Modelling long-range transport of CFCs to Mace Head, Ireland

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SUMMARY

A Lagrangian particle-based long-range dispersion model is used to describe the transport of inert chlorofluorocarbon (CFC) tracer species to Mace Head, a remote location on the Atlantic coast of Ireland, from emission sources in the European continent and from across the ocean in North America. A six-month analysis covering the period January to June 1996 of model and observed trace-gas concentration reveals a high level of model skill with good correlations and no significant bias in our description of long-range transport over the 100–2000 km scale from Europe to Mace Head. Model simulations have also supported previous suggestions that CFCs detected at Mace Head on two occasions during 1994 were of North American origin. A detailed analysis is made of the meteorological processes involved in one of these transatlantic episodes. We estimate that, on average, North American sources may account for only a few per cent of the CFC-11 and other man-made trace gases observed at Mace Head above the northern hemisphere baseline concentrations. During long-range transport events, North American sources may contribute pollutant peaks at Mace Head which are about one order of magnitude smaller in concentration than those from European sources.

KEYWORDS: Global emissions Long-range dispersion model Pollution

1. INTRODUCTION

The long-range transport of man-made pollutants on a global scale first became evident following the atmospheric testing of nuclear weapons in the 1940’s and 1950’s through the observation of the widespread nature of the resulting fallout. Subsequently, continental-scale long-range transport has been invoked to explain a variety of otherwise unrelated observations including episodes of black snow in 1881 in Norway (Bernes 1991), the deposition of Saharan dusts in the Caribbean (Savoie et al. 1989), the transport of pollen grains from South American 4500 km to the Island of Tristan da Cunha (Gregory 1978), the global increase in carbon dioxide (Keeling et al. 1989), the spread of pesticides such as DDT into the Arctic region (Pacyna and Oehme 1988) and episodes of poor visibility there (Rahn and Heidam 1981). With the commercial availability of perfluorocarbon tracers and highly sensitive electron-capture detectors, it has become feasible to conduct perfluorocarbon tracer dispersion experiments over an increasing range of scales, and detailed studies across the North American continent have been reported (Ferber et al. 1986; Draxler and Heffter 1989). Long-range transport is now well understood as an atmospheric phenomenon and its mechanisms have been elucidated over all scales up to the global scale (Pierrehumbert 1991; Maryon and Buckland 1994).

In this study, our concern is with long-range transport on continental and intercontinental scales, including the suggestion that North American sources of ozone and its precursors—hydrocarbons, carbon monoxide and oxides of nitrogen—may influence levels in Europe. This suggestion has been made following a series of measurement campaigns up the east coast of North America in which a substantial export of ozone and carbon monoxide was quantified into the North Atlantic region during summertime. This led Parrish et al. (1993) to speculate that this export may in turn influence the ozone and carbon monoxide levels over Europe. Furthermore, European pollution sources of ozone and its precursors could conceivably influence Asian levels, and emissions there influence North American levels. Such possible links between emissions in one continent and pollutant levels in another are of significant interest to policy makers. It is widely recognized that

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human activities have led to increased emissions of methane, carbon monoxide and oxides of nitrogen and that these increases may have more than doubled tropospheric ozone levels over Europe since pre-industrial times (Volz and Kley 1988; Hough and Derwent 1990). If this increase has been northern hemispheric in scale then it would have made a significant contribution to the estimated increase in the greenhouse effect since pre-industrial times (Houghton et al. 1996). The magnitude of possible intercontinental contributions to any ozone increases therefore becomes of interest to policy makers concerned with regional-scale ozone pollution problems.

The initial aim of the present study was to assess the UK Meteorological Office’s long-range dispersion model, NAME, as a means of investigating the long-range transport of chlorofluorocarbons (CFCs) from Europe and America by comparing model predictions with observations at Mace Head on the Atlantic coast of Ireland. Previous modelling of long-range transport to Mace Head has been based on climatological data (Simmons et al. 1996) or on back-trajectory analyses. In contrast NAME attempts to model the transport of CFC’s directly using analysed meteorological fields from the UK Meteorological Office’s numerical weather-prediction model. A detailed analysis was also made of two possible transatlantic episodes, to establish whether CFCs detected at Mace Head during these periods were in fact of North American origin. The opportunity was also taken to attempt to throw some light upon the synoptic machinery underlying episodes in which North American sources lead to higher-than-background concentrations of trace gases in western Europe.

In section 2 an account is given of the measurement, and previous efforts at source attribution, of trace gases at Mace Head. Section 3 gives a brief introduction to the processes governing dispersion over long range in the atmosphere. Section 4 describes the numerical dispersion model applied in the present study, and section 5 the results of a continuous six-month simulation of the combined emissions from Europe and North America. Section 6 concentrates on two possible North American episodes, those of March and November 1994, and includes a detailed analysis of the meteorological background to the March episode. Further discussion and the conclusions are contained in section 7.

2. Observations of Trace Gases at Mace Head, Ireland

Man-made halocarbons and radiatively active trace gases have been monitored continuously at Mace Head, Ireland at two-hourly intervals since 1987 and forty-minute intervals since 1994 (Simmons et al. 1996). The Mace Head monitoring site is situated on the Atlantic Ocean coastline (53.32°N, 9.90°W) and receives air masses with an uninterrupted fetch over the ocean for about two-thirds of the time. The trace gases monitored include the major chlorofluorocarbons (CFC-11, -12 and -113), the halocarbons (carbon tetrachloride, methyl chloroform and chloroform), the radiatively active gases (carbon dioxide, methane, nitrous oxide and ozone) and the ozone precursor (carbon monoxide).

Three independent methods have been used to sort the continuous monitoring data into broad air-mass origins. These methods include sorting based on the concentrations of man-made halocarbons (Rasmussen and Lovelock 1983); isentropic back-track trajectories (Merril 1994) and daily wind sector allocations provided by the European Monitoring and Evaluation Programme Meteorological Synthesizing Centre-West in Oslo (Lemhau et al. 1988). In this way, the trace-gas concentrations associated with northern hemisphere mid-latitude background air have been separated from those concentrations associated with air of European origins. The trends in the long-term mean concentrations of most man-made species appear to be significantly different in each of the air-mass regions (Simmonds et al. 1996).
One feature of the Mace Head monitoring record has been the lack of an obvious contribution from North American pollution sources. Following the suggestions made by Parrish et al. (1993) concerning the export of ozone and carbon monoxide into the North Atlantic region, a more detailed examination of the Mace Head records was made. The first approach used air-mass back-trajectories to identify the difference in ozone and carbon monoxide concentrations associated with those trajectories which passed over Canada and the United States of America compared with those passing over Greenland and Iceland. Four years of data were analysed, but the monthly mean differences attributable to Canada and the United States were small and the analysis was inconclusive (Derwent et al. 1998).

