air mass as it travels from Europe to the Arctic via the short path.

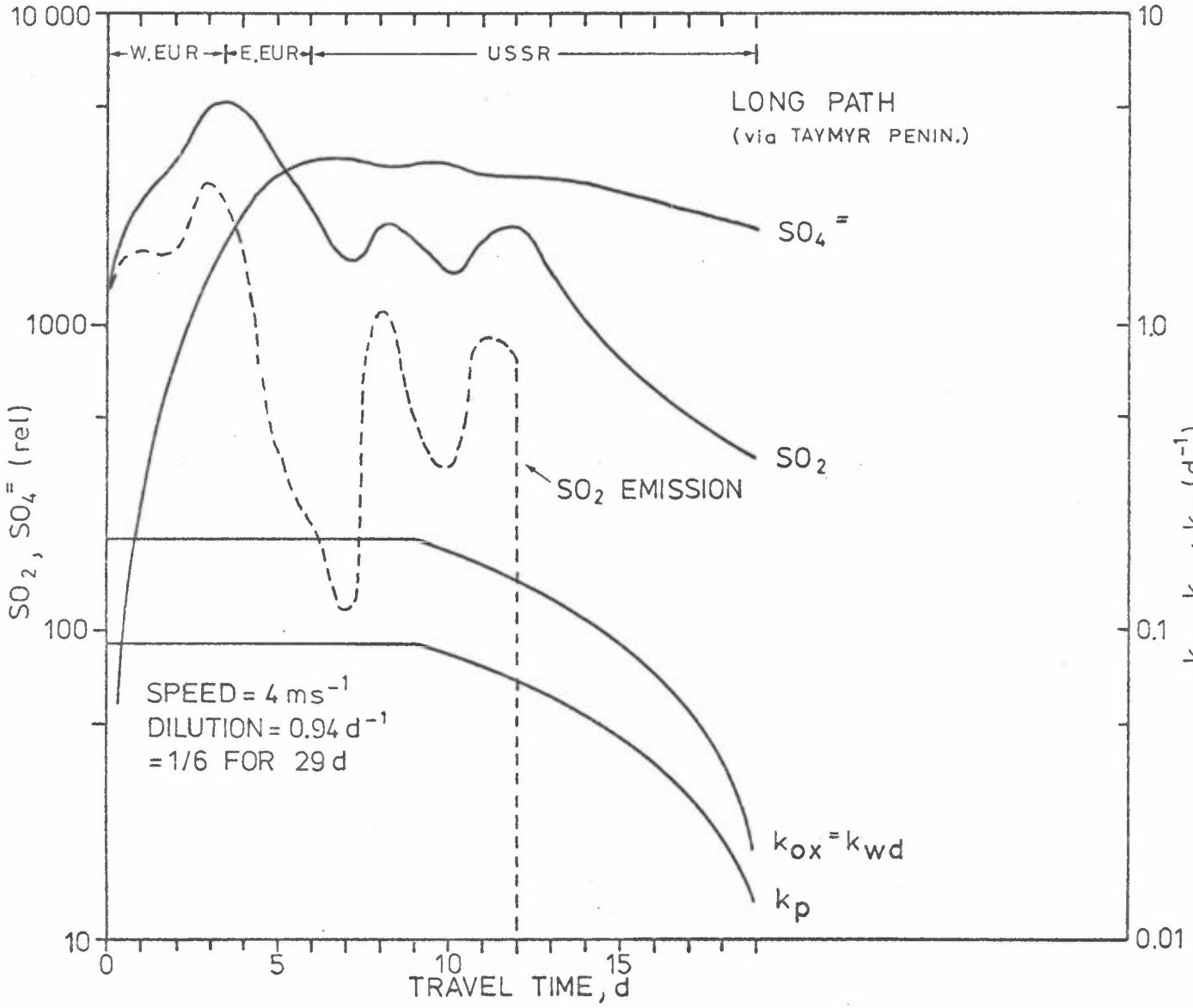

Figure 7: Simulated tranformation of a polluted air mass as it travels from Europe to the Arctic via the long path.

by extending the SO_2 trace without the USSR sources of Moscow and Sverdlovsk. For the short and long paths the decreases of SO_2 concentration are roughly 2X and 7X, respectively.

