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The transatlantic transport of sulfur

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ABSTRACT

This paper examines the transport of North American sulfur emissions across the North Atlantic Ocean to Europe. A review of available precipitation sulfate data from the North Atlantic and adjacent coastal regions yields a concentration field which is consistent with known source distributions and meteorological factors. The marine background precipitation excess sulfate concentration is found to be 6–8 μeq l^{-1} and $[SO_4^{=*}]$ to decrease from >50 μeq l^{-1} with offshore flows at the North American east coast to 8–15 μeq l^{-1} with onshore flows at the European west coast. This decay is consistent with a distance constant of 2400 km and a residence time of ~ 80 h, and in turn, corresponds to a transatlantic flux of anthropogenic sulfur of 0.3–0.4 Tg a^{-1}. A second independent estimate, based on the application of a climatological dispersion model, which accounts for long-term average diffusion, wet and dry deposition, and SO_2 to $SO_4^=$ transformation, yields a flux of North American anthropogenic sulfur at the European west coast of 0.2 Tg a^{-1}, in agreement with the first estimate.

At the distance of the European west coast, North American anthropogenic emissions account for ca. 4 μeq S l^{-1} in precipitation—less than the marine background of 6–8 μeq l^{-1}, and much less than the annual average $[SO_4^{=*}]$ value of ca. 30 μeq S l^{-1} appropriate for much of the coastal region. It is concluded that, on average, the amount of North American anthropogenic sulfur reaching Europe is small compared to that from other sources.

1. Introduction

The intense interest in the transport and deposition of acidic pollutants in North America and Europe has naturally led to questions of the origin of the background levels of acidic constituents in air and precipitation, and of the possible transport of such material across the Atlantic Ocean. Since the time of the OECD Programme on Long-Range Transport of Air Pollutants in Europe (OECD, 1979) the question of the origin of excess precipitation sulfate associated with off-ocean trajectories has not been resolved (Eliassen and Saltbones, 1983; Fisher and Clark, 1985). On the other side of the Atlantic, Galloway et al. (1984b) have estimated that 37% of man-made sulfur emissions in eastern North America is advected eastward over the Atlantic. Approximately one-half of this is carried above approximately 1500 m, where it is available for extended transport over the ocean by the prevailing westerly winds. Measurements of precipitation composition made over the Atlantic Ocean have shown that elevated concentrations of acidic species are associated with air mass trajectories which have previously passed over the heavily industrialized areas of North America (e.g., Nyberg, 1977; Jickells et al., 1982). Unfortunately, such over-ocean measurements are sparse.

The purpose of this paper is to provide an estimate of the amount of anthropogenic sulfur transported across the North Atlantic Ocean from North America to Europe. As a first approach we examine the available precipitation sulfate concentration data from sites on and adjacent to the North Atlantic. Based on the concentration decrease with distance from North America an over-ocean residence time for sulfur

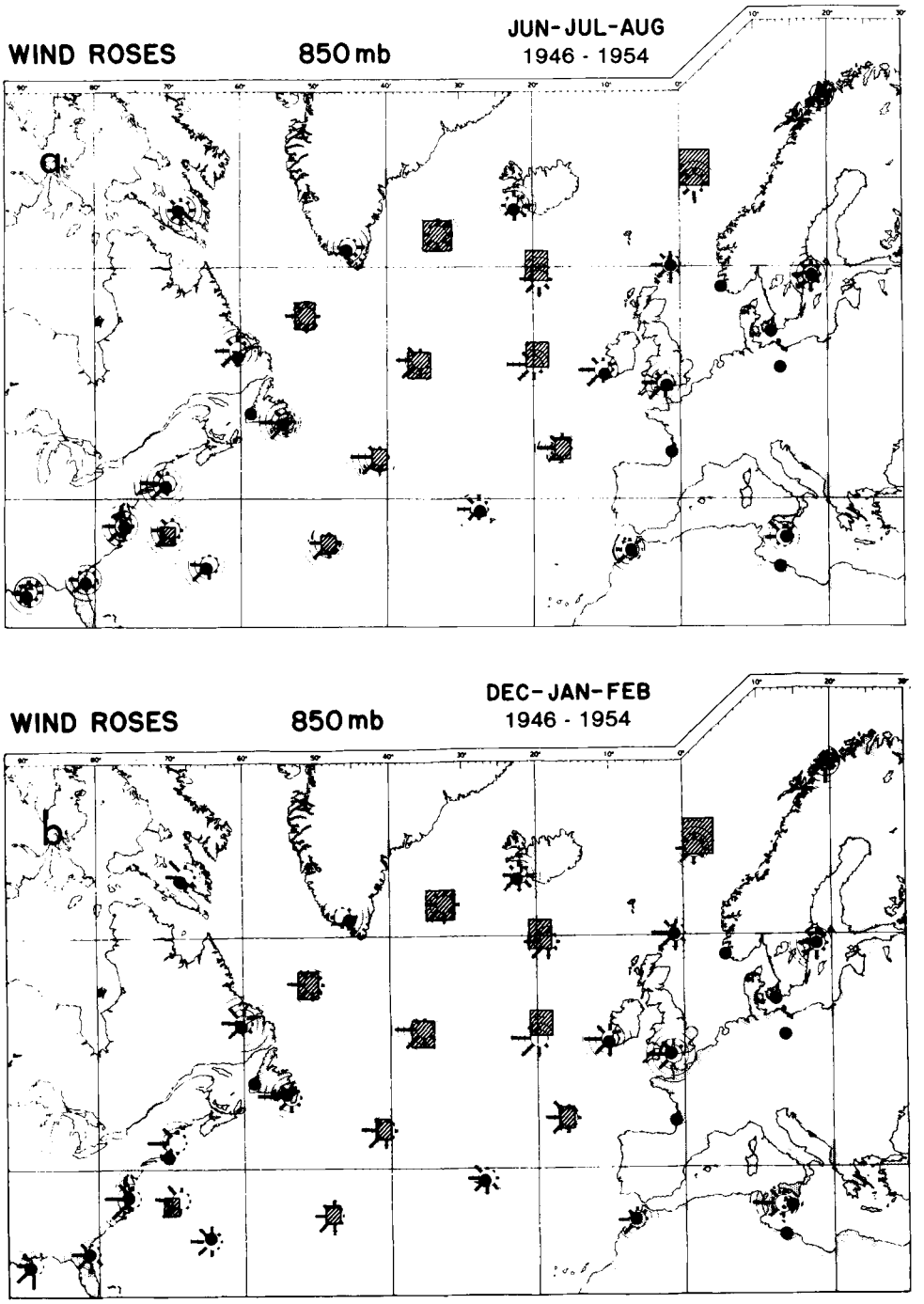

Fig. 1. Climatological 850 mb wind direction frequencies over the North Atlantic Ocean: (a) June–July–August; (b) December–January–February (US Navy, 1955). Bars represent percentage frequency of wind observed from each direction. Each circle equals 10%. Shaded boxes represent ship locations.

is obtained and used to calculate the eastward flux. A second approach makes use of a simple climatological dispersion model, whose parameter values have been derived independently, to calculate the long-term average transport from North American emission sources to the European west coast.

