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BLACK AND WHITE EPISODES, CHEMICAL EVOLUTION OF EURASIAN AIR MASSES, AND LONG-RANGE TRANSPORT OF CARBON TO THE ARCTIC.

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ABSTRACT

Long-range transport of pollution aerosol from central Europe to southern Scandinavia has been recognized for a decade. Depending on the trajectory, high concentrations of sulfate in southern Sweden are accompanied by greater or lesser carbon, and have become known as black or white episodes. It is now known that, when conditions are right, pollution aerosol (and elemental carbon) can be transported order-of-magnitude longer distances, from Eurasia to the Arctic. A simple model of long-range transport of SO_2, $SO_4^=$, and trace metals has been constructed to account for the large changes between Eurasia and the Arctic. This paper presents a broadened interpretation of black and white episodes which, in combination with atmospheric measurements at other locations in Eurasia and environs, allows this transport model to be checked empirically. It also allows the atmospheric lifetime of carbon to be compared with lifetimes of other primary aerosol pollutants. The results confirm most features of the model, and suggest that black and eastern white episodes are highly related, early stages of systematic atmospheric aging, later stages of which can be observed as far away as the Arctic. The lifetime of elemental carbon appears to be similar to that of other primary, submicron elements, but is still very uncertain.

Transport of air pollutants to Scandinavia as black and white episodes

Transport of polluted air masses from the highly industrialized regions of central Europe to Scandinavia has been known for over a decade [1-3, for example]. This transport is highly episodic, with each episode having high concentrations of submicron aerosol particles (including the associated $SO_4^=$, H^+, etc.) and sometimes high concentrations of pollution gases such as SO_2 and NO_2. Episodes occur when air moves directly from European source regions to Scandinavia. The major environmental effects, which have been the driving force for the European OECD/LRTAP studies, are the acidification of precipitation and its effects on the environment [1,4]. Changes in SO_2 and $SO_4^=$ during transport have been extensively measured and modeled by the OECD/LRTAP program [3,5].

Although the greatest attention has been paid to the various forms of sulfur being transported, an entire suite of aerosols and gases is carried to Scandinavia as well. Important constituents of the aerosol include Fe, Mn, V, Pb, Cd, etc., and, of course, elemental C. Although the latter may have natural sources such as forest fires, these sources should not be important during winter, when most of the pollution episodes occur in Scandinavia. Elemental C, or more properly the darkness which it imparts to samples of aerosol, has long been paid special attention in urban studies because it can be measured rapidly, inexpensively and reproducibly. Typically, the mass of the aerosol in a given region is estimated from the grayness of a filter, by using empirical calibration curves for that region which relate grayness to the total mass of aerosol. An example of this procedure is the OECD technique [6], described by Brosset and Åkerström [7], which uses reflectance measurements on filters to estimate their grayness. Unfortunately, total masses derived in this way are often referred to as "soot", a historical misnomer which can be confused with the

modern meanings of "soot" (graphitic C with or without the other mass closely associated with it [8]) unless clearly specified.

Soot was rarely measured in remote regions, however. Some of the first investigations of this type were those of Brosset and co-workers, who have considered both soot and sulfate in Scandinavia since the earliest studies of long-range transport to that region. Brosset and Nyberg [2] found that concentrations of each were linearly related in winter episodes, those which Rodhe et al. [9] and Nordø et al. [10] later showed to be associated with air flow from central Europe. Those episodes were also accompanied by high concentrations of nitrate and metals such as Mn, Fe and V. The particles were of moderate acidity, with much or most of the H^+ neutralized by NH_3. Occasionally, high concentrations of SO_2 were also observed [11, 12].

Subsequent measurements by Brosset revealed, however, that similarly high concentrations of sulfate could be accompanied by drastically reduced soot when air masses were transported eastward to Sweden from over the North Sea. Such episodes, which occured primarily during summer, were termed "white episodes" by Brosset, to distinguish them from the "black episodes" of winter [11]. The white episodes were also characterized by much higher concentrations of hydrogen ions and much lower concentrations of metals and nitrate than were found in black episodes. Although the source of the sulfate, soot, etc. of white episodes could not be documented with certainty because of long over-water trajectories, it was presumed to be Europe as well.

Later, another degree of complexity was added to black and white episodes when Brosset discovered that white episodes could occur with transport from the east as well as with transport from the west [12]. Properties of particles in the eastern white episodes were nearly the same as

those in western white episodes. Interestingly, eastern white episodes were not found during summer; the first cases were observed during February and March (1976). Trajectory analysis by the Norwegian Institute for Air Research showed that air masses of eastern white episodes had passed over eastern Europe or the western USSR some days earlier.

It should be noted that Brosset's classification of episodes as black or white was never intended to be rigid. Rather, the two types of episodes were deemed only to represent particles of "essentially different genetic origin", and the existence of transition stages was recognized [12]. This point is important for the discussion below.