The response of $SO_4^=$ is much more sluggish, because of the time delay introduced by the oxidation from SO_2, so that the effect of the USSR sources is more difficult to estimate from these figures alone. A very broad maximum of $SO_4^=$, from eastern Europe through the entire western USSR, is predicted by these calculations. It is only partially confirmed by the annual mean observations and predictions of the OECD program (Ottar, 1978), which seems to predict a somewhat sharper maximum of $SO_4^=$ (over eastern Europe) than do the present calculations. Much of this difference may be due to differences in rate constants between the two sets of calculations. The constants used here were tailored specifically for winter conditions, and are all slower than those used by OECD. The net effect is to disperse the S more widely in our system than in theirs.

The broad maximum of $SO_4^=$ predicted here agrees well with that seen on the winter map for Europe and the Arctic (Rahn and McCaffrey, 1979c). An extension of either $SO_4^=$ trace to 10 days travel in the Arctic (roughly simulating the time needed to reach Barrow) yields a $SO_4^=$ concentration 4X lower than that of the Eurasian maximum, which agrees with actual observations of 6 $\mu g\ m^{-3}$ in Europe and 1.5 $\mu g\ m^{-3}$ at Barrow. This demonstrates that it is possible to find a set of rate constants for the atmospheric sulfur system that can make the observed $SO_4^=$ concentrations at Barrow consistent with a complex source pattern over Eurasia and 10 000-to-15 000-km transport.

There are three other interesting features of these plots that should be noted. First, the two paths give essentially identical concentrations of SO_2 and $SO_4^=$ reaching the borders of the Arctic, although at quite different locations. This effect will contribute to the surprising homogeneity of concentration of the Arctic aerosol. Second, the time lag in response of $SO_4^=$ to SO_2 sources creates on interesting out-of-phase relationship to the source pattern within the USSR, with source regions there

having gentle minima in $SO_4^=$. Third, although it is not shown here, analogous calculations indicate that the trace of V ought to be intermediate to those of SO_2 and $SO_4^=$, assuming that the source patterns of SO_2 and V are the same (which is almost certainly not the case). This supports qualtitatively the observation that the relative decrease of V towards the Arctic is greater than that of $SO_4^=$. But V is predicted by these calculations to decrease by only about double the factor for sulfate, whereas in reality it decreases by 10 times as much. This would suggest that sources later along the path are deficient in V relative to earlier sources. Industrial emission of SO_2 in the Sverdlovsk region, which should not be accompained by much V, may explain part of the low V at Barrow. Another possibility would be that the USSR as a whole, and perhaps eastern Europe as well, emits relatively less V than does western Europe. Statistics on this are not yet available, however.

In order to estimate better the contributions of each segment of the path to the final SO_2 and $SO_4^=$ concentrations at the edge of the Arctic, the emission from each box was transported individually along the rest of the path and the resulting concentration expressed as a percentage of the final concentration. The results, together with the corrected emission figures for each path, are given in Table 2 and are shown graphically in Figures 8 and 9. They are summarized by region in Table 3, both for these calculations and for similar calculations with a transport speed of 8 m s^{-1}. The range of these results should include most actual cases of interest.

These results show a number of interesting features. (1) The contribution of the USSR seems not to be negligible under any of the circumstances considered here. It can amount to 18-93%, depending on substance, pathway and transport speed. The mean contribution of the USSR for all cases treated here is 71% for SO_2 and 32% for $SO_4^=$. (2) The contribution of the USSR is greater (a) for SO_2 than for $SO_4^=$, presumably because of the shorter atmospheric residence time for SO_2, (b) for the long path than for the short path, because of additional sources at Sverdlovsk late in the long

Table 2: Corrected SO_2 emissions and fractional contribution of each box to SO_2 and $SO_4^=$ at the edge of the Arctic.