2. Precipitation sulfate over the North Atlantic

2.1. Approach

The region of interest is the North Atlantic Ocean, bounded by the coastlines of North America and Europe, and by approximately 30° and 70° N latitude. Within the conceptual framework of a steady-state box model budget for the North Atlantic, the western boundary inflow is assumed known, and the eastern boundary outflow is the term to be determined. Other budget terms—wet deposition, dry deposition, outflow to the North and South, vertical outflow, and natural emissions from the ocean—are not explicitly considered. The assumptions inherent in this approach are first, that the prevailing westerly flow regime over the North Atlantic is sufficiently consistent, on average, to transport material across the ocean and second, that the concentration of sulfur in precipitation at a given distance from the North American continent is a reasonable indicator, on average, of the amount of sulfur which has been transported that distance.

Fig. 1 shows summer and winter wind directional frequencies and speeds for the North Atlantic, based on a 10-y record for the 850 mb level (U.S. Navy, 1955). The strong westerly component of the flow over the entire region of interest gives support to the first assumption above. The chemical composition of precipitation depends upon the composition of the air in which the precipitation formed and fell, i.e., what remains after transport from the continent, past deposition, and recent over-ocean inputs. Direct air concentration measurements would of course be better; however, the above-surface measurements required are almost non-existent. Thus, precipitation composition, averaged over suitably long periods (seasons, years) where possible, is used as an indicator of atmospheric composition.

In contrast to continental regions, the precipitation regime over the oceans is not well known. Fig. 2 (Elliott and Reed, 1984) shows a recent climatological estimate for the North Atlantic. For most of the region of interest, annual precipitation ranges from about 50 cm in the southeast to more than 100 cm between Newfoundland and Iceland. Precipitation is greater near the North American coast and along the Norwegian coast. Most adjacent coastal stations and shipboard measurements show a clear summer minimum and winter maximum in precipitation amount. Because the ocean surface is cool relative to off-continental air in summer and warm in winter there is a tendency for temperature inversions over the surface in summer and fairly strong vertical mixing in winter. In turn these conditions favour decoupling of the upper layers with stratus cloud or fog in summer, and more turbulent, better mixed winter conditions with convective cloud. The combination of higher precipitation and convective activity in winter over the ocean would be expected to result in more effective removal of atmospheric constituents, from a deeper layer of the atmosphere. Thus, distant transport across the ocean would be more likely in summer.

2.2. Available data

Data used in this study are listed in Table 1 and plotted in Fig. 3. For each location, the latitude, the longitude and the sampling period are given. Most data are from 1978–1982 although some go back as far as 1968. The number

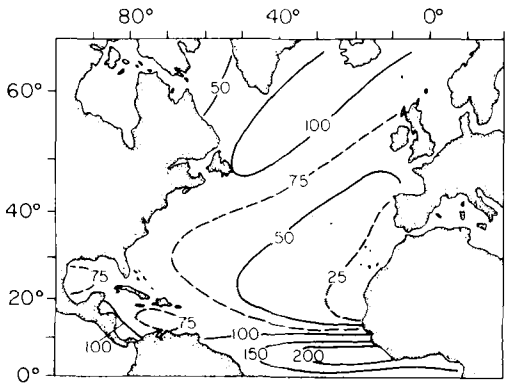

Fig. 2. Annual precipitation (cm) over the North Atlantic Ocean (adapted from Elliott and Reed (1984)).

Table 1. *Precipitation-$SO_4^{=*}$ data for North Atlantic Ocean*

Location (lat., long.)	Period	Number of samples	$SO_4^{=*}$ (μeq l^{-1}) gm or median	pwm	am	Footnote
Lewes, De. (38°46′N 75°00′W)	1978–1979	47	40.6	45.6	55.4	1
	May 1980–Apr 1981	—	—	53.2	—	2
	May 1981–Oct 1981	24	58.0	—	65.4	3
Brookhaven, N.Y. (40°52′N 72°53′W)	1978–1979	45	39.6	38.7	52.6	1
Hubbard Brook, N.H.	Jul 1975–Jul 1980	100	43	—	52.2	4
(43°57′N 71°42′W)		47	60.2	—	—	†
Shelburne, N.S. (43°43′N 65°15′W)	1980–1982	35	30	29	36	5
Kejimkujik, N.S. (44°26′N 65°12′W)	1980–1982	35	27	27	32	5
Gander, Nfld. (48°57′N 54°34′W)	1980–1982	29	22	34	30	5
Goose Bay, Nfld. (53°19′N 60°22′W)	1980–1982	34	16	15	15	5
Sable Is., N.S. (43°56′N 60°01′W)	1980–1982	21	32	45	47	5
Greenland (65°11′N 43°50′W)	(See Reference)	—	1.0	—	1.1	6
Western Atlantic	May–Oct 1981	20	8.4	—	12.1	3
(New York–Bermuda–Nassau)		10	12.7	—	18.2	†
Bermuda	Apr 1980–May 1984	268	14.1	—	18.5	7
(32°19′N 64°45′W)		120	17.5	—	23.8	†
Northeastern Atlantic	1973–1975	20	16	—	19	8
(52–62°N 18–33°W)		9	18	—	26	†
(64–66°N 02–05°E)	1973	3	37	—	76	9
		2	34	—	34	†
(57°N 20°W)	1979–1981	276	6.8	12.4	—	10
(66°N 02°E)	Apr 1979–Oct 1981	42	24	34	44	11
(65–70°N 0–02°W)	1979	—	—	6	—	12
Irafoss, Iceland	Oct 1979–Sep 1982	477	—	20.6	—	13
(64·05′N 21°10′W)		75	—	23.1	—	†
Faeroes, Denmark (62°18′N 07°04′W)	1978–1980	34	31	30	38	14
(61°24′N 06°40′W)	Oct 1978–Sep 1982	797	—	58.1	—	13
		169	—	50.0	—	†
Bantry Bay, Ireland (51°41′N 09°45′W)	Nov 1984–Feb 1985	28	11.8	9.1	14.5	7
Valentia, Ireland	1968–1974	—	—	—	25	15
(51°56′N 10°15′W)	1975–1977	—	—	30	—	16
	1978–1980	35	29	30	34	14
	Oct 1979–Sep 1982	529	—	16.9	—	13
		225	—	15.0	—	†
Belmullet, Ireland (54°07′N 10°00′W)	1968–1974	—	—	—	25	15
Lerwick, U.K. (60°14′N 01°19′W)	1978–1979	—	—	55	—	17
Bettyhill, U.K. (58°32′N 04°14′W)	1978–1980	—	—	32	—	17
Inverpolly, U.K. (58°08′N 05°14′W)	1978–1980	—	—	17	—	17
Broadford, U.K. (57°12′N 05°51′W)	1978–1980	—	—	26	—	17
Torlundy, U.K. (56°48′N 05°06′W)	1979–1980	—	—	43	—	17