These initial results suggested that North American pollutant emissions become well dispersed into the mid-latitude background on the time-scale of the transport to Europe from North America. A possible explanation is that convection over the North American continent efficiently transports surface man-made emissions into the free troposphere, thus diluting pollutant concentrations sufficiently for them to be undetectable above background levels following transport across the North Atlantic. Other processes will be examined in this study.

In a second approach, the air-mass back-trajectories to Mace Head were sorted according to a stricter set of criteria which required: (i) trajectories to join up the eastern seaboard of North America with Europe, (ii) the trajectory to be wholly in or close to the boundary layer throughout its length, (iii) the trajectory not to have passed over any part of Europe before arrival at Mace Head. This approach identified five occasions during the period 1990 to 1994 where material detected at Mace Head may have originated from North American sources. Of these periods, one during December 1992 has already been identified by Bandy et al. (1994) as including a long-range transport event from the North American continent. An additional event has been identified during April 1992 from measurements of black-carbon aerosol and carbon monoxide (Jennings et al. 1996).

3. **Dispersion over long range**

The processes by which turbulent diffusion occurs in the atmosphere have been exhaustively studied, and are described in many texts (see, for example, Pasquill and Smith (1983)). Diffusive spread occurs wherever a plume of suspended material much exceeds in size the scales of the eddy motions. It is characterized by the transfer of energy to smaller scales in an inertial subrange. Turbulent diffusion is relatively important for the first 12 hours at least after material is released into the atmosphere and, depending upon the meteorological conditions, occasionally for as long as 48 hours. It is usually assisted by vertical wind shears in spreading material in the early stages. Sooner or later, however, the dispersion comes to be dominated by the largely two-dimensional processes of differential or chaotic advection: the material is stretched and folded with a tendency to exponential separation of initially juxtaposed parcels (Lin 1972). This mechanism is also known as the enstrophy cascade process. Characteristic structures are the drawing out of filaments of pollutant into 'tendrils', or, in the vicinity of a slow-moving depression, winding the material up into a whorl, as is commonly exhibited in satellite cloud photographs. The two-dimensional stretching and folding can lead to a lamellar structure where adjacent striations incorporate material from different origins (Ottino 1991)—a process often simply described as mixing or stirring. These contrasts can be blurred by the continuing operation of diffusion which, however, remains subordinate to the large-scale processes. Less well understood is the dispersive influence of intermediate scales (say between one and a few hundred km) characterized by upscale transfers of energy; here the tendency to transport or diffuse material must depend on the relative scales of the plume and the
motion systems. A description of the dispersive machinery operating in the troposphere for a week or two following a release is given in Maryon and Buckland (1994).

In the present study we are presented with a situation in which multiple areal sources of a contaminant are mixed in the early stages, over North America and adjacent sea areas, helped by the diffusive processes of the energy cascades. Eventually large amounts of material are caught up in broadly westerly flows and transported across the Atlantic Ocean, the cloud undergoing, meanwhile, the deformations associated with the enstrophy cascade process. The motion scales of the reverse, upscale energy cascade may be of critical importance here, as they will be associated with synoptic and subsynoptic transports with a vertical component; vertical exchanges of material may play a key role in the delivery (or otherwise) of significant quantities of material to the observing site.

4. THE NAME LONG-RANGE DISPERSION MODEL

The UK Meteorological Office's long-range dispersion model, NAME, was developed following the Chernobyl incident to simulate the medium- and long-range transport of airborne pollutants (Maryon et al. 1991). The model provides estimates of air concentrations, and the deposition of pollutants to the ground by both wet- and dry-deposition processes. NAME is implemented operationally as an emergency response model, and, as a World Meteorological Organization Regional Specialist Meteorological Centre, the UK Meteorological Office has international responsibilities for providing forecasts of the spread of radionuclides or other hazardous materials in the event of a major atmospheric release. Unfortunately, only limited high-quality observational data is available for assessing long-range models; the model has, however, been applied with some success to the Chernobyl incident (Ryall et al. 1994) and to the Kuwait oil fires (Browning et al. 1991). Early results from the European Tracer Experiment (Maryon and Ryall 1996) are also encouraging.

The model is of a Lagrangian type, in which emissions are modelled by releasing large numbers of 'particles' into the 'model atmosphere'. The particles are carried along passively by the ambient three-dimensional wind, with turbulent dispersion simulated by random walk or Wiener techniques. Each particle represents a mass of released pollutant, which is reduced over time by both wet- and dry-deposition processes if applicable. All meteorological data, such as wind and temperature fields, precipitation, surface heat flux etc., are obtained from global, regional and mesoscale versions of the UK Meteorological Office's numerical weather-prediction model, the Unified Model (Cullen 1993). These provide data at six-, three- and one-hour intervals, with horizontal resolutions of about 90, 50 and 16 km, respectively. A nested structure is used, whereby the meteorological data used are taken from the highest resolution data available at that time and place. In this study assimilated meteorological data at the regional scale were used. The vertical coordinate of the Unified Model is the hybrid η (eta) coordinate, which graduates from terrain-following in the lower atmosphere to constant-pressure in the upper atmosphere.

(a) Advection and diffusion

The NAME model has a range of options available for advection and diffusion. Here we give a brief description of those schemes used in this study. More detailed descriptions of the parametrizations available to NAME are given in Physick and Maryon (1995) and Ryall and Maryon (1996).

Particles are advected each time-step using

\[ x_{t+\Delta t} = x_t + \{u(x_t) + u'(x_t) + u''(x_t)}\Delta t, \]
where \( \mathbf{x} \) is the particle position vector, \( \mathbf{u}(\mathbf{x}) \) is the wind velocity vector, \( \mathbf{u}'(\mathbf{x}) \) is the turbulent velocity vector for small-scale turbulence, \( \mathbf{u}'(\mathbf{x}) \) is the velocity vector for low-frequency horizontal meandering, and \( \Delta t \) is the time-step.