Box	Short path			Long path		
	SO_2 emission 10^3T S y^{-1}	f_{SO_2}	$f_{SO_4^=}$	SO_2 emission 10^3T S y^{-1}	f_{SO_2}	$f_{SO_4^=}$
1	1273	1.1	6.8	1273	0.3	4.6
2	1719	2.4	10.7	1719	0.6	7.2
3	1691	3.8	12.2	1691	0.9	8.2
4	2891	10.3	23.9	2891	2.4	16.3
5	1696	9.6	16.1	1696	2.3	11.1
6	370	3.3	4.0	370	0.8	2.8
7	196	2.8	2.4	222	0.7	1.9
8	155	3.5	2.1	118	0.6	1.2
9	1203	43.1	17.4	1117	9.5	12.3
10	247	14.0	3.7	473	6.4	5.8
11	38	3.4	0.6	343	7.3	4.6
12	20	2.7	0.3	903	29.6	12.7
13	-	-	-	800	38.7	11.4

f = Fractional contribution of emissions in box to final concentration of SO_2 or $SO_4^=$, in percent.

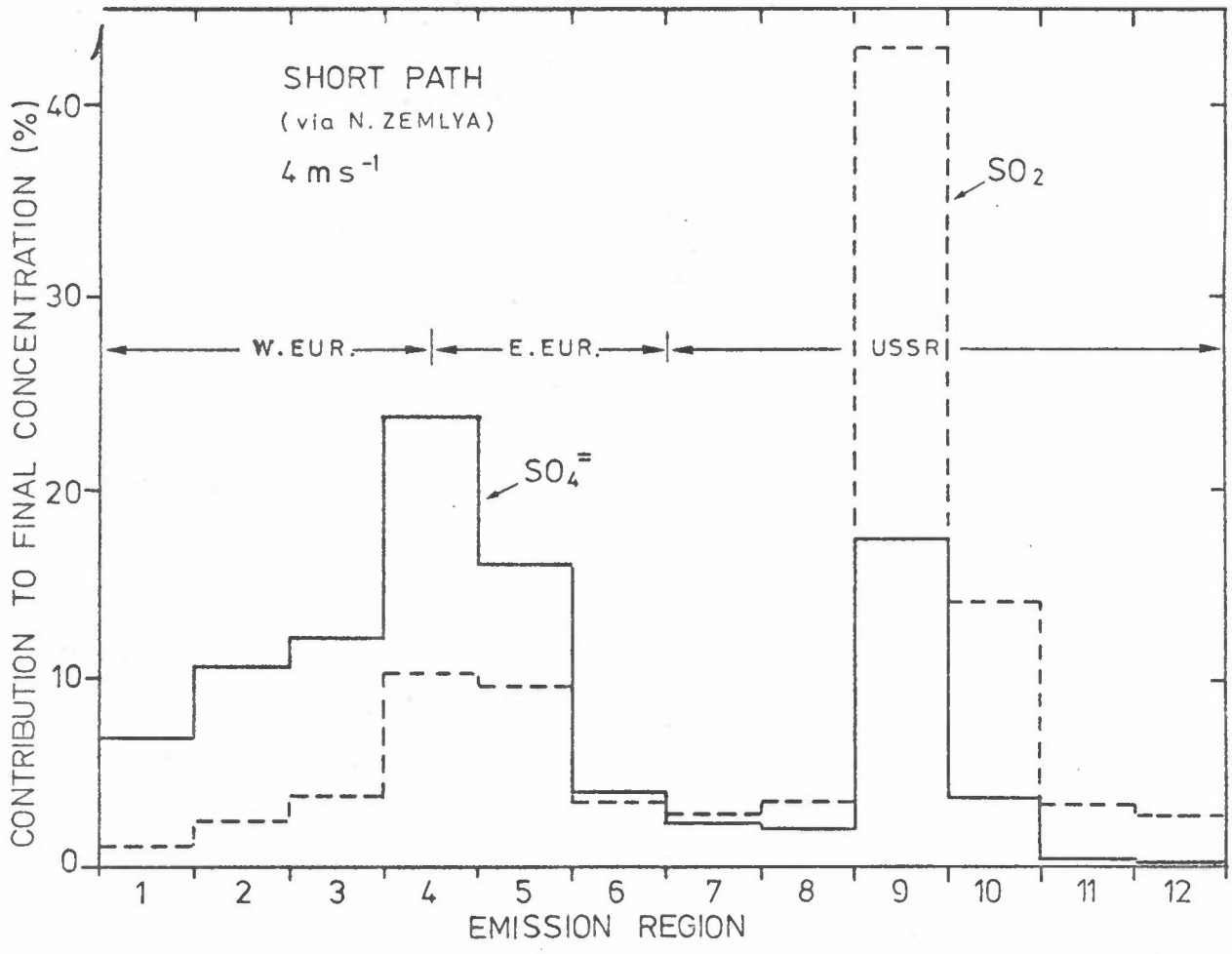

Figure 8: Fractional contribution of each box to Arctic SO_2 and $SO_4^=$, short path.