Table 1—*continued*

Location (lat., long.)	Period	Number of samples	$SO_4^{=*}$ (μeq l^{-1}) gm or median	pwm	am	Footnote
Lephinmore, U.K. (56°06′N 05°11′W)	1978–1980	—	—	32	—	17
Goonhilly, U.K. (50°03′N 05°11′W)	Oct 1979–Sep 1982	493	—	40.0	—	13
		205	—	31.3	—	†
	1980	—	—	44	—	17
Soyland, Norway (58°41′N 05°59′E)	Jul 1972–Mar 1975	—	—	18	—	18 †
Skei i Jolster, Norway (61°34′N 06°29′E)	Jul 1972–Mar 1975	—	—	13	—	18 †
Fitjar, Norway (59°55′N 05°19′E)	Aug 1972–Mar 1975	—	—	18	—	18 †
Fillefjell, Norway (61°11′N 08°07′E)	Jun 1973–Mar 1975	—	—	11	—	18 †
Bjornoya, Norway (74°31′N 19°01′E)	Jan 1978–Dec 1982	—	—	50	—	19
Jergul, Norway (69°24′N 24°36′E)	Jan 1978–Dec 1982	—	—	31	—	19
Tustervatn, Norway (65°50′N 13°55′E)	Apr 1978–Sep 1982	779	—	14.4	—	13
		251	—	8.1	—	†
	Oct 1978–Sep 1979	13	—	—	8	20†
Kaarvatn, Norway (62°47′N 08°53′E)	Apr 1978–Sep 1982	712	—	12.8	—	13
		295	—	12.5	—	†
	Oct 1978–Sep 1979	26	—	—	4	20†
Skreaadalen, Norway (58°49′N 06°43′E)	Apr 1978–Sep 1982	787	—	32.5	—	13
		81	—	6.9	—	†
	Oct 1978–Sep 1979	31	—	—	12	20†

† Value is for precipitation associated with air of N.A. origin (see text).

1. MAP3S/RAINE (1982). Total S, not excess S. Several months of data missing in period.
2. Church et al. (1982).
3. Galloway et al. (1983). Western Atlantic samples are from ships during westerly storms.
4. Munn et al. (1984). Values derived from reference as precipitation-weighted-mean of medians from all groups, and groups 3, 4, 5, 7, 10, respectively.
5. CANSAP (1982a, 1982b, 1983). The pwm values are arithmetic means of 3 annual pwm values.
6. Galloway (1985). Based on a review of S concentrations in surface snow and ice cores.
7. Data supplied by J. N. Galloway. See also Galloway et al. (1982, 1983).
8. Nyberg (1977). Data from OWS Alpha (62° N 32° W), India (59° N 19° W), Juliet (52° N 20° W) and Lima (57° N 20° W) grouped. Samples marked with † were associated with 5-day, 850 mb back trajectories which had originated over North America. OWS Kilo (45° N 16° W) data were not used since they were associated with trajectories from remote areas.
9. Nyberg (1977). Data from OWS Mike.
10. Buijsman et al. (1984), and E. Buijsman (personal communication, 1985): data from OWS Lima.
11. W. Asman (personal communication, 1982): Data from OWS Mike, considered representative for June–November period. Uncertainty in weighted mean estimated at ~30%.
12. Varhelyi and Gravenhorst (1983). Number of samples and type of mean value unknown.
13. Data from the European Monitoring and Evaluation Program supplied by H. Dovland. Sector allocations (9: N, NE, . . ., NW, Undecided) are based on 4 96h 850 mb trajectories per day. If more than ½ the trajectory positions (every 2h, 4 d^{-1}) are in a sector, the event is allocated to that sector. Distances between 150 and 1500 km are used.
14. EDIS (1981); NESDIS (1983); NESDIS (1984).
15. Fisher (1982). Bulk samples.
16. Mészaros (1982). Based on WMO BAPMoN data. Data uncertainty estimated at ±50%.
17. Barrett et al. (1983). Monthly bulk samples are collected at all sites except Goonhilly, where daily samples are taken.
18. OECD (1979). Data for precipitation associated with air from oceanic sector.
19. EMEP/CCC (1984).
20. Data supplied by H. Dovland and J. Saltbones. These averages are based on cases in which the air had not passed over any man-made emissions.