Transport of carbon and other pollutants from Eurasia to the Arctic

Recently, a series of studies has demonstrated that aerosols and gases are transported from Eurasia to the Arctic regularly during the winter half-year [13-19]. Constituents to which the most attention has been paid include SO_2, sulfate, V and Mn. But the Arctic aerosol of winter also contains large amounts of sooty carbon, to the point that all filter samples are noticeably gray. This was a surprise, because the aerosols of other comparably remote regions such as the equatorial Atlantic and Pacific Oceans, Mauna Loa, and the Antarctic, are essentially colorless. (By contrast, summer samples in the North American Arctic are nearly colorless, and those from the Norwegian Arctic are only light gray.) Winter transport to the Arctic is controlled near the surface by the Icelandic low-pressure and Asiatic high-pressure systems and is episodic, as is that from central Europe to Scandinavia. At 5,000 to 10,000 km, it is the longest routine transport of pollution aerosol presently known.

One of the outstanding chemical features of the Arctic aerosol is its high secondary/primary ratio relative to near-source aerosol. For example, the winter sulfate/V ratio in the Arctic is about 3×10^3, a full

order of magnitude higher than the values of 0.3×10^3 found in regional eastern North America and Eurasia during winter [14]. Because the winter Arctic aerosol appears to be derived in large measure from midlatitude polluted areas such as eastern North America or Eurasia, study of it and its precursors offers an ideal opportunity to observe systematic chemical and physical evolution of polluted air masses on a scale not previously possible. Physical properties of interest include the particle-size distribution of the aerosol as well as its scattering and absorption of light. Chemical properties of interest include particle/gas ratios such as $SO_4^=/SO_2$, secondary/primary ratios within the aerosol such as $SO_4^=/V$, and primary/primary ratios within the aerosol, such as Mn/V, for elements of different particle sizes. Of particular interest for this paper is the behavior of elemental carbon relative to other primary submicron elements such as Mn and V.

A simple description of certain systematic chemical changes during transport from Eurasia to Barrow, Alaska has been offered by Rahn and McCaffrey [14] . It starts with the mean chemical properties of polluted air masses of central Europe during winter, which are assumed to reasonably represent the true sources of Arctic pollution. These air masses are then allowed to age for times up to 20 days, which should be long enough to include all reasonably direct transport to the Arctic. During this time, gases are allowed to react, particles and gases are removed, and the air masses are steadily diluted by external air with or without aerosol or trace gases. Smoothly varying rate constants for removal, oxidation and dilution are used. Calculations are carried out in one-day time steps.

An example of the results of Rahn and McCaffrey [14] is shown in Figure 1. The k's are rate constants for removal of particles, oxidation of SO_2, and wet and dry removal of SO_2, in units of d^{-1}. These results