The horizontal turbulent velocity components are from the standard random-walk formulation:

\[
\begin{align*}
\mathbf{u}'_{t+\Delta t} &= \mathbf{u}'_t \left( 1 - \frac{\Delta t}{\tau_u} \right) + \left( \frac{2\sigma_{u,2}^2 \Delta t}{\tau_u} \right)^{1/2} \mathbf{r}, \\
\mathbf{u}'_{l,t+\Delta t} &= \mathbf{u}'_{l,t} \left( 1 - \frac{\Delta t}{\tau_{u,l}} \right) + \left( \frac{2\sigma_{u,l,2}^2 \Delta t}{\tau_{u,l}} \right)^{1/2} \mathbf{r}_l,
\end{align*}
\]

where \( \mathbf{r} \) and \( \mathbf{r}_l \) are random Gaussian variables of zero mean and unit variance, \( \tau_u \) and \( \tau_{u,l} \) are the Lagrangian time-scales for the \( x \) component of turbulence and meandering respectively, and \( \sigma_{u,2}^2 \) and \( \sigma_{u,l,2}^2 \) the corresponding horizontal velocity variances. The expressions for \( v' \) and \( w' \), the \( y \) and \( z \) components respectively, are similar, though \( w'_l = 0 \). Note that the turbulent velocities consist of two components, the first term representing a "memory", or damping, of previous motion and the second term an innovation. The vertical component includes an additional term, the drift velocity, designed to prevent model particles accumulating at levels of small \( \sigma_w \).

The time-step is chosen to be short in comparison with the Lagrangian time-scale,

\[ \Delta t = e_1 \tau_u, \]

where \( e_1 \) is a small number such as 0.05 or 0.1.

(b) Diffusion coefficient scheme

These random-walk schemes were developed to simulate near-source diffusion, and are computationally very expensive. For greater economy a simplified scheme based on diffusion coefficients can be employed. The \( u' \) turbulent component for small-scale turbulence is determined from

\[
\mathbf{u}' = \sqrt{\left( \frac{2K_u}{\Delta t} \right)} \mathbf{r},
\]

where the diffusion coefficient \( K_u \) is determined from the turbulent velocity variance and the Lagrangian time-scale:

\[ K_u = \sigma_u^2 \tau_u. \]

Similar expressions are used for the \( v' \), \( w' \), \( u'_l \) and \( v'_l \) components. This is a Wiener diffusive process resulting in a parabolic spread, which experience has shown to be an adequate scheme for dispersion modelling at long range.

(c) Turbulence profiles

The random velocity components are functions of the vertical profiles of the vertical velocity variances and Lagrangian time-scales, which are derived from published empirical fits to observational data, combined with Unified Model products such as the friction velocity \( u_* \), surface temperature \( T_s \) and surface heat flux \( H \). Separate formulae are used for stable and unstable conditions. For unstable boundary layers these are based on the turbulence profiles of Hibberd and Sawford (1994), with a mechanical component (and neutral limit) from Brost et al. (1982). For stable boundary layers the profiles of Hanna (1982)
TABLE 1. HOMOGENEOUS TURBULENCE STATISTICS USED IN STUDY.

<table>
<thead>
<tr>
<th>Stable ABL</th>
<th>Unstable ABL</th>
<th>Above ABL</th>
<th>Meander (ABL only)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \sigma _{u} , \sigma _{c} )</td>
<td>( u _{*} )</td>
<td>( (0.4w ^{2} _{<em>,} + 3u _{</em>} ^{2})^{1/3} )</td>
<td>0.25 m s(^{-1})</td>
</tr>
<tr>
<td>( \sigma _{w} )</td>
<td>( 0.65u _{*} )</td>
<td>( (0.4w ^{2} _{<em>,} + 1.1u _{</em>} ^{2})^{1/3} )</td>
<td>0.1 m s(^{-1})</td>
</tr>
<tr>
<td>( \tau _{u} , \tau _{c} )</td>
<td>( 2\sigma _{u} /C _{0})</td>
<td>( e = 0.6w ^{2} _{<em>,} /z _{l} + 1.2u _{</em>} ^{2} /k z _{l} )</td>
<td>300 s</td>
</tr>
<tr>
<td>( \tau _{w} )</td>
<td>( 0.05 \frac{\gamma _{l}}{\sigma _{w}} )</td>
<td>( 2\sigma _{w} /C _{0})</td>
<td>( e = 0.6w ^{2} _{<em>,} /z _{l} + 1.2u _{</em>} ^{2} /k z _{l} )</td>
</tr>
</tbody>
</table>

ABL = atmospheric boundary layer, \( z _{l} \) is the boundary-layer depth, \( t \) is time between model fields and \( u _{10} \) is the 10 m wind velocity, the convective velocity scale \( u _{*} = u _{*}(z _{l} /|L|) \).

are used. In the long-range scheme, simplified homogeneous turbulence profiles are used whereby the turbulence is assumed uniform within the boundary layer. The homogeneous values are determined from the mid-boundary-layer values of the inhomogeneous profiles. Table 1 summarizes the homogeneous profiles which were used for this study.

The stability of the boundary layer is determined from the Monin–Obukhov length

\[
L = \frac{\rho c _{p} T_{n} u _{*} ^{3}}{kg H},
\]

where \( \rho \) is air density, \( c _{p} \) is the specific heat at constant pressure, \( k \) is von Karman’s constant and \( g \) is the acceleration due to gravity. Negative values of \( L \) indicate unstable boundary layers and positive values indicate stable boundary layers.

An economical option of reflecting particles from the top of the boundary layer and the surface was adopted, so that entrainments across the inversion capping the boundary layer occur through the change in the boundary-layer depth from one model time-step to the next, or by particles being advected to regions of differing boundary-layer depths.

For this study the fully memory-based scheme (2) was applied for meandering, with the simplified far-field \( K \)-based scheme (3) for the turbulent components. As the species being considered in this study are inert, no deposition or loss processes were applied.

\( d \) Boundary-layer depths

The correct determination of the boundary layer depth, \( z _{l} \), is crucial for modelling the dispersion of airborne pollutants; for example, turbulence is usually greater in the boundary layer than in the free troposphere, resulting in more rapid horizontal and vertical diffusion, but usually slower transports. In NAME, boundary-layer depths are directly calculated from Unified Model wind and temperature profiles, using either a Richardson-number or parcel technique.