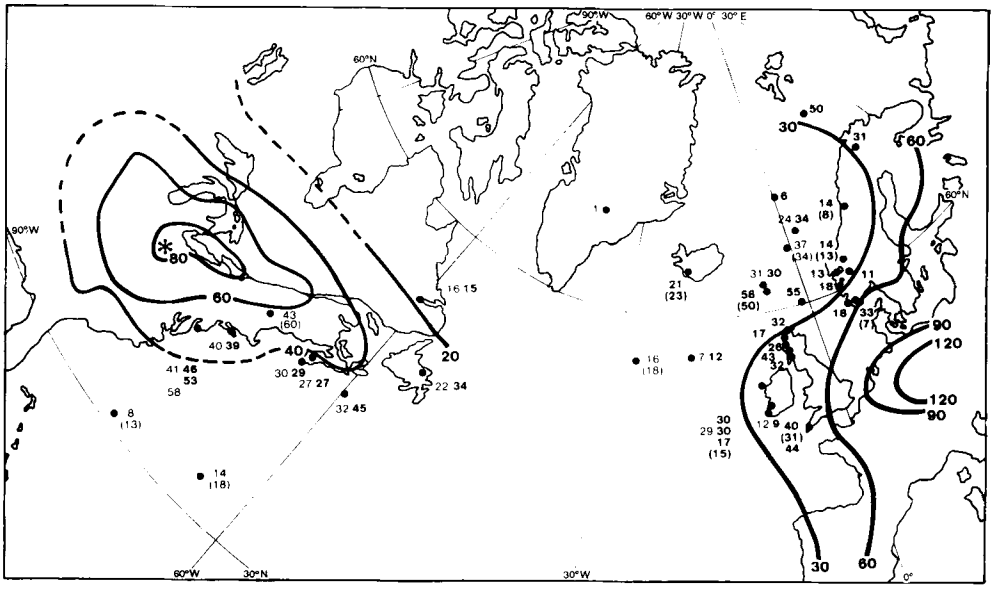

Fig. 3. Excess precipitation-sulfur concentrations (μeq l^{-1}) for the North Atlantic Ocean and adjacent areas. • denotes approximate emissions centroid for eastern North America. See text for details.

of samples contributing to the averages are given. Median values (or geometric means) were the preferred measure of central tendency in the data sets; precipitation-weighted means and arithmetic means are also given, where available. Footnotes to Table 1 contain information on the data and references. Uncertainties in such a data set are many: they arise, in part, because the data are from several sources whose sampling periods, times and methods differ. Thus, we look for large-scale spatial consistency in the data, rather than for quantitative detail.

2.3. *North Atlantic background*

It is first instructive to establish a "background" value of precipitation sulfur concentration over the North Atlantic Ocean. Even the most remote parts of the North Atlantic are undoubtedly affected by anthropogenic emissions on occasion; therefore, we use data which investigators have associated with air flows from "clean" regions, and other very low values as defining background. Only a few such data are available.

Estimates of the North Atlantic background have been obtained from three Norwegian stations during the European Monitoring and Evaluation Program, EMEP (Table 1, Footnote 20). Based on events in which air had not previously crossed anthropogenic emission areas, average [$SO_4^{=*}$] were 8, 4 and 12 μeq l^{-1} for the year October 1978–September 1979. Nyberg (1977) found that the concentration of excess S in precipitation associated with air coming from the high pressure area near the Azores ranged from 2–11 μeq l^{-1} in five events, with a median value of ~6 μeq l^{-1}. Based on a tabulation by Galloway (1985), the concentration of $SO_4^{=*}$ in surface snow and ice cores (resulting from wet plus dry deposition) in Greenland ranges from 0.5 to 1.7 with a median value of 1.0 and an arithmetic mean of 1.13 ± 0.51 μeq l^{-1}. Galloway et al. (1983) found average excess S concentrations in easterly storms, measured on board cruise ships travelling between Bermuda and the east coast, and at Bermuda, to be 6.1 and 11 μeq l^{-1}, respectively. On the basis of these data, representative background precipitation $SO_4^{=*}$ concentrations for the North Atlantic are thought to be approximately 6–8 μeq l^{-1}.

These values are slightly higher than background concentrations reported by Galloway et al. (1984a) for a number of remote global sites. This suggests that over the North Atlantic, in the absence of direct influence from North American and European sources, that precipitation sulfur

concentrations are almost as low as those in remote parts of the world.

2.4. North American east coast

Barrie and Hales (1984) have compiled precipitation data from four major North American networks for the year 1980. The annual precipitation-weighted $SO_4^{=*}$ concentration field is reproduced in Fig. 3. In the east coastal region of North America south of ~50°N concentrations are in the range of ~30–60 μeq l^{-1}. Maximum values occur in the region near 40°N and the lower values occur in the Atlantic Provinces. North of 52°N concentrations fall below 20 μeq l^{-1}. More recent 1981 data from Lewes, De. (Galloway et al. 1983) and the Shelburne and Kejimkujik CANSAP stations (Sirois, 1983) are consistent with this picture. Less reliable data (monthly samples, less strict quality control) from the WMO BAPMoN station at Sable Island yield a 1978–1980 median value of 32 μeq l^{-1}.

2.5. European west coast

Data from the European Monitoring and Evaluation Program for the years 1978–1982 (EMEP/CCC, 1984) are also shown on Fig. 3. The 30 μeq l^{-1} (0.5 mg S l^{-1}) contour passes northward through Portugal to the west of the British Isles and then turns to the northeast through Scandinavia. Low EMEP station density in the area south of 60°N through which this contour passes makes its placement somewhat uncertain; however, additional data are available from a number of near coastal stations in the British Isles. Data from Inverpolly, 17 μeq l^{-1}, from Valentia (EMEP), 17 μeq l^{-1}, and from Bantry Bay, 12 μeq l^{-1}, suggest that typical concentrations at coastal sites unaffected by local sources may be significantly less than indicated by the 30 μeq l^{-1} contour in Fig. 3 between 50° and 60°N.

Norwegian near-coastal stations at Karvatn and Tustervatn have 1978–1982 pwm $[SO_4^{=*}]$ values of 13 and 14 μeq l^{-1}, respectively (Table 1, footnote 13), indicating a decrease in concentration north of 60°N. The higher concentrations at Jergul and Bear Is. result, respectively, from a combination of topography and European emissions, and from a combination of very low precipitation and high sea spray. The EMEP station on Iceland has a 1979–1982 pwm $[SO_4^{=*}]$ value of 21 μeq l^{-1} (Table 1, footnote 13), and the WMO site on the Faeroes, a 1978–1980 median value of 31 μeq l^{-1} (EDIS, 1981; NESDIS, 1983; NESDIS, 1984). Other reported data from the Faeroes (Table 1; Varhelyi and Gravenhorst, 1983) appear to show a large influence of sea spray or local sources. Limited data are also available from Ocean Weather Ship Cumulus at position Mike (66°N 2°E) in the North Atlantic. W. Asman (personal communication, 1982) reports, for the period June to November for years 1979–1981, a median value of 24 μeq l^{-1}, with an estimated uncertainty of ~30%. He considers these data to be influenced by emissions from Europe.