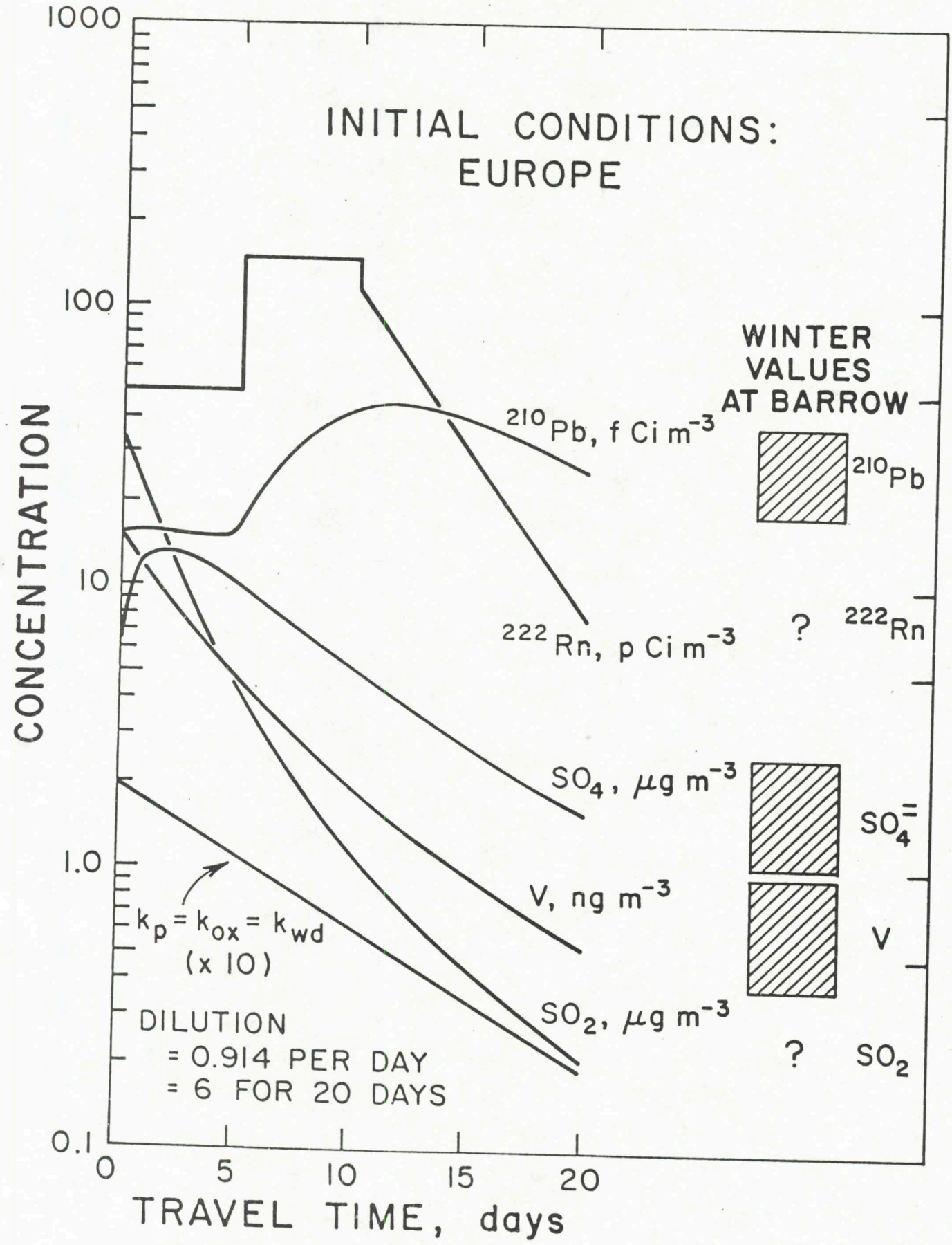

Figure 1. Simulated aging of a polluted air mass as it travels from Europe to the Arctic during winter (after Rahn and McCaffrey [14]).

showed that the aerosol at Barrow was quantitatively compatible with a European precursor, provided that rate constants decreased by roughly an order of magnitude during transport and that large-scale dilution was only a factor of 6. The shapes of the curves had a number of interesting features, such as rapid changes during the first days followed by slower changes, and a maximum in the secondary sulfate after 2-3 days. These features, which were consequences of the particular forms of evolution chosen for the rate constants, seemed generally reasonable, because polluted air masses ought to be most reactive when they are youngest and have highest concentrations of reactants such as SO_2, and because SO_2 is known to be dry-deposited rapidly near its source, particularly in summer [20]. Eurasian air moving northward during winter should be no exception. The Arctic during winter is cold, dark and dry, and may inhibit later transformation and removal in polluted air masses to an unusually great degree [14, 15]. In addition, there are both theoretical reasons and observational evidence that the rate of dilution of polluted air masses by surrounding air decreases with time [3, 21].

It would be highly desirable to check the aging model of Rahn and McCaffrey empirically. The most direct way to do this would be through a Lagrangian experiment in which Eurasian air masses were actually followed on their way to the Arctic. No such experiment has been performed, however, nor is it likely to be undertaken in the near future because of the high costs and logistical and political complexities involved. Failing this, a pseudo-Lagrangian approach may be taken, in which atmospheric measurements taken at different sites downwind of different source regions within Eurasia are compared. Such an approach is possible provided the ages of each aerosol are known and the various parent air masses are similar. The success of such a venture can never be predicted theoretically,

because it is not known whether the parent air masses within Europe are sufficiently similar or whether the ages of the aerosols can be deduced with sufficient accuracy. The only way to test these assumptions is to perform the experiment and look for coherence of results. This paper presents such an experiment and evaluates its results.

At the time that Rahn and McCaffrey [14] was written (late 1978), it was not possible to attempt to verify Figure 1 with a pseudo-Lagrangian approach, because atmospheric data were available only for the initial and final points of the trajectory. Somewhat later, data from northern Norway and the Norwegian Arctic became available, but they were judged to represent conditions after the critical first few days when the most rapid changes occurred. New data, from earlier stages of aging, i.e., nearer the source, were needed to complete the picture.

Black and white episodes as related to a simplified theory of atmospheric aging

We have recently become aware that black and white episodes, as observed by Brosset and co-workers at Onsala in southern Sweden, can provide the important missing data on the first days of aging of Eurasian air masses. In combination with data from northern Norway, the Norwegian Arctic, Barrow, and source regions in Eurasia, they offer a chance to observe details of the progressive aging of Eurasian air masses on the time scale of 0-20 days, and hence to check the entire simulations of Rahn and McCaffrey [14]. The result is the first verified model of large-scale chemical changes during true long-range transport of polluted air masses. Exposition of this idea, together with its consequences and implications, forms the rest of this paper.