In the Richardson-number technique the presence of turbulent motion is inferred from the value of the gradient Richardson number, \( Ri \), which is calculated from

\[
Ri = \frac{g \partial \theta / \partial z}{T (\partial u / \partial z)^{2}},
\]

where \( \partial \theta / \partial z \) and \( \partial u / \partial z \) are gradients of potential temperature and wind, respectively, and \( T \) is temperature. This is calculated for each model level starting at the surface until the
Richardson number exceeds a critical value, \( R_i \), taken as 1.3. This high value is derived from earlier investigations, and is intended to offset the loss of detail in the model profiles due to discretization.

In the parcel method the boundary layer is assumed to be the depth of atmosphere over which potential temperature is constant. This is found by following the dry adiabatic lapse rate (DALR) from a near-surface (1.5 m) temperature, and determining the height at which it intersects the model environment curve (as defined by the model temperature profile). A detailed description of both techniques is contained in Maryon and Best (1992).

The boundary-layer depth is taken as the maximum of the Richardson-number and parcel-method values. This generally results in the Richardson-number method being used in stable conditions and the parcel method in unstable conditions. A minimum boundary-layer depth of about 80 m is used, and a maximum boundary-layer top of 5000 m.

\[ (e) \quad \text{Attribution} \]

When studying atmospheric pollution problems it is important to be able to establish the particular sources that contribute to pollution levels at a given receptor. In NAME, particles are labelled with their location and time of origin, so it is possible to identify the various sources that contribute to a defined receptor and quantify their relative contributions.

5. LONG-TERM SIMULATIONS

\[ (a) \quad \text{Model set-up} \]

A continuous six-month simulation was performed covering the period January to June 1996. Both US and European sources of CFC-11 were represented within a model domain [90°W to 40°E, 25°N to 80°N]. Emissions on a \( 1^\circ \times 1^\circ \) grid were based on the global fluorocarbon inventory of Prather et al. (1987), scaled by the release rates in each country and weighted within each country by population density (McCulloch et al. 1994). The emissions were then scaled to give a total European contribution of 20 000 tonnes per year, as estimated by Simmonds et al. (1996) for 1994 emissions. A constant release rate is assumed throughout the year and all sources are assumed to be surface releases. The initial coordinates of a particle are chosen randomly from within each \( 1^\circ \times 1^\circ \) source area.

A cut-off release rate is defined such that particles are only released from sources that contribute in excess of 0.1% of the global total. This results in 63% of the emissions being represented, but reduces the number of \( 1^\circ \times 1^\circ \) sources required from 1203 to just 147. This concentrates the available particle numbers on the small number of major sources with high emission densities, which represent the bulk of the emissions. As the remaining emission cells are distributed over a much larger area with lower emission densities, their individual contribution to Mace Head concentrations is likely to be small.

Boundary-layer mean concentrations were calculated every 15 minutes (the model time-step used) over a grid volume defined by the boundary-layer depth and an area \( 1.25^\circ \times 0.833^\circ \), centred on Mace Head. These model concentrations were compared with observations of CFC-11 taken at 40-minute intervals at Mace Head. For statistical comparisons, model values were linearly interpolated in time to correspond with the observation. A three-hour moving-average filter was applied to both model and observed data to reduce noise. The comparisons are made with the short-term fluctuations observed above background levels, which represent years of emissions since industrial manufacture began in the 1930's.
Results

Model and observed time series are shown in Fig. 1(a) and a cross plot showing the best-fit line in Fig. 1(b). Observed data are plotted inverted for easy comparison, and with the background concentration level subtracted (assumed constant and taken as the minimum observed value during the period). The overall correlation between observed and model traces is good, with a product moment correlation coefficient of 0.841. The model appears to reproduce both the timing and shape of most of the main features observed during the six-month period, although there are cases where observed peaks were not modelled and modelled ones not observed. Overall the model seems to overestimate concentrations, with the best-fit line suggesting a factor of 1.699.

A high degree of confidence can be placed on the accuracy and reliability of the observations which are reckoned to be accurate to ±0.2%, 0.5 parts per thousand (p.p.t) (Cunnold et al. 1994), so that differences between the model and observed data are predominantly the modelling errors and source terms. The main problem areas can be identified as:

(i) Errors in the distribution, timing and magnitude of emissions. CFC emissions are falling rapidly as legislation banning their manufacture and use takes effect, so that actual emissions are likely to be lower than those assumed, with the reduction varying from country to country. It is also probable that CFC-11 emissions vary in time, depending on time of day and year, though such variations have yet to be quantified.

(ii) Errors and limitations in the meteorological data. Analysis fields from numerical weather-prediction models can only approximate the meteorological situation, particularly over data-sparse regions such as the North Atlantic Ocean. In addition, the resolution of model data (50 km in this study) limits the scale of features represented.

(iii) Limitations in model physics. A range of assumptions is made in the model advection and diffusion schemes. In addition important processes such as venting and mixing by deep convection are not well represented by the model.

(iv) Limitations in the model configuration. Owing to the inert nature of CFCs it is possible that sources from outside the computational domain or released prior to the start of the simulation may have contributed to concentrations at Mace Head. Problems can arise from using too few particles to represent a source, as this can result in insufficient particle densities to represent the shape of the plume properly. By using a cut-off release rate a number of smaller sources are ignored, which may result in the model missing or under-predicting some pollution peaks. The resolution of the analysis grid can also have an effect.

Despite the range of potential problems and errors, the match between model predictions and observations is encouraging, suggesting that NAME represents the important mechanisms involved.

Estimating emissions

If we assume that limitations in the meteorological data and model physics do not result in systematic errors, then any overall bias between observations and model data would be mostly due to errors in the magnitude of emissions. If we further assume that the distribution of emissions is reasonably accurate we can then make an estimate of the actual emissions by scaling the emissions by the ratio of model to observational data (as determined by linear regression). Considering European contributions only, this results in estimated CFC-11 emissions in Europe for 1996 of 11.8 thousand tonnes per year, a little over half (1/1.699) the emissions estimated by Simmonds et al. (1996) for 1994. Given the
Figure 1. Comparison between model-predicted and observed chlorofluorocarbon CFC-11 concentrations at Mace Head: (a) the time series, with the observed data inverted and background subtracted (see text), and (b) a cross-plot and best-fit line. Observed data are at 40 min intervals, and a three-hour smoothing filter is applied to both observed and model data.
TABLE 2(a). Correlations and estimated European emissions for trace gases which have been phased out.