3. Results

3.1. Dependence of sulfur concentrations on distance from North America

Precipitation sulfate concentrations from Table 1 are plotted in Fig. 4 as a function of distance from the estimated centroid of eastern North American emissions (c.f., Fig. 3). To make use of as much of the available data as possible, precipitation-weighted mean values are included where medians are not available. Open symbols repre-

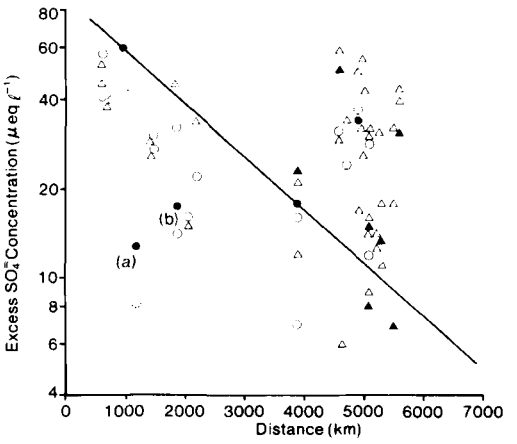

Fig. 4. Variation of excess precipitation sulfate concentration (μeq l^{-1}) with distance from emissions centroid (Fig. 3). Key: circles—median values; triangles—precipitation-weighted-mean values; open—all data; solid—data associated with westerly flows. See text for discussion of line.

sent all data at a site, and solid symbols denote only data associated with flow from North American source regions (see Subsection 3.2.). Out to a distance of about 4000 km concentrations decrease with distance and show large scatter. The lower concentrations from the cruise ships travelling between New York, Bermuda and Nassau are likely the result of a seasonal bias (summer collection only) since this region experiences relatively little flow from the region of continental emissions to the northwest during summer (Fig. 1).

The European west coast region data, centred around 5000 km, show a large spread in concentration. As discussed briefly in Subsection 2.5, sites having higher concentrations (i.e., >ca. 20 μeq l^{-1}) fall into one of the following categories: suspected contamination from a local source—Faeroes, Goonhilly; known to be influenced by European emissions—Skreaadalen, Jergul, OWS (66N, 2E); monthly bulk samples with attendant likelihood of dry deposition influence—Lerwick, Torlundy, Bettyhill, Lephinmore, Broadford; large sea-spray correction and resultant uncertain excess sulfate value—Bear Island, Faeroes; WMO monthly data—Faeroes, Valentia. Thus, despite the fact that these sites were selected initially as having the greatest potential to provide data representative of clean conditions, there is reason to suspect that they do not do so. Stations which have lower concentrations are known, with the exception of Inverpolly and OWS (VG), to be sites where sampling representative of clean conditions can be, and has been carried out. These include Fitjar, Soyland, Valentia (EMEP), Kaarvatn, Tustervatn, Skei i Jolster, Bantry Bay, Fillefjell and Skreaadelen (ocean sector). (A minor influence is the fact that most available values here are precipitation-weighted means rather than the corresponding, slightly lower medians—cf. Table 1).

3.2. *Dependence of precipitation composition on wind direction*

Data sets in which measurements were stratified according to wind or trajectory direction are listed in Table 2 (solid symbols in Fig. 4). The table permits a comparison for each site of concentrations based on all measurements with those stratified by source region. Individual concentration values are less important here than the ratios, since differing means and source region definitions were used.

For the first six entries, i.e., out to ~4000 km in Fig. 4, precipitation associated with flow from North America has higher concentrations than does all precipitation by a factor of from 1.1 to 1.8. For the last seven entries, where the influence of European emissions becomes stronger, the concentrations associated with flow from the direction of North America are less than for all data, by a factor ranging from 0.2 to 0.9. This simply means that under conditions of eastward transport, precipitation sulfate concentrations are larger, at the east coast of North America, and smaller, at the west coast of Europe, than would be indicated by using all available precipitation data (i.e., associated with winds from all directions). This also means that the transatlantic transport of sulfur would be smaller than inferred from the use of all data. The final column in Table 2 shows that in all but one case concentration values associated with flow from remote regions are less than those associated with *all* data.

3.3. *Distance constant and residence time*

Pollutants being transported away from the North American continent experience several processes which may result in a long-term average decrease in their concentration in an easterly direction, i.e., towards Europe. These include transport northward into the sub-polar and polar regions, transport southward to the tropical Atlantic, vertical mixing to heights above which precipitation scavenging is usually effective, and dry deposition to the ocean surface. The concentration of sulfur in precipitation at any time and distance is a result of the past history of the pollution-containing air mass which has experienced these various processes. With the exception of that sulfur transported above levels at which scavenging occurs the concentration decrease with travel distance from Fig. 4 provides a means of estimating the over-ocean distance constant and effective average residence time of sulfur.

The line drawn in Fig. 4 is a subjective estimate of the decrease of concentration with distance based on *wind-direction stratified data only* (solid symbols—cf. Subsection). In drawing this line, the three data points (solid symbols) in the

Table 2. *Precipitation sulfur concentration as a function of source region*

Location (reference)	Sulfur (hydrogen) concentration (μeq l^{-1}) All Data (A)	North America (NA)	Europe (E)	Remote (R)	NA/A	R/A
Hubbard Brook, N.H. (Munn et al., 1984)	43	60(1)	—	10	1.4	0.2
Long Island, N.Y. (2) (Raynor & Hayes, 1981)	52	93	—	49	1.8	0.9
Cruise Ships: New York, Bermuda, Nassau (Galloway et al., 1983)	12	18	—	6	1.5	0.5
Bermuda (4) (this paper: JNG data)	14	18	—	11	1.3	0.8
Ocean Weather Ships (4) (Nyberg, 1977)	16	22	106	6	1.4	0.4
Irafoss, Iceland (5) (this paper: EMEP data)	21	23	29	21	1.1	1.0
Norwegian Coast (5, 6) (OECD, 1979)	32	18	76	18	0.6	0.6
Tustervatn, Norway (5, 6) (this paper: EMEP data)	14	8	23	8	0.6	0.6
Kaarvatn, Norway (5, 6) (this paper: EMEP data)	14	13	17	13	0.9	0.9
Skreaadalen, Norway (5, 6) (this paper: EMEP data)	33	7	36	7	0.2	0.2
Valentia, Ireland (5) (this paper: EMEP data)	17	15	17	15	0.9	0.9
Goonhilly, U.K. (5) (this paper: EMEP data)	40	31	70	33	0.8	0.8
Faeroes, Denmark (5) (this paper: EMEP data)	58	50	84	43	0.9	0.7