The key point in the development of our new approach was a broader interpretation of black and white episodes that recognized and stressed their basic similarities rather than the more obvious differences that

have been discussed in the literature to date. Black and white episodes are surprisingly similar in several ways: (1) the sulfate/soot and sulfate/Mn ratios, which are secondary/primary ratios that have been used to distinguish black from white episodes, are not always so different between the two types of episodes as one might imagine. (Here we restrict ourselves to eastern white episodes because they occur during winter, the season of maximum transport to the Arctic, and have more easily defined source areas than do western white episodes.) Data from Brosset [12] show that there is typically a factor-of-two range of each ratio within a given type of episode, whereas mean sulfate/soot ratios are a factor of three different between the types of episodes and sulfate/Mn ratios are only a factor of two different. In fact, sulfate/Mn ratios overlap considerably between the two types of episodes. Thus, the secondary/primary ratios of black and eastern white episodes differ from one another by a factor of three at most; the variation within each class is the same order as the separation between classes. (2) A factor-of-three change in the secondary/primary ratio sulfate/V is easily predicted from Figure 1 to occur during the first days of aging, when the rate constants are varying smoothly. (3) Black and white episodes can occur at the same site during the same season (spring). Sometimes they can even come right after one another. For example, during February 1976, episodes of the two types alternated regularly, sometimes occurring only one day apart [12]. Brosset [12], in his Table 11, reports four such alternations between 14-24 February 1976. (4) Meteorological maps for 14-24 February 1976 reveal surprisingly similar placements of synoptic systems for the two types of episodes. (5) Air-mass trajectories for the two types of episodes at Onsala [12] are not so different in length. Both come rather directly from heavily polluted areas: central Europe to the south for black episodes

and eastern Europe - western USSR for eastern white episodes. (6) Black and eastern white episodes must be traceable back to a similar polluted precursor. Because each type of episode has about the same final concentration of sulfate (15-20 $\mu g\ m^{-3}$), each must have started with approximately the same concentration of SO_2, and thus roughly the same concentrations of other primary pollutants such as Mn, Fe, etc. (7) White episodes may be derived from black episodes, because the initial conditions for a white episode, 10-15 $\mu g\ m^{-3}$ SO_2 and low concentrations of primary particles [12], are characteristic of a partially aged polluted air mass, which could have been a black episode or something related to it.

In view of these considerable similarities between black and eastern white episodes, we have adopted the following working hypothesis: Black and eastern white episodes represent progressive stages of a unified process of atmospheric aging of similar parent air masses, although from different regions within Eurasia. The main variable causing the differences between black and eastern white episodes is time, or extent of aging; this alone is enough to explain many of their apparently unrelated features. According to this view, there is a natural progression in the blackness of a pollution aerosol relative to its total mass. In the beginning, near the source, primary constituents like soot are of major importance, and the aerosol is very black. As the aerosol ages, these primary constituents decrease in concentration monotonically, whereas secondary substances like sulfate increase monotonically relative to the primary constituents, and may even pass through a maximum in absolute concentration. Thus the relative blackness of an aerosol decreases steadily during aging.

Here we wish to acknowledge that relating black and eastern white episodes to nothing more than time, or extent of aging, is an obvious oversimplification. It is, however, deliberate on our part, an attempt to

investigate just how little is really required to explain the essence of black and white episodes, namely the sulfate/soot or secondary/primary ratio. The goal of this paper is to create a first level of explanation only, by keeping the number of variables as small as possible. In this way, we hope to reveal the fundamental process of atmospheric aging and its most basic results. The results, discussed below, suggest that this approach has merit. In the future, after the successses and shortcomings of this first approach have been treated, a second, somewhat more sophisticated, approach will be taken to explain the remaining properties of black and white episodes by this basic theory and additional variables.

An empirical, pseudo-Lagrangian aging diagram for Eurasian air masses

It is not difficult to assign approximate times of aging to black and eastern white episodes. The presumed source for black episodes is central Europe. From southern Sweden to central Europe is 700-800 km, or roughly 1 day's travel at 6-7 m s^{-1}, the speed commonly cited for long-range transport in Europe [3]. The source for eastern white episodes is presumably eastern Europe and the western USSR. From southern Sweden to major pollution sources in the western USSR is roughly 1500 km, or about double the distance to central Europe. Because of the slower travel speeds expected for trajectories from the east, which are more associated with smaller pressure gradients near high-pressure areas, we take a travel time of three days from these eastern sources. This figure is confirmed by the 850-mb trajectories shown by Brosset [12], which take three days to arrive in southern Sweden from eastern source areas. To complete the picture, we assign approximate travel times of 5 days to northern Norway (Skoganvarre and Jergul) under normal "direct" northward flow, and 10 days to the Norwegian Arctic (Spitsbergen and Bear Island) via the "return-flow" pathway [15]. We thus have data for Eurasian air masses at aging times of 0 days

(central Europe), 1 day (Onsala, black episodes), 3 days (Onsala, eastern white episodes), 5 days (northern Norway), 10 days (Norwegian Arctic), and 20 days (Barrow). Figure 2 shows the sources, the measurement sites, and the presumed transport paths. We stress that the transport times to northern Scandinavia and the Arctic are still imperfectly known, and will probably be adjusted in the future. The values used here are first approximations, for demonstration purposes only.