<table>
<thead>
<tr>
<th>Species</th>
<th>Correlation with Model CFC-11</th>
<th>European Emissions (NAME Model) (kT yr⁻¹)</th>
<th>European Emissions (Simmonds et al. 1996) (kT yr⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CFC-11</td>
<td>0.841</td>
<td>11.8</td>
<td>18.7-20.8</td>
</tr>
<tr>
<td>CFC-12</td>
<td>0.806</td>
<td>21.2</td>
<td>25.9</td>
</tr>
<tr>
<td>CFC-113</td>
<td>0.719</td>
<td>9.00</td>
<td>14.7</td>
</tr>
<tr>
<td>Methyl chloroform</td>
<td>0.754</td>
<td>42.9</td>
<td>82.8</td>
</tr>
<tr>
<td>Carbon tetrachloride</td>
<td>0.735</td>
<td>8.23</td>
<td>5.6</td>
</tr>
</tbody>
</table>

TABLE 2(b). Correlations and estimated European emissions for radiatively active gases and chloroform.

<table>
<thead>
<tr>
<th>Species</th>
<th>Correlation with Model CFC-11</th>
<th>European Emissions (NAME Model) (kT yr⁻¹)</th>
<th>European Emissions (Simmonds et al. 1996) (kT yr⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon monoxide</td>
<td>0.826</td>
<td>108 000</td>
<td>53 000-108 000</td>
</tr>
<tr>
<td>Nitrous oxide</td>
<td>0.802</td>
<td>2690</td>
<td>1300-2700</td>
</tr>
<tr>
<td>Methane</td>
<td>0.828</td>
<td>40 720</td>
<td>22 600-48 000</td>
</tr>
<tr>
<td>Chloroform</td>
<td>0.510</td>
<td>35.77</td>
<td>not available</td>
</tr>
</tbody>
</table>

rate of decline of CFC-11 emissions found by Simmonds et al. (1996) over the 1987–1994 period, this figure seems reasonable, and cannot be taken to indicate significant bias in the model.

It is interesting to compare the model CFC-11 predictions with observations of other species at Mace Head. Table 2 lists the correlations obtained and the estimated emissions. Figure 2 compares scaled model products with observations for each of the species.

The correlations obtained for carbon monoxide, methane, nitrous oxide and CFC-12 of 0.825, 0.828, 0.802 and 0.806, respectively, are close to the CFC-11 correlation of 0.841. This suggests that the distribution of emissions used for CFC-11 also represents the distribution of emissions for these species. This is not unreasonable given that the distribution is based on industrial output and population density. Correlations with CFC-113, methyl chloroform and carbon tetrachloride are slightly lower but still fairly good, ranging from 0.719 to 0.754, again indicating emission distributions broadly similar to CFC-11. In contrast the chloroform correlation is poor at 0.510. Inspection of the chloroform observations reveals that chloroform peaks are observed on many occasions where the model predicts low levels. The diurnal nature of many of these peaks (see 29 April to 20 May, Fig. 2) suggests that local sources (not included in the model inventory and not important for the major man-made halocarbons) may be present. These diurnal peaks can also be seen on the methane trace. These local sources are the subject of a separate investigation.

Also listed in Table 2(a) are 1994 emissions estimated by Simmonds et al. (1996) using a much simpler long-range transport model. With the exception of carbon tetrachloride these are all higher than the model-derived values for 1996. This is reasonable, as lower values should be expected for 1996 as the manufacture of CFC's has been phased out by international treaty (World Meteorological Organization 1988). Estimated emissions
for the radiatively active trace gases, methane, nitrous oxide and carbon monoxide, show broad agreement, falling at the high end of the Simmonds et al. values (Table 2(b)). They also show broad agreement with independent emissions from the European Commission CORINAIR study (McInnes 1994) of 45 000, 1900 and 69 000 thousand tonnes per year, respectively.

Despite the uncertainties in current emission inventories and the assumptions made in deriving emission estimates (such as assuming the same emission distribution as CFC-11), the broad agreement between model-derived emissions and other estimates is encouraging, indicating no significant bias in our model estimates of long-range transport and dispersion over the 100–2000 km scale.

(d) The North American contribution

The proportions of the predicted air concentration due to North American sources are shown in Fig. 3. Whilst European sources clearly dominate, the model predicts regular low-level contributions of up to 2.0 p.p.t. from North America, especially between major European episodes where the prevailing winds at Mace Head are predominantly westerly. On a number of occasions, material is attributed both to North America and European sources at the same time, resulting from the mixing of air from both Europe and North America prior to arrival at Mace Head.

During this six-month period the percentage of Mace Head material originating from North American sources averaged 4.2%, though this figure varies from week to week depending on the dominant weather patterns. For example, in the week starting 22 January 1996, less than 0.25% is attributed to North America, but in the week starting 19 February 1996, over 58% was attributed to North American sources.

This implies that low, but potentially significant, levels of a range of pollutants may reach Western Europe from North America, especially those species which are chemically inert and are not readily removed by wet- and dry-deposition processes.

6. The episodes of March and November 1994

The second part of this study was to investigate two potential transatlantic episodes in some detail: 21 March 1994 and 12–13 November 1994. For each episode two simulations were performed, starting at least eight days before the potential event. One simulation covered the domain [110°W to 10°E, 20°N to 75°N] with North American emissions only, and the second covered the domain [20°W to 40°E, 30°N to 75°N] with European emissions only. The aim of performing separate North American and European model runs was to distinguish clearly material arriving at Mace Head as of US or European origin. Again all simulations were performed using Unified Model regional meteorological data.

In this study the cut-off rates for CFC-11 were 0.03% of global emissions for North American sources and 0.05% for the European sources, resulting in approximately 87% and 83% of North American and European emissions, respectively, (within the computational domain) being represented. Additional comparisons were made against observations of CFC-12, CFC-113, methyl chloroform and carbon dioxide. Model concentrations for the different species were determined by scaling the global CFC-11 predictions according to the relative emissions calculated in section 5. The same emissions distribution was assumed for carbon dioxide, but scaled to give a North American emission of 5 346 000 thousand tonnes per year (Marland et al. 1994).

To illustrate the distribution of material at a given time, both horizontal and vertical particle plots are used. These show the instantaneous position of all particles at the time of
Figure 2. Model versus observed concentrations at Mace Head for a range of species. Model curves are CFC-11 predictions scaled to obtain best-fit to observations, and observed data are inverted.
the plot (i.e. particle trajectory end points), and give a detailed picture of the distribution of particles.