(1) Derived from reference as precipitation-weighted mean of sector medians.
(2) H^+ data precipitation-weighted means: S data not available.
(3) Arithmetic means.
(4) Medians.
(5) Precipitation-weighted means.
(6) Sectors for "North America" and "Remote" are the same.

upper right of the figure were given less weight. Data from the Faeroes and Goonhilly sites are thought to be influenced by local sources (cf. Subsection 2.5), and the OWS (64–66°N 02–05°E) data point is based on only two event samples. Data from two other sites (Western Atlantic ships and Bermuda—points (a) and (b), respectively, in Fig. 4) were also given less weight because these sites lie to the south of the main region of pollution outflow from North America (Fig. 1 and Whelpdale et al., 1984). Their lower concentrations probably result from the relatively more frequent inclusion of "clean" precipitation episodes in the average values. (See also Subsection 3.1.) The slope of the line in Fig. 4 is uncertain. To provide an indication of the effect that a change in slope would have, we calculate that an increase or decrease in slope to envelope the solid symbols (expecting those discussed above) between 4000 and 6000 km would result in a change in the distance (time) constant derived below of approximately $\pm 25\%$.

This simple model of an exponential decrease in concentration yields a distance constant of 2400 km. Using a mean over-ocean wind velocity of 8 ms^{-1} from maps of Lau et al. (1981) results in a time constant of approximately $3\frac{1}{2}$ days. Although the time constant derived is not strictly

the same as the average residence time (Rodhe, 1972), the value is similar to those reviewed and proposed by Rodhe (1978), 90 h.

3.4. *Transatlantic flux*

On the basis of the change in precipitation composition as a function of distance from North America it is possible to estimate the amount of sulfur crossing the Atlantic. We use the assumption stated earlier that the composition of precipitation is a measure of the amount of sulfur in the atmosphere at that distance, and in turn, that the eastward flux also diminishes according to the same distance constant.

Estimates of the flux of sulfur leaving the North American continent to the east are available. Galloway et al. (1984b) calculate that 4.3 Tg S a^{-1} ($\pm 60\%$) is transported eastward from North America. This figure was based on climatological wind data, average S concentration profiles and 1980 eastern North American emissions of 12.5 Tg S a^{-1}. J. Shannon (personal communication, 1985) has recently used the ASTRAP model to calculate an average eastward flux of 5.5 Tg S a^{-1} based on 1980 North American emissions of 15.0 Tg S a^{-1} and 1976–1981 meteorological data. We choose an eastward flux of approximately 5 Tg S a^{-1} at the North American east coast, along with a distance constant of 2400 km, to obtain a total S flux at the European west coast (4800-5500 km distant) of approximately 0.7 Tg S a^{-1}.

This value for the transatlantic flux includes both an anthropogenic and a natural component. The anthropogenic component will decrease with distance across the ocean in the absence of anthropogenic sources, but the background component would likely decrease more slowly or not at all (depending on the relative magnitude of continental and marine natural sources affecting the region), and then only down to the marine background level established earlier. The magnitude of the natural component can be estimated as the flux from natural sources leaving North America, ca. 5–10% of the total 4.3 Tg S a^{-1} (Galloway and Whelpdale, 1980), or by noting from Fig. 4 the relative magnitudes of total and background precipitation sulfur concentrations at the European west coast, ca. 11 and 7 μeq l^{-1}, respectively. Both methods indicate that approximately one-half of the total flux derived above may be background. Thus, the anthropogenic transatlantic sulfur flux is estimated to be approximately 0.3–0.4 Tg a^{-1}.

A number of uncertainties are associated with this estimate. As noted earlier the precipitation data themselves are of variable quality. Improvements in sampling, e.g., careful event sampling, and more selective meteorological stratification, might be expected to yield lower concentrations near Europe and higher concentrations near North America. This would result in a shorter distance constant and smaller flux. Transport above a height at which scavenging is effective (cf. Galloway et al. 1984b) would result in a larger flux than calculated, but would not necessarily be reflected in the composition of precipitation at the European west coast, but rather as deposition further inland, the result of large-scale fumigation.

4. A model calculation of transatlantic flux

4.1. *Model formulation*

An independent estimate of the long-term average transport of sulfur from North America to Europe can be made using a simple model of the "Rodhe-Venkatram" type. Such a model describes the long-term spread of sulfur from a point source of strength Q within an angle θ and a mixed layer of depth h. The model includes an assumed linear transformation of sulfur dioxide to sulfate. This is necessary because sulfate travels farther than sulfur dioxide due to less efficient dry deposition.

The fluxes of SO_2 and $SO_4^=$ may be defined, respectively, as

$$G = C_1 u\theta rh;\ S = C_2 u\theta rh, \tag{1}$$

where C_1 and C_2 are the concentrations of sulfur dioxide and sulfate, respectively, u is an average wind velocity and r is the distance from the source. G and S change with travel time t due to deposition, described by average rates λ and $\bar{\lambda}$, and to transformation with an average rate k_t. The resulting mass balance equations are

$$\frac{dG}{dt} = -\lambda G - k_t G; \qquad \frac{dS}{dt} = \bar{\lambda} S + k_t G, \tag{2}$$

where sulfur dioxide and sulfate are both measured in sulfur units.

These first-order equations with constant coefficients can be solved for G and S. Substituting r/u for the travel time t, one obtains

$$G = (1-\beta)Q \exp\left[-(\lambda + k_t)\frac{r}{u}\right], \tag{3}$$

$$S = \frac{k_t}{\alpha}(1-\beta)Q \exp\left(-\lambda\frac{r}{u}\right)\left[1-\exp\left(-\alpha\frac{r}{u}\right)\right] + \beta Q \exp\left[\bar{\lambda}\frac{r}{u}\right], \tag{4}$$

where

$$\alpha = \lambda - \bar{\lambda} + k_t. \tag{5}$$

To parameterize the faster transformation to sulfate observed just after emission, a small part β of the sulfur is assumed to be emitted directly as particulate sulfate. $\beta = 0.05$ gives reasonable agreement between observed and calculated particulate sulfate concentrations over Europe (Eliassen and Saltbones, 1983).