The available data on SO_2, $SO_4^=$, C, V and Mn for these six sites have been plotted as an empirical, pseudo-Lagrangian aging diagram in Figure 3. Several remarks about the construction of this figure are in order. Nonmarine sulfate was determined from total sulfate, whenever possible, by using Na to subtract marine sulfate, according to the following formula:

$$SO_4^=_{nonmarine} = SO_4^=_{total} - Na\left(\frac{SO_4^=}{Na}\right)_{seawater}$$

Similarly, crustal Mn and V have been removed by calculating them from Al:

$$Mn_{noncrustal} = Mn_{total} - Al\left(\frac{V}{Al}\right)crust$$

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

The noncrustal Mn and noncrustal V thus calculated will both be approximately submicron, and so can be compared reliably to the submicron nonmarine $SO_4^=$.

We attempted to normalize the entire plot to the maximum mean 24-h concentrations found at each site during episodes. This was made difficult, and probably only partially successful, by the different sampling times at the different sites, which ranged from one day to one week. When 24-h data were available, typical winter maxima were chosen. When only longer-period data were available, a factor was determined which related these maxima to 24-h maxima. For central Europe, winter mean values were

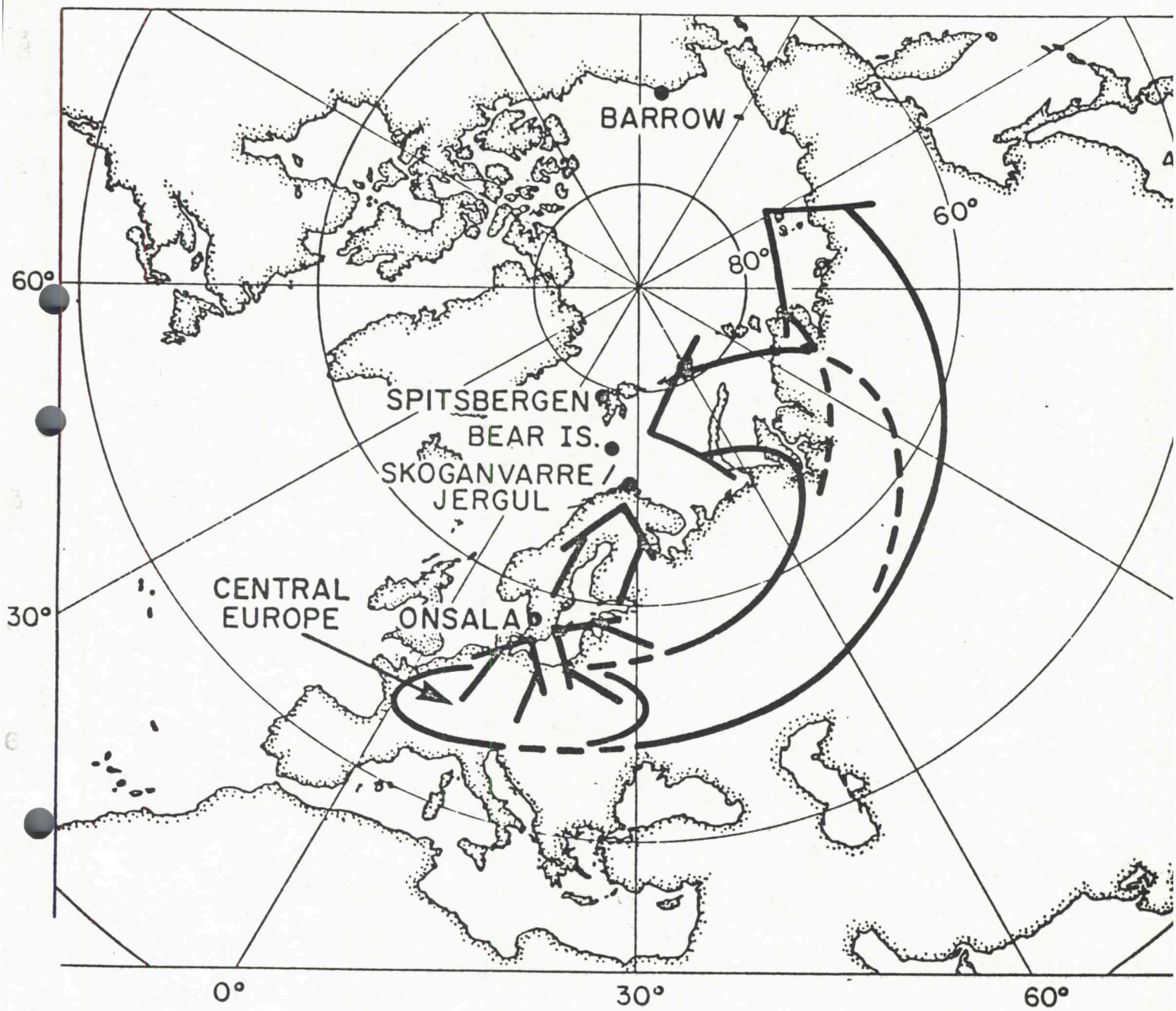

Figure 2. Locations of measurement sites for aged air masses discussed in this paper, and principal atmospheric pathways from Eurasian source regions to each site.