(a) *March 1994 episode*

In Fig. 4 observed air concentrations are plotted against those predicted by the NAME model. Note that the vertical axis minima and maxima are chosen arbitrarily to display the

![North American Component of CFC-11 Air Concentration at Mace Head](image)

Figure 3. Model-predicted CFC-11 concentrations due to North American sources only.
observational data—the minima do not represent the baseline for that species. The model data is presented relative to the scale minima used. The model clearly indicates a broad peak during the 21 March 1994, originating from North American sources only. Unfortunately there is a period of missing halocarbon monitoring data from 0914 to 1753 UTC on the 21st which, with the relatively low levels of pollutants observed above background and noise levels, makes comparisons difficult. However, there is clear evidence of a rise in CFC-11 levels in the early hours of the 21st, and a fall in the early hours of the 22nd, agreeing well with the model timings. A complete observation record was available for CO₂, which shows a clear peak with the same timings. Whilst it is recognized that CO₂ is
not an inert tracer like the other gases, it is unlikely to have suffered significant depletion over the North Atlantic. The CO$_2$ trace exhibits a shorter peak from 0900 to 1200 UTC on 20 March, though this is not observed on any of the other trace-gas concentration records.

This relatively clear-cut episode lent itself to an analysis of the meteorological factors controlling the direct transport of pollutant across the North Atlantic.

**(b)** A synoptic view of the episode of March 1994

The transport and dispersion as interpreted by the NAME model is examined here in the light of the evolving synoptic patterns. Of necessity this is a much simplified account of synoptic developments during ten days or so of disturbed North Atlantic weather, and of three-dimensional plume configurations which were highly complex, and evolving rapidly in time. For convenience, some of the depressions and weather fronts have been labelled with letters: these identifiers are unrelated to any published by national meteorological services. The important sources of CFCs used in the modelling of this event were the line of cities along the east American seaboard, Washington to New York, and the industrial areas fringing the Great Lakes (Chicago, Detroit, Cleveland and Buffalo).

(i) The early stages—13–17 March 1994. The period 13–17 March 1994 was one of persistent cols, ridging or light winds over the source region, with the result that large amounts of material collected between New England and the Carolinas. Some of the material drifted north-east towards Newfoundland, was caught up in a frontal system, and ascended to engage a strong westerly flow aloft, over the North Atlantic. This high-level material was swept rapidly east-south-east, crossing the south-west of Britain, the tendril ultimately extending from Hudson Bay to central Europe—the first instalment of successive transports of material along this mid-sometimes upper-tropospheric route. A second strand was wound into a depression over the Atlantic east of Chesapeake, and was again lifted, reaching 500 hPa by midday on 15 March, and with sparse material near the tropopause a day later. Both these transports are well shown in Fig. 5(a). For our purposes, however, the important feature was the great mass of pollutant ‘ponded’ over the seaboard states which was transferred into the Atlantic in two phases. In phase 1, the material south of Long Island (roughly) was pushed south-east by the flow around a deep new low centred east of Chesapeake (low E), the forward edge of the pollutant cloud coincident with—and delineating—a cold front (front F). As low E drifted north-east to Nova Scotia, the northern part of the reservoir (phase 2 material, north of Long Island) was engaged, and swung south-east into the Atlantic behind a second cold front (front G). The two phases were linked by dense strands of material around 40°N. Cross-sections show uplift of pollutant to 500 hPa or more associated with the frontal activity. Figure 5(a) illustrates the situation at this point (1200 UTC 17 March).

(ii) The Atlantic passage—18–20 March 1994. By 18 March high pressure over the eastern North Atlantic Ocean had sunk southwards to allow a zonal, broadly west-southwesterly flow which would sweep the pollutants very directly towards western Europe. A new low (H) had developed off Newfoundland in the circulation of low E by the 18th, around which ships reported 25 to 45 kt surface westerlies. The phase 1 material aligned with cold F reached 33°W by 1200 UTC 19 March and 23°W by 0000 UTC on the 20th. Phase 1 was to play no great role in the episode, however: overtaken by the dynamics of front G, perhaps, the material along F was stretched northwards, attenuated, and lifted. The interlinking material behind F became indistinguishable from the pollutant masses associated with the advancing cold G (1200 UTC 18 March, Figure 5(b)). The south to north uplift, evidently a ‘conveyor belt’ feature (Browning 1985) ahead of G, is clearly
Figure 5. Particle positions predicted by the NAME model and observed mean sea-level pressure pattern at 1200 UTC (a) 17 March 1994, (b) 18 March, (c) 19 March, (d) 20 March, and (e) 21 March (boundary-layer particles only). Letters denote synoptic features of the pressure pattern which is detailed in the text.
Figure 5. Continued.
shown in the NAME output, implying that the Unified Model winds interpreted the broader features of the frontal dynamics well. Some of this high-level material was carried back to the north and west around the persistent lows in the Labrador/Newfoundland area. Over the Atlantic, cold G, with phase 2 pollutant, gradually caught up with cold F, although a thin band of phase 1 material remained distinguishable for some time after F was dropped from the synoptic charts. Boundary-layer material reached 15°W by 1200 UTC on the 20th (Fig. 5(d)).

Meanwhile, an interesting development took place behind phase 2: yet another low (I) developed off Delaware and swung north towards the persistent low-pressure areas around Labrador (Fig. 5(c)). During the formative stage, pressure was again slack over the source regions, allowing further pollutant collections (phase 3) which, like the earlier phases, were pulled south-eastwards into the Atlantic as low I developed. Between phases 2 and 3 a warm sector (warm J, cold K) associated with low I pushed north-east over the north-west Atlantic during 19 and 20 March, containing relatively sparse pollutant concentrations: clearer tropical maritime air (but incorporating some older residues from earlier North American emissions) had been enfolded into the highly polluted air off the Continent (Figs. 5(c) and (d)). The cool north-westerly outflows from America had also had the effect of deepening the boundary layer over parts of the North Atlantic: values had increased from about 400 m, through 800 m in phase 2 to as much as 1600 m in the phase 3 areas behind cold K. Plume cross-sections frequently show a very sharp increase in boundary-layer depth as the air moves from land to sea. A weak ridge developed between G and warm J in mid-ocean, containing dense masses of phase 2 material, with some at high level (above 300 hPa). The upper-level portion probably derived from pronounced uplift apparent along
front K, where the upper flow no doubt formed part of a warm conveyor belt, sweeping around the top of the wave and above the ridge (1200 UTC 20 March, Fig. 5(d)). If that were so, it constituted a phase 3 contribution at high level.