The deposition D of sulfur (mass per unit area and time) is related to the change in the flux of total sulfur:

$$\frac{d}{dr}(G+S) = -\theta r D. \tag{6}$$

Addition of the two eq (2) gives

$$u\frac{d}{dr}(G+S) = -\lambda G - \bar{\lambda} S. \tag{7}$$

Elimination of $(d/dr)(G+S)$ from (6) and (7), and use of (1) gives a relationship between the deposition D and the concentrations:

$$D = h(\lambda C_1 + \bar{\lambda} C_2). \tag{8}$$

4.2. *Parameter values*

Parameter values for the model calculations are derived independently of results from Section 3, in order to obtain a second, unrelated estimate of transatlantic flux. For wet deposition, removal rates are derived following Rodhe and Grandell (1972, 1981). A constant scavenging coefficient λ_p is assumed to apply during precipitation. Considering the sequence of dry and wet periods experienced by a pollutant-carrying particle or trajectory, the rate of wet removal that applies may be taken as a stochastic process $\lambda(t)$ defined by

$$\lambda(t) = \begin{cases} 0 \text{ if no precipitation at } t \\ \lambda_p \text{ if precipitation occurs at } t. \end{cases} \tag{9}$$

Assuming that $\lambda(t)$ is a Markov process with stationary transition probabilities, the lengths of the dry and wet periods are exponentially distributed. If τ_d and τ_p are the average lengths of dry and wet periods experienced by the moving particle, Rodhe and Grandell showed that the average residence time T_p for the pollutant with respect to wet deposition is

$$T_p = \tau_d p_d + \frac{\tau_d + \tau_p}{\tau_p}\lambda_p^{-1}, \tag{10}$$

where p_d is the probability of a dry period at the release point of the trajectory. The quantities τ_d and τ_p have been estimated by Hamrud et al. (1981) for Europe from trajectories and precipitation data. τ_d was found to be 51 hours in summer and 53 hours in winter, whereas τ_p was 11 hours in both cases. We choose $\tau_d = 50$ hours and $\tau_p = 10$ hours and assume that these apply reasonably well to transatlantic transport. The Eulerian probability is approximated from the Langrangian data, i.e., $p_d \sim \tau_d/(\tau_d + \tau_p) = 0.83$. Finally, we choose $\lambda_p^{-1} = 1$ hour, a value thought to be fairly representative for both sulfur dioxide and particulate sulfate. Inserting these values in (10) gives

$$T_p = 50 \text{ hours} \cdot 0.83 + 6 \cdot 1 \text{ hour} \sim 47.5 \text{ hours}.$$

For pollutants that are reasonably efficiently removed by rain, T_p is determined mainly by τ_d, and is relatively insensitive to λ_p^{-1}. Thus, somewhat different λ_p^{-1}-values for sulfur dioxide and particulate sulfate would not make much difference for T_p. We have therefore assumed $T_p = 47.5$ hours for both species.

To estimate dry deposition, a deposition velocity v_d of 0.8 cms^{-1} is assumed for sulfur dioxide and the corresponding quantity w_d for particulate sulfate is taken as 0.1 cms^{-1}. A mixing height h of 2000 m (i.e., the layer throughout which the sulfur is assumed to be dispersed) is chosen to represent transatlantic transport. This implies residence times with respect to dry deposition of $h/v_d = 69.4$ hours for sulfur dioxide and $h/w_d = 555$ hours for particulate sulfate.

The assumptions made about wet and dry deposition are now combined to evaluate the average removel rates (e.g., Rodhe, 1978)

$$\lambda = T_p^{-1} + \frac{v_d}{h} = 9.85 \cdot 10^{-6}\,\text{s}^{-1}, \qquad \bar{\lambda} = T_p^{-1} + \frac{w_d}{h} = 6.35 \cdot 10^{-6}\,\text{s}^{-1}, \tag{11}$$

The transformation rate k_t is taken as $3 \cdot 10^{-6} s^{-1}$ ($\sim 1\%$ per hour), which is thought to be a representative annual average. Finally, the average windspeed was assumed to be 11 ms^{-1}. The parameter values chosen are summarized in Table 3.

4.3. *The flux of North American sulfur at the European west coast*

First, the flux of total sulfur $G + S$ through a circle of radius $r = 5500$ km, which is roughly the distance from the main North American emissions to the European west coast, is calculated using eqs. (3) and (4). An annual Eastern North American sulfur emission of 15 Tg is assumed, and taken as a point source. This calculation gives

$$G_0 + S_0 = 0.021\ Q = 0.32 \text{ Tg a}^{-1}.$$

That is, about 2% of the sulfur remains airborne after 5500 km. Assuming further that the transport from North America hits Europe roughly 50% of the time, the corresponding flux is found by dividing the above quantity by two; thus, the flux of North American anthropogenic sulfur across the European west coast is roughly 0.16 Tg a^{-1}.

The average concentrations of airborne sulfur C_1 and C_2 due to North American emissions can also be estimated. If Europe is assumed to cover an angle $\theta = d/r = 4500/5500 = 0.82$, from eq. (1) we obtain

$$C_1 = 7 \cdot 10^{-3}\ \mu g\ m^{-3} \quad \text{as S},$$
$$C_2 = 0.1\ \mu g\ m^{-3} \quad \text{as S},$$

at the west coast of Europe, averaged over the assumed 50% of the time when the transport is from North America to Europe.