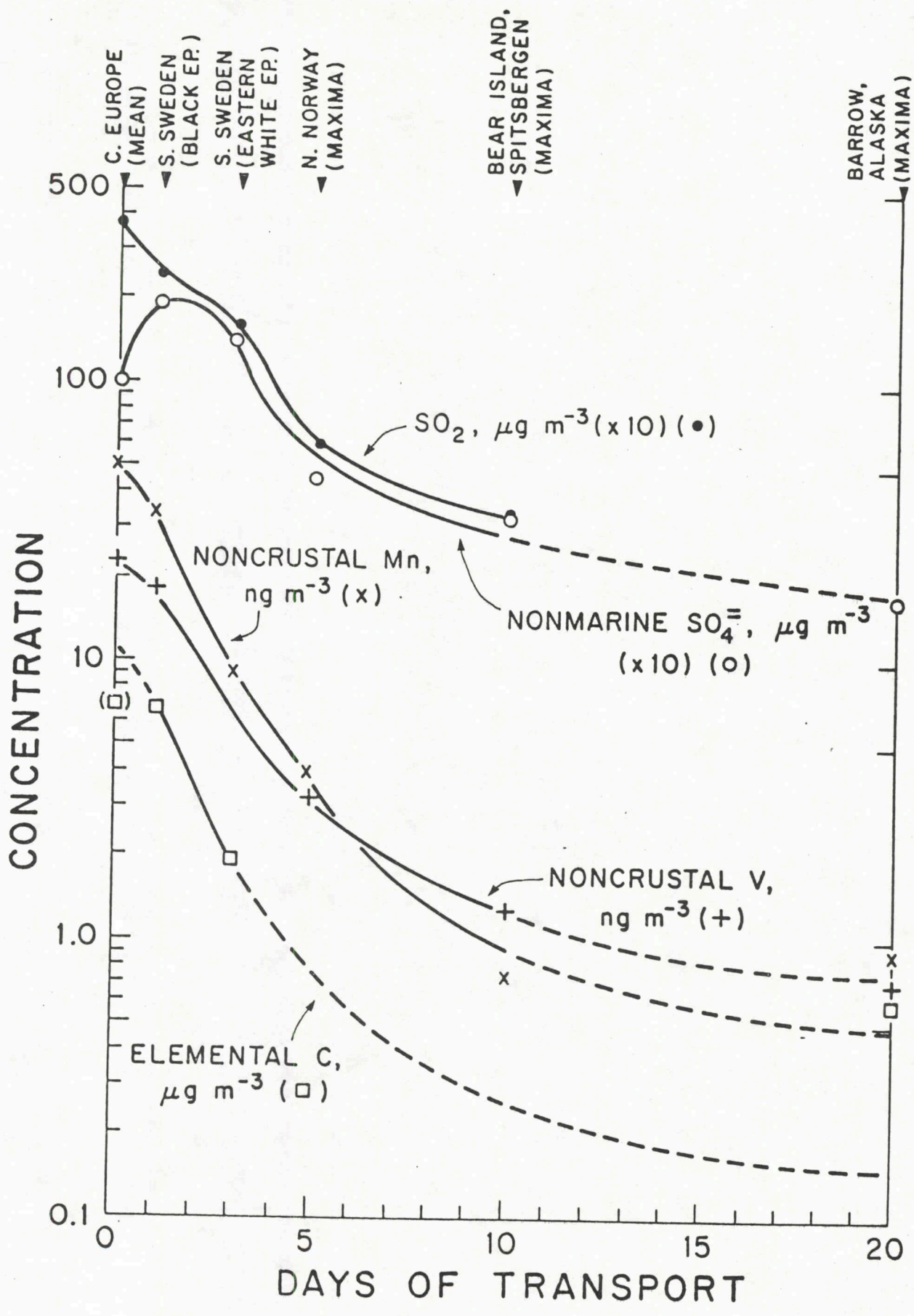

Figure 3. Empirical, pseudo-Lagrangian aging diagram for polluted air masses travelling between Eurasia and the Arctic during winter.

used because we could not assume that transport, especially to the Arctic, was automatically preceded by higher-than-normal concentrations at the source.