A cross-section along 43°N at 1200 UTC 19 March (Fig. 6(a)) intersects a thin band of phase 1 material at 33°W, with dense phase 2 boundary-layer material to about 47°W. High-level material associated with the J–K system is evident at about 60°W (and further north there is much uplifted material 55–75°W associated with the persistent depressions of eastern Canada).

The cross-section for 1200 UTC 20 March (Fig. 6(b)) illustrates the complexity of the Atlantic-wide plume. The phase 1 material had now mostly dispersed; the phase 2 forward slope was about 1:45—slopes in the cross-sections no doubt often reflect wind shears in the vertical or a history of ascent along a frontal surface or conveyor. The cross-section suggests the densest phase 2 boundary-layer material was around warm J at about 27°W; the western parts of the cross-section again show old upper-level material engaged by low L and its predecessors in that area. There is perhaps an impression of descent, or at least suppressed vertical motion, behind cold G in these figures.

(iii) The episode at Mace Head—21–23 March 1994. Unless otherwise stated, the descriptions in this section refer to the simulation of events at Mace Head. For this episode we are primarily interested in boundary-layer pollutant, and can make the assumption that the boundary layer is well mixed following the Atlantic crossing, even given a history of vertical transports and interchanges. The time series of modelled air concentrations (Fig. 4) essentially reflects only the presence of particles in the grid cell containing Mace Head, not events at the station itself. The first particles reached the grid cell by 2000 UTC 20 March, as the boundary-layer depth in the cell was descending to an overnight value of 400 m. The dense ‘front’ of phase 2 material was close to Mace Head itself by midnight on the 21st, while some was pulled south-east towards Land’s End, and later Biscay, by a depression (low L) which had formed over the South-West Approaches and drifted into France. The cloud was especially dense at 15–20°W at this time, where it was organized by warm J. Behind J a tongue of tropical maritime air had produced a relatively clear slot ahead of phase 3 material; an incursion of clean boundary-layer air eventually extended north to 60°N.

By 1200 UTC 21 March (Fig. 5(e); note this is restricted to boundary-layer particles) the episode had reached its peak. Warm J had closed with G by this time; the forward edge of the (phase 2) boundary-layer material seemed to lie along G as always, as far as G can still be identified on synoptic charts: the particle plot may even give an improved analysis! These forward G residuals were aligned Iceland–Outer Hebrides and north—south through central Ireland. The densest material was close behind on J, especially north from County Mayo. The peak concentration values were measured in the Mace Head grid cell towards 1600 UTC, as the boundary-layer depth fell to its lowest value around 250 m (from an earlier 600 m)—no doubt the fall was associated with the arrival of maritime air and relative stability. The cross-section (1200 UTC 21 March, Fig. 6(c)) shows the surface and perhaps upper frontal positions over the British Isles and the clear slot of tropical maritime air 20–25°W; phase 3 material was evident behind cold K. During the 21st, the modelled concentrations show a strong inverse correlation with the boundary-layer depth at Mace Head, although it is likely that both are reflecting frontal passages rather than exhibiting dilution phenomena.

On 22 March a very strong south-westerly flow around what had become a major Icelandic low engulfed the British Isles, sweeping the weather systems through (low I attained depths of about 940 hPa). It was increasingly difficult now to relate the distribution
Figure 6. Vertical cross-sections showing particles at 1200 UTC in the regions (a) 42–44°N, 19 March 1994, (b) 49–51°N, 20 March, and (c) 55–57°N, 21 March. Darker shading indicates boundary-layer particles.
of the material to the historic synoptic analysis in a consistent fashion. The frontal situation was complex, the winds strong and ageostrophic and vertical transfers no doubt marked. The particle plots suggest that the boundary layer along G was losing its identity and being absorbed by warm J and a J–K occlusion, although upper levels may as yet have had a separate existence: resolving the situation would require a detailed analysis for which there is no space or necessity here. The clear slot crossed (the analysed) cold K at about 53°N and was pulled into the heart of low I as an intrusion, which rapidly attenuated and lost its identity. The events reflected perhaps greater complexity of ageostrophic and chaotic advection than is easily handled by synoptic analysis. The polar front extended west across the Atlantic, with minor waves. Thus at midday, K trailed back to a returning warm (M) along 52°N, with a small wave depression at 25°W; from there the front continued southwest as cold N. This system prolonged the Mace Head episode through another day.

To conclude, at 0000 UTC 22 March a belt of phase 2 particles, much stretched and narrowed over the preceding 24 hours, lay north-east/south-west through Scotland and Ireland. Concentrations at the Mace Head grid cell plunged shortly after midnight with the passage of cold K. Throughout the 22nd lesser quantities of phase 3 pollutant continued to stream across Mace Head. Warm M returned to Mace Head around 2000 UTC giving a temporary clear patch of warm-sector air, but with the proximity of the rippling cold N to the north, further pulses of pollutant, registering concentrations about one fifth of the maximum, affected Mace Head at intervals, the last five-hour pulse immediately behind cold N, which passed south through Mace Head about 1100 UTC on the 23rd. This terminated the episode.
(c) The episode of 10–11 November 1994

Figure 7 shows predicted versus observed air concentrations for 9–13 November 1994. Overall there is good agreement between the main characteristics of the observed and model curves. Peaks in the observed levels of CFC-11 are clearly evident on the 10th and 11th, with corresponding model peaks. The model unambiguously identifies these peaks as originating from Europe; Fig. 8(a) shows the predicted plume due to European emissions at 1200 UTC 11 November 1994. This shows the cause of the episodes to have been a high pressure system to the north-east of Britain drawing polluted air from central Europe north-westwards across the UK. An east–west cross-section through 54°N in Fig. 8(b) shows much of the material trapped in the lower levels of the atmosphere, most probably due to a capping inversion associated with the high pressure.

From 1800 UTC 11–14 November 1994 the model predicts low levels of CFC-11 (<1 p.p.t.) for much of the period, originating from the USA. Because the levels are low it is not possible to compare the observed and predicted traces quantitatively. However, the model simulations do support the view that pollutants measured during this period are most likely to have originated in North America. Figure 9 shows the plume due to North American emissions only at 24-hour intervals from 1200 UTC 10 November 1994. The material arriving over Mace Head from the 12–13 November 1994 appears to have been transported westwards across the North Atlantic by a complex frontal system. The frontal system was both 'stretched' and undergoing significant vertical mixing as it crossed the Atlantic Ocean, resulting in low pollutant concentrations by the time it reached Mace Head.