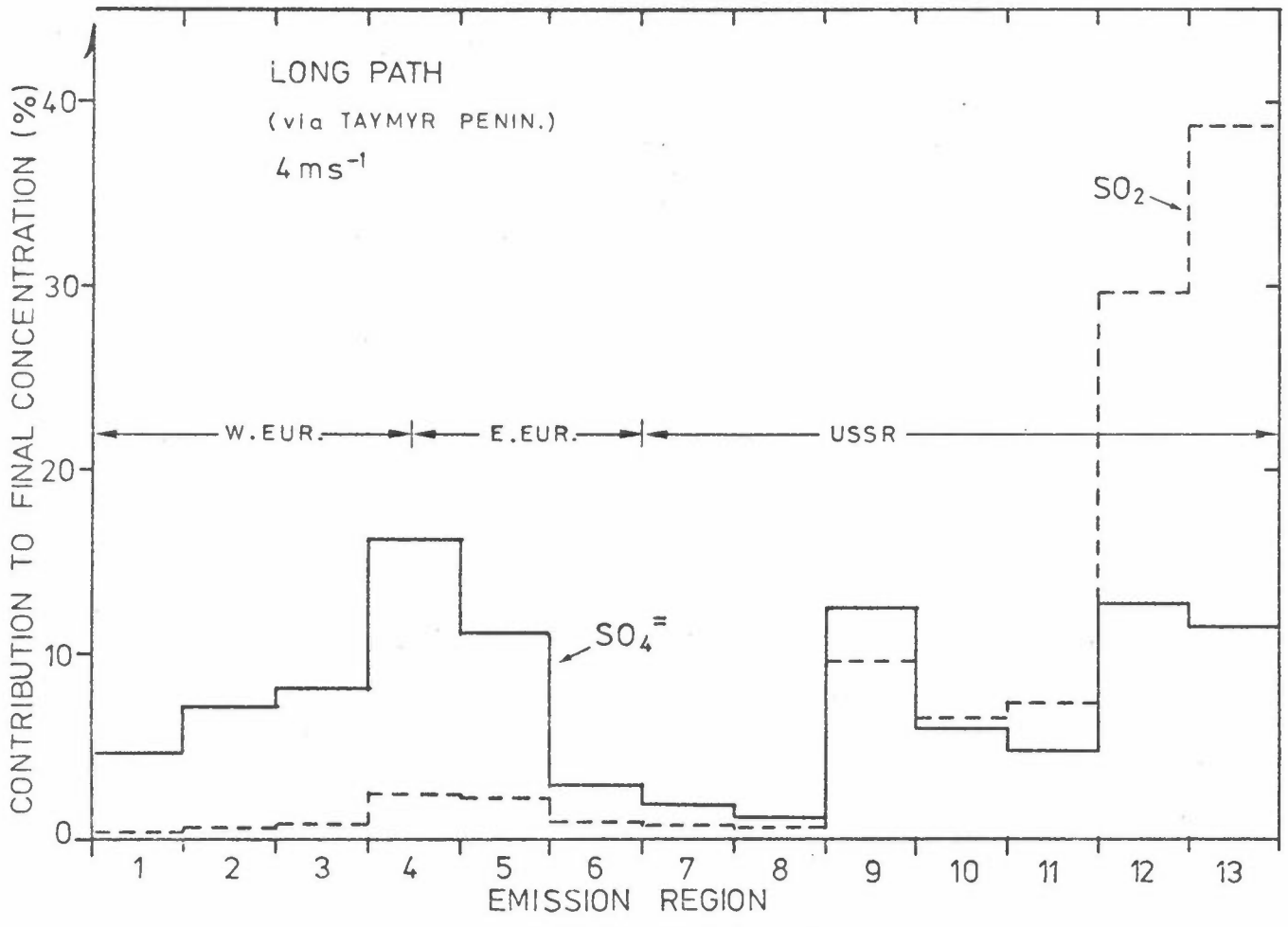

Figure 9: Fractional contribution of each box to Arctic SO_2 and $SO_4^=$, long path.

Table 3: Regional contributions to Arctic SO_2 and $SO_4^=$, percent.

		Western Europe (boxes 1,2,3, 0.5x4)	Eastern Europe (boxes 0.5x4,5,6)	USSR (boxes 7 ff)
Short path (via Novaya Zemlya)	SO_2 4 m s^{-1}	12	18	70
	8 m s^{-1}	28	25	47
	$SO_4^=$ 4 m s^{-1}	42	32	26
	8 m s^{-1}	52	30	18
Long path (via Taymyr Pen.)	SO_2 4 m s^{-1}	3	4	93
	8 m s^{-1}	13	12	75
	$SO_4^=$ 4 m s^{-1}	28	22	50
	8 m s^{-1}	40	24	36
Means and ranges	SO_2	14(3-28)	15(4-25)	71(47-93)
	$SO_4^=$	40(28-52)	27(22-32)	32(18-50)

path, (c) and for 4 m s^{-1} transport speed than for 8 m s^{-1} speed, because with slower speeds there can be greater removal of SO_2 between the major European sources and the major USSR sources. The two extreme cases are SO_2 along the long path at 4 m s^{-1} (93% from the USSR) and $SO_4^=$ along the short path at 8 m s^{-1} (18% from the USSR). (3) Other calculations not given here show that if the source pattern for V is taken to be the same as that for SO_2, and V is given the same atmospheric residence times as $SO_4^=$ along the paths, the fractional contributions of the various boxes to Arctic V are very similar to those for $SO_4^=$, being only about 20% more from the USSR than is $SO_4^=$. Increasing the separation between the European and USSR maxima of emissions increases slightly the differences in their contributions to V and $SO_4^=$ near the Arctic. This surprising similarity between