Elemental carbon for black and eastern white episodes was derived by multiplying Brosset's "soot" data for 1975-76 [12] by 0.2. This factor represents the approximate mass fraction of carbon in the fine-particle mode of typical urban aerosols, and was derived from data on elemental C in the New York aerosol of the late 1970's [22] and the mass of the accumulation mode there during summer 1976 [23]. The corresponding ratio in Charleston, West Virginia, which is in an area generally richer in sulfate than is New York, was 0.13 in summer 1976 [24]. The value for elemental C in central Europe, which is only an estimate, was derived by scaling the annual concentrations in New York City to European regional winter conditions. The scaling factors were 1/3 to regional, 1.3 to winter, and 37/16 to Europe (based on SO_2 in Europe and North America). The concentration of elemental carbon at Barrow was derived from broad-band transmittance measurements of filters taken during March 1978, by converting absorption to mass of C, March data to winter data, and finally winter data to typical episodic values.

Other data for central Europe came from the ECE EMEP study for 1977-78 [25] (sulfate and SO_2) and from Hoste et al. [26] (Mn,V of Belgium in 1972-73). Data for black and eastern white episodes came from Brosset [11, 12], supplemented by data from the Swedish EMEP station at Rörvik [25]. Data for northern Norway in 1971-72 came from Skoganvarre (K.A. Rahn, unpublished), and from the EMEP site at Jergul during 1977-78 [25]. Data for Bear Island and Spitsbergen in 1977-78 came from the Norwegian Institute for Air Research and from the University of Rhode Island. Barrow data of 1977-78 came from studies by the University of Rhode Island.

In spite of the large number of assumptions and approximations used to construct Figure 3, it presents a coherent, interpretable picture of atmospheric aging. It is surprisingly similar to the simulations of Rahn and McCaffrey [14] shown in Figure 1, and reproduces their basic features such as initially rapid decreases followed by a slackening in the rate of decline, the greatest changes in composition within the first 4-5 days, and a maximum in the sulfate curve. We first considered that this sulfate maximum might be an artifact, but the OECD Lagrangian calculations for individual days show a maximum for sulfate about 800 km downwind of its main sources [3], roughly the distance that Onsala is downwind from central Europe during black episodes. There are some differences from the simulations, though: the peak in $SO_4^=$ occurs earlier, SO_2 parallels $SO_4^=$ and the rest of the aerosol after 3-4 days (it becomes more nearly inert than in the simulations), and the curves are flatter in the last 10 days than expected (less dilution or shorter travel distances than expected). New information from Figure 3 is that Mn decreases faster than V (probably an effect of the former's somewhat larger particle size), and that elemental C has a larger particle size than V and/or is somehow associated with Mn in the aerosol. Few data on the particle size of C are available to compare with V, other than experience with cascade impactors that C is generally submicron. Two pieces of evidence support the association of C and Mn in the aerosol: (1) their probable common source in metallurgy, and (2) the study of Linton et al. [27], who showed that Mn is preferentially volatilized during combustion, then condenses in cooler temperatures onto the surface of pre-existing particles, such as fly ash or soot.

The progressive primary-to-secondary transformation of the aerosol can be seen from the various ratios plotted in Figure 4. Note that the $SO_4^=/SO_2$ ratio rises more rapidly initially than does the $SO_4^=/Mn$, $SO_4^=/V$, or $SO_4^=/C$ ratio. This may be an indication of early, rapid dry removal of SO_2 near its source. Depending on the particular indicators chosen, the secondary/primary ratio of the aerosol increases by factors of 4 to 20 during aging. The approximate factor-of-three increase from black to eastern white episodes is compatible with that predicted from Figure 1. The V/Mn ratio increases by about a factor of 4 between Eurasia and the Norwegian Arctic, in the same direction but larger than the factor of 2 predicted by Rahn [18] if particles down to radius 0.3 μm were removed. The reason for this discrepancy is not yet clear.

Two other interesting features of Figures 3 and 4 are the excesses of Mn (2x) and elemental C (4x) at Barrow compared to values extrapolated from the other points. The Mn at Barrow is actually higher in concentration than it is in the Norwegian Arctic. This excess Mn at Barrow has been noted by Rahn [18]; it appears as well at Mould Bay, NWT, also in the North American Arctic [17]. The North American Arctic would thus seem to be influenced at least partially by an additional source of aerosol, probably in the central USSR, which seems to emit enriched Mn, C, and probably other elements as well.

Conclusions

A number of basic conclusions are suggested by the above analysis: (1) Large-scale aging of polluted air masses can be observed in and around Eurasia during winter, once a small set of principal pathways is understood. The regularity of the empirical pseudo-Lagrangian aging plot (Figure 3) and the similarity to the simulated aging (Figure 1) suggest that aerosols at different aging times have had common precursors, and were produced by