7. Discussion and conclusions

Simulations have been carried out with the UK Meteorological Office long-range dispersion model, NAME, of a continuous six-month release of CFCs from Europe and North America, and of two episodes in which it seemed likely that North American emissions had played a role, for comparison with the detailed observational record at Mace Head on the Atlantic coast of Ireland. The correlation between model predictions and observations over the period January to June 1996 is encouraging, with the model reproducing well the main characteristics of the observed short-term fluctuations of CFC-11 concentrations at Mace Head without any clear bias. The model identifies the main sources as being of European origin, though North American sources contribute approximately 4% to the Mace Head predicted total. Estimated European emissions for a range of inert species, determined by fitting model CFC-11 predictions to observations of several species, are reasonably consistent with independent estimates and emission trends.

A similar contribution from North America might be expected from other species that are not readily depleted by chemical conversion, wet- or dry-deposition processes. These include carbon monoxide, carbon dioxide, ozone and the oxides of nitrogen. However, it is likely that levels of soluble and reactive species would be substantially lower, as significant scavenging by wet-deposition processes is likely in frontal zones as the pollutants are transported across the North Atlantic Ocean.

In two separate cases, model simulations have indicated that material detected at Mace Head originated from North American sources some days previously, supporting back-trajectory and wind-sector analyses. As discussed below, in each of the cases the transport appears to be closely associated with low pressure systems tracking eastwards across the North Atlantic Ocean. This is in contrast to European episodes which are often related to slow-moving high pressure systems.
Figure 7. Model-predicted and observed concentrations of species at Mace Head for the November 1994 episode (see text). (a) The fraction of material arriving at Mace Head originating from North America. In (b)-(e) model values are plotted relative to the minimum on the vertical axis, as in Fig. 4.
Figure 8. For 1200 UTC 11 November 1994: (a) particle distribution predicted by the NAME model and observed mean sea-level pressure pattern (see text), and (b) the corresponding vertical cross-section showing particles in the region 54–56°N.
The analysis of the March 1994 episode has led to the identification of the following factors which were unique to the events over the North Atlantic region, leading to the March 1994 episode of relatively high North American CFCs at Mace Head. The factors include:

- A period of accumulation in the source region providing a dense pool of pollutant. This implies col or anticyclonic conditions.
- Efficient transfer of the pool in bulk to the western North Atlantic. In March 1994 this was accomplished by repeated north-westerly winds on the flank of depressions developing in the cyclogenetic region off the North American coast, and then drifting north-east towards Newfoundland.
- A strong, uninterrupted zonal or broadly west-south-westerly flow from the western North Atlantic to western Europe, for direct and rapid transport. This incidentally facilitates identification of the North American source by clearing European pollutants which have gathered over the eastern North Atlantic. A strong, persistent Icelandic low, dominating the northern and eastern North Atlantic, would be a typical predisposing feature. In the episode of March 1994 such a low developed as a result of the explosive deepening of one of the aforementioned family of depressions.
- Protection from processes which would destroy or excessively dilute the pool. The most important are: (i) Strong attenuation due to chaotic advection (dispersion in tendrils, etc.); characteristically this reflects the presence of strong horizontal shear. Under this heading might be included the enfolding of slots of uncontaminated air by similar processes. (ii) Removal by vertical transports in cyclonic and frontal activity, for example, the warm conveyor belt, and also by strong convective activity. (iii) Dilution by mixing through deep boundary layers. This, with horizontal turbulent diffusion, is less critical than (i) or (ii).

By corollary, favourable situations for the maintenance of the pool, once under way, would be location in a region of relatively stable, albeit mobile, air, such as a ridge of high pressure following a cold front, where it may even benefit from mass descent. In a mobile flow driving systems towards high latitudes, a sizeable pool cannot be maintained for very long: the phase 2 pool was still coherent when the pollutant first reached Mace Head early on 21 March 1996, but was distorted beyond recognition by 22 March.

The regional Unified Model products used by NAME have handled the frontal dynamics realistically, within the limitations of the model resolution. NAME in addition was restricted to meteorological fields at three-hourly intervals. The cross-sections show that large amounts of material were uplifted, much being transferred polewards at high level. Whether the depletions of pollutant by mass ascent are quantitatively accurate is yet to be established: at the time of the integrations NAME lacked schemes for the venting of boundary-layer aerosol by strong convection, and for small-scale entrainment processes. As observed by Maryon and Buckland (1994), pollutant has a strong tendency to align itself along or delineate fronts, no doubt reflecting convergence along the system (or in post-frontal diffuence) and large-scale motions parallel to the front. In modelling, however, discretization can blur the relation of the plume to the frontal surfaces and dynamics. Particles passing inadvertently through the front due to a long time-step will of course engage contrasting and inappropriate wind systems, so that disproportionate amounts of material may be lost along the warm conveyor, for example.

A final question concerns the scope for the mixing of European and North American emissions. The early phase, and most important, would be an enfolding of one field of pollutants into the other, in an exactly similar way to the enfolding of polluted and cleaner
Figure 9. Particle positions predicted by the NAME model and observed mean sea-level pressure pattern (see text) at 1200 UTC: (a) 10 November 1994, (b) 11 November, (c) 12 November and (d) 13 November.
air illustrated in Figs. 5(d) and (e), for example. In some situations more of an interleaving or laminar mixing might take place, and this would enhance further merging by the small-scale diffusive processes. The time-scales suggest that broad juxtaposition of material from the different source would be more important than fine mixing and diffusion for significant events—the time needed for material to become well interdiffused would allow considerable dilution by the other ongoing processes. Indeed, it is commonly the case that North American pollutant emissions become well dispersed into the mid/high-latitude background on the time-scale of the North Atlantic transfer.

The horizontal and cross-sectional plots in Figs. 5 and 6 underline the extraordinary complexity of a major plume embedded in realistic three-dimensional flows. Concepts and paradigms derived from historical studies of contour charts, idealized weather systems—even two-dimensional satellite imagery—scarcely prepare one for the convoluted morphologies produced by the mix of chaotic advection and vertical motion. It is impossible to study these patterns without regretting the lack of opportunity properly to elucidate the evolution of some of the more striking or curious structures—a project requiring high time resolution (only 12-hourly NAME products were obtained in this study) and three-dimensional visualization.

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