the contribution to primary V and secondary $SO_4^=$ must reflect the relatively short atmospheric residence times of SO_2 (compared to the length of a box) and the relatively long atmospheric residence times of aerosol V and $SO_4^=$, so that $SO_4^=$ can be effectively emitted in nearly the same box as is V. This result, that identical source patterns of SO_2 and V produce nearly identical fractional contributions to Arctic $SO_4^=$ and V, is very important because it allows us to deduce something of the nature of the V source pattern relative to that of SO_2 from $SO_4^=/V$ ratios in the Arctic and environs. Because of the relative insensitivity of final $SO_4^=/V$ ratios to path length or direction, this ratio appears to be mostly fixed by the SO_2/V ratio of the sources to which the air parcel has been exposed (on the mean, of course, and within a given season, when systematic differences in rate of oxidation of SO_2 can be neglected). Thus, short-term differences in $SO_4^=/V$ ratio in the Arctic seem to indicate different sources more than different path lengths or speeds of transport. Furthermore, differences in sources later in the path (i.e., within the USSR) will affect the final ratios more than differences in sources early in the path (i.e., within Europe). Thus, to explain major short-term variations in $SO_4^=/V$ ratios at an Arctic site, say Bear Island, it now seems most reasonable to look first for differences in path that could have brought the air parcel over different types of pollution sources. There