The average deposition D_0 per unit area and time along the coast when this transport occurs can be estimated from these concentrations by means of (8), giving

$$D_0 = 0.042 \text{ gm}^{-2} \text{ a}^{-1}.$$

Table 3. *Parameter values for model calculation*

Symbol	Parameter	Value	
h	mixing height	$2 \cdot 10^3$	m
k_t	transformation rate	$3 \cdot 10^{-6}$	s^{-1}
P_d	Eulerian probability for dry period	0.83	
$T_d(SO_2)$	average residence time with respect to dry deposition (SO_2)	69.4	h
$T_d SO_4^=$	average residence time with respect to wet deposition $SO_4^=$	555	h
$T_d(SO_2, SO_4^=)$	average residence time with respect to wet deposition (SO_2, $SO_4^=$)	47.5	h
u	average wind speed	11	ms^{-1}
v_d	deposition velocity (SO_2)	0.8	cm s^{-1}
w_d	deposition velocity $SO_4^=$)	0.1	cm s^{-1}
α	$\lambda - \bar{\lambda} + k_t$	$6.5 \cdot 10^{-6}$	s^{-1}
β	initial sulfate emission fraction	0.05	
λ	average removal rate (SO_2)	$9.85 \cdot 10^{-6}$	s^{-1}
$\bar{\lambda}$	average removal rate ($SO_4^=$)	$6.35 \cdot 10^{-6}$	s^{-1}
λ_p^{-1}	wet removal rate during precipitation	1	h^{-1}
τ_d	average Lagrangian dry period	50	h
τ_p	average Lagrangian wet period	10	h

Such a long distance away from the source, this deposition will mainly be wet deposition of sulfate. An independent estimate of this deposition can therefore be obtained by assuming a scavenging ratio W of $7 \cdot 10^5$ and a typical annual precipitation amount P_0 of 900 mm. This gives

$$D_0' = C_2 W P_0 = 0.06 \text{ gm}^{-2} \text{a}^{-1},$$

reasonably close to the deposition D_0 above. Note that the implied concentration WC_2 of (anthropogenic) sulfate in rain amounts to 0.07 mg l^{-1} of sulfur, or 4.4 μeq l^{-1}, in close agreement with the value derived from Fig. 4 at this distance (ca. 11 μeq l^{-1} – background). Very rough estimates of the annual deposition of North American anthropogenic sulfur in Europe can be calculated from D_0, recalling the assumption that the transport from North America to Europe occurs only 50% of the time. For Norway and the UK (324 and $224 \cdot 10^3$ km^2 respectively) this deposition amounts to roughly 7000 and 5000 tonnes of S per annum.

5. Concluding discussion

Two distinct approaches have been used to estimate the amount of sulfur carried across the North Atlantic Ocean from North America to Europe. The first made use of available precipitation sulfur data from sites on and adjacent to the North Atlantic. The features of the large-scale average sulfur-concentration pattern can be explained on the basis of known anthropogenic sulfur emissions source distributions on both continents and the average meteorological regime over the North Atlantic. The decrease in concentration with distance from North American sources permits the derivation of a distance decay constant and an average residence time, which in turn leads to an estimate for the amount of sulfur reaching the European west coast. The anthropogenic component of this flux is approximately 0.3–0.4 Tg S a^{-1}.

The second method employs a climatological dispersion model, which accounts for long-term average diffusion, wet and dry deposition, and a linear transformation process for SO_2 to $SO_4^=$. Parameter values used in the model were derived independently of the first approach: average SO_2 residence time = 28 h, average $SO_4^=$ residence time = 44 h, SO_2 to $SO_4^=$ transformation rate = 1% h^{-1}, average wind speed = 11 ms^{-1}, and frequency of westerly flow = 0.5. The flux of North American anthropogenic sulfur across the European west coast was calculated to be approximately 0.2 Tg a^{-1}, in agreement with the first estimate. Use of the same wind speed and average $SO_4^=$ residence time values in the two methods would improve the agreement slightly.

Thus, we conclude that the transatlantic flux of sulfur amounts to a few tenths of a Tg a^{-1}. As discussed in earlier sections, uncertainties in this estimate arise from the precipitation data used, the simplicity of the models, and the parameter values used in the climatological model. Refinement of this estimate and quantification of the uncertainties will require further careful measurements of precipitation and air concentrations over the ocean and at the European and North American coasts, with directional stratification. Vertical profiles of the concentrations of the important sulfur species (and other anthropogenic substances) at various distances from North America over the ocean would help elucidate the important transport levels in the atmosphere. On the modelling side, more comprehensive, physically-based models will be needed; but, in turn, these require a much more detailed meteorological and chemical data base over the ocean than can be anticipated in the near future.

Indirect evidence that the sulfur flux estimates are not unreasonable comes from three sources. Two independent model estimates of S deposition at the European west coast from Section 4.3 are in agreement. Second, the model estimate of the anthropogenic component of $SO_4^=$ in precipitation at the European west coast, 4.4 μeq l^{-1}, from Section 4.3 is in close agreement with the value one can derive from Fig. 4 (ca 11 μeq l^{-1} – background). Finally, using the precipitation field in Fig. 2 along with the over-ocean sulfur concentration profile in Fig. 4, one can derive a wet deposition flux of approximately 5.5 Tg S a^{-1} for a 15° latitude band stretching from North America to Europe. This is more than the total S leaving North America, and indicates that the transatlantic flux is not likely to be larger than estimated.

Based on the precipitation data analysis, the background sulfur concentration in precipitation over the North Atlantic is ca. 6–8 μeq l^{-1}. On average, at the European west coast, the North

American anthropogenic contribution is ca. 3–4 μeq l^{-1}, to make up the total of ca. 11 μeq l^{-1} observed in Fig. 4. The model calculation is similar. Figs. 3 and 4 also show that at most European coastal stations average sulfur concentrations are significantly larger as a result of the influence of European emissions. Therefore, on average, the North American anthropogenic contribution to S in precipitation is relatively small at the European west coast. The widespread influence of European emissions on precipitation composition, even at "clean" coastal sites, emphasizes the need for improved sampling strategies to refine these estimates. As shown by the model calculations, the deposition of North American anthropogenic sulfur in Europe is also relatively small. For Norway and the United Kingdom, for example, it amounts to an estimated 7 and 5 kT a^{-1}, respectively, compared with their total respective depositions of approximately 300 and 1000 kT S a^{-1} (Eliassen and Saltbones, 1983).

This analysis has evaluated only the long-term, climatological precipitation-sulfur concentration field and transatlantic transport. It is important to make a distinction between the average situation portrayed in the above calculations and what happens on shorter time scales. As indicated in Subsections 2.1, seasonal differences in transport are likely because of the different stability and wind regimes over the ocean in winter and summer. As several of the meteorological analyses of single precipitation events referenced earlier have shown, on specific occasions the signal from North American sources in measured concentrations may be strong. An evaluation of the significance of shorter-term contributions must await a more refined analysis.

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