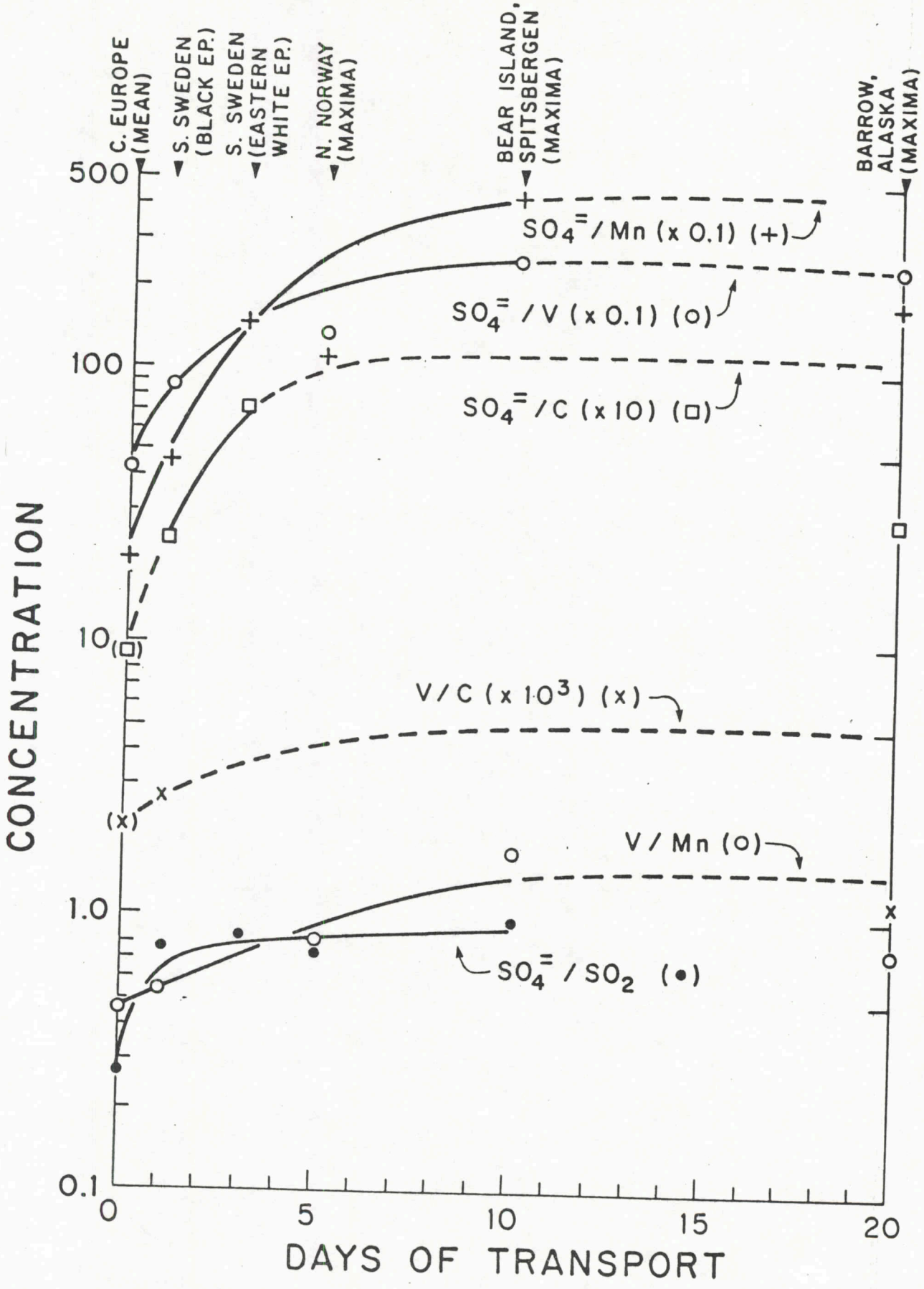

Figure 4. Evolution of various secondary/primary and primary/primary ratios during winter transport of polluted air masses from Eurasia to the Arctic. All data from Figure 3.

a highly unified and regular process of atmospheric aging. The main variable of this process, at least as far as these species are concerned, is time alone. This confirms our working hypothesis stated above. (2) Black and eastern white episodes represent early stages of this aging process, and have ages of roughly 1 and 3 days, respectively. The major difference between black and eastern white episodes is time, or degree of aging, which is reflected in the secondary/primary (sulfate/soot) ratio of the aerosol. Although there may well be differences in sources or aging between black and eastern white episodes, or between any of the other aerosols discussed here, they do not have to be invoked to explain the basic patterns of the empirical aging diagram. (3) The rate of aging decreases rapidly during transport, and reaches very small values in the cold, dry and dark Arctic night. The major transformation from primary to secondary character of the Eurasian aerosol is largely completed within the first 4-5 days. (4) The relative atmospheric lifetime of elemental C compared to other primary pollutants such as V or Mn is not yet clear. In the first days of transport, it would appear to be similar to them, and to Mn in particular; subsequent behavior is obscured by the apparent additional source influencing Barrow. Further data at intermediate points are needed to resolve this question.

An intriguing question for the future is the extent to which the large-scale features of atmospheric aging revealed here are general, i.e., can be found downwind of other major polluted areas. Eastern North America and eastern Asia are the two other source regions near which such studies might be carried out. In each of these cases, however, the downwind transport is over oceans and at lower latitudes, where meteorological and chemical conditions, hence atmospheric aging, may be quite different from that of the Eurasian case.

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