is evidence that such source-related differences are in fact seen at Bear Island; they will be examined more in the future.

One final note: longer-term (i.e., seasonal) systematic variations of trajectory to the Arctic, and hence the pattern of effective SO_2 and V sources, may well exist and affect the seasonal $SO_4^=/V$ ratios. This effect is harder to isolate than the shorter-term variations, though, because it is mixed with the seasonal variations of rate of oxidation of SO_2. There is some indirect evidence that the threefold increase of the $SO_4^=/V$ ratio at Barrow and Bear Island in summer may be too large to be explained by seasonal conversion to $SO_4^=$ alone. If this should indeed be the case, systematic V-poor pollution sources may be required.

Conclusions and implications

Several main conclusions and implications emerge from the above calculations and interpretations: (1) The USSR appears to be an important source of Arctic sulfate (20 to 50%) and of Arctic SO_2 (50 to 90%) during winter. Future models of generation and transport of Arctic aerosol will thus have to include the USSR. (2) The calculated importance of the USSR needs to be verified experimentally. The most direct way to do this would be with a series of 3 to 5 measuring stations between Novaya Zemlya and the New Siberian Islands, spaced so as to coincide with the various maxima and minima of transport proposed here. Such stations would logically be cooperative projects with the USSR. If this is not possible, shorter-term experiments in adjacent areas, such as manned ice islands, would be an alternative. Highest priority, though, should be given to establishing cooperative projects with the USSR. (3) For Arctic-oriented trajectory calculations, the present EMEP grid may have to be expanded eastward, if the SO_2 sources near Sverdlovsk and the path over the Taymyr Peninsula are as important as they seem to be at present. This would also require an inventory of emissions for the new portion of the grid. Such an expanded grid would probably also be useful for interpreting EMEP data from northern

Scandinavia, which should be subject to the same return-flow episodes that affect Bear Island. Before the large task of expanding the grid is undertaken, however, a preliminary study of return-flow trajectories to Bear Island and Jergul should be carried out in order to verify that important trajectories do indeed pass eastward of the present grid. (4) Because of the apparent importance and usefulness of the $SO_4^{=}/V$ ratio in Arctic air chemistry, an emissions inventory of V in Europe and the USSR is needed. (5) Further attention should also be given to the Sverdlovsk area and its emissions, in order to document better its effect on the Arctic aerosol.

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ABSTRACT (max. 300 characters, 5-10 lines)

There is now considerable evidence that Eurasia is the main source region for the pollution component of the winter Arctic aerosol, and that one of the major transport paths is from Europe eastward into the western USSR, then northward into the Arctic. Under these conditions, gases and aerosols emitted in the western USSR will contribute to the overall burden of pollution reaching the Arctic. This report attempts to evaluate the relative contribtions of Western Europe, Eastern Europe and the USSR to Arctic SO_2 and $SO_4^=$ for two possible pathways through the western USSR, by means of numerially simulated aging of air manes traversing these paths and emission of SO_2 according to the recent EMEP survey. The results indicate that on the mean, the USSR may contribute 47-93% of Arctic SO_2 and 18-50% of Arctic $SO_4^=$, depending on pathway and transport speed.

**Kategorier: Åpen - kan bestilles fra NILU A
Må bestilles gjennom oppdragsgiver B
Kan ikke utleveres C