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The origin of high particulate concentrations over the United Kingdom, March 2000

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Abstract

An episode of exceptionally high PM₁₀ and PM_{2.5} levels was observed during the night of the 2–3 March 2000 throughout England and Wales. The weather was characterised by strong westerly winds and widespread rainfall associated with a low pressure system to the north of Scotland, conditions usually associated with relatively clean, unpolluted air. Possible sources included volcanic ash from an eruption on 26 February 2000 in Iceland, or dust from large sandstorms over the Sahara. A combination of atmospheric transport modelling using the Lagrangian dispersion model NAME, an analyses of satellite imagery and observational data from Mace Head has shown that the most likely origin of the episode was long range transport of dust from the Sahara region of North Africa. Further modelling studies have revealed a number of previously unidentified dust episodes, and indicate that transport of dust from the Sahara can occur several times a year. Dust episodes are of interest for a number of reasons, particulate levels can be elevated over a wide area and in some instances can significantly exceeded current air quality standards. If a natural source is identified over which there can be no control, there are implications for the setting of air quality standards. © 2001 Published by Elsevier Science Ltd.

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1. Introduction

There is increasing evidence that particulate matter has an adverse effect on health, recent epidemiological studies have shown a correlation between air pollution and mortality (Schwartz and Marcus, 1990; Dockery et al., 1993; Pope et al., 1995; Schwartz et al., 1996). In the European Community (EU), concentration limits for PM₁₀ (particles with a diameter <10 µm) have been established under the new Air Quality Directive (Directive 1999/30/EC). The Stage 1 limits to be

achieved by 2005 are an annual mean value of 40 µg m⁻³ with 24 h limit of 50 µg m⁻³, not to be exceeded more than 35 times a year. By 2010, the Stage 2 limits come into force, where PM₁₀ concentrations must not exceed an annual mean value of 20 µg m⁻³, or a daily mean value of 50 µg m⁻³ on more than 7 days a year.

Particulates have a variety of natural and anthropogenic sources, which can be broadly separated into primary and secondary (APEG, 1999). Primary particulates are those which are emitted directly into the atmosphere such as vehicle and industrial emissions and wind-blown dust. Secondary particulates are formed through chemical transformations of gas-phase species

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in the atmosphere. Much attention has been given to man-made sources, initially to domestic coal combustion and more recently to diesel vehicular traffic (QUARG, 1996). In comparison, little attention has been given to natural sources of airborne particles such as wind-blown dusts and sea-salts in the United Kingdom. With the advent of continuously recording monitoring instruments and telemetric reporting, it has become much easier to follow the time series of particulate matter and to observe the occurrence of pollution episodes across the United Kingdom monitoring network. Continuous monitoring of airborne particles with aerodynamic diameters $<10\mu\text{m}$, PM_{10} , began in 1993 and with diameters $<2.5\mu\text{m}$, $\text{PM}_{2.5}$, in 1998.

The majority of particulate pollution episodes have either occurred during wintertime in still, cold weather conditions or during summertime, associated with hot, sunny photochemical pollution conditions (Harrison et al., 1997). A number of particulate pollution episodes have, however, occurred in the British Isles under weather conditions that are far removed from those generally associated with pollution events. One possible cause of these particulate pollution events is the advection of air masses, heavily loaded with dust from the Saharan region of North Africa. Long range transport of Saharan dust across the Mediterranean Sea into southern and central Europe (Rodríguez et al., 2001; Schwikowski et al., 1995; Avila and Peñuelas, 1999; Prodi and Fea, 1979; Chester et al., 1984) and across the tropical Atlantic Ocean to the Caribbean and both North and South America (Rajkumar and Siung Chang, 2000; Prospero, 1999; Prospero et al., 1981; Carlson and Prospero, 1972) is well established. Rodríguez et al., estimate 10–23 exceedances of the $50\mu\text{g m}^{-3}$ PM_{10} standard in Southern Spain and 4–7 exceedances in Northern Spain. Long range transport of Saharan dust to the British Isles appears much less frequent, though specific events have been identified (Reiff et al., 1986; Stevenson, 1969) and red dust deposits are regularly reported over the British Isles following rainfall in air masses originating over the Sahara.

This paper reports how long range transport of Saharan dust led to the occurrence of elevated levels of both PM_{10} and $\text{PM}_{2.5}$ in urban and rural areas across the British Isles during the early months of 2000. The levels approached the national and internationally accepted air quality standards and guidelines set for the protection of human health (EPAQS, 1995; WHO, 1995). A sophisticated Lagrangian dispersion model has been used to study the frequency of long range transport from the Saharan region to the British Isles over a 5 yr period and the likely occurrence of elevated PM_{10} and $\text{PM}_{2.5}$ concentrations.

2. The episode

2.1. Observations

Fig. 1 shows observed values of PM_{10} , $\text{PM}_{2.5}$ and nitrogen oxides at a number of sites across the United Kingdom between 1 and 4 March 2000. All the sites are urban with the exception of Narberth (Wales) and Rochester (East of London) which are considered rural, and Middlesbrough (North East coast of England) which is an industrial site. Data were retrieved from the National Air Quality Information Archive web site <http://www.aeat.co.uk/netcen/airqual/index.html> funded by the Department of Environment, Transport and Regions (DETR). Significant peaks in PM_{10} are evident at all sites except for those in Scotland and Northeast England, and similar peaks are seen in $\text{PM}_{2.5}$ observations at London and Rochester, the only sites for which $\text{PM}_{2.5}$ data are available. No corresponding peaks are evident in the nitrogen oxides data, which indicates that the source was not industrial or from traffic. The highest peak occurs in Plymouth, in the Southwest, with a maximum mean hourly concentration of $292\mu\text{g m}^{-3}$, the highest value detected in Plymouth since measurements began. Maximum concentrations generally reduce from west to east, with maximum values in eastern sites dropping to $\approx 100\mu\text{g m}^{-3}$. The rise in PM_{10} levels occurs some 2–3 h earlier in the west, which indicates rapid west to east transport. In contrast the end of the episode occurs later towards the south, resulting in longer periods of high PM_{10} levels in the south and west, and shorter periods of high PM_{10} in the north and east. The strong correlation between PM_{10} and $\text{PM}_{2.5}$ suggests a common source, consisting of material with a broad size distribution. $\text{PM}_{2.5}$ concentrations are about half of those of PM_{10} levels, indicating a higher number density of $\text{PM}_{2.5}$ particles.

2.2. Meteorology

The synoptic chart for 00Z 3 March in Fig. 2 shows a low pressure centred to the Northeast of Scotland, bringing strong west to northwesterly winds across the whole of the United Kingdom. An associated frontal system is evident, with a warm front clearing the south of the country by midnight, and a strong cold front trailing across central parts of the United Kingdom at midnight, moving rapidly south and clearing the south coast by midday on the 3 March. Extensive precipitation was associated with the passage of the cold front.

Detailed analyses of the timing of the pollution episodes reveals that high particulate levels occurred after the passage of the warm front, and that particulate levels fell to background levels with the passage of the cold front. Elevated particle levels were not observed either before or after the passage of the warm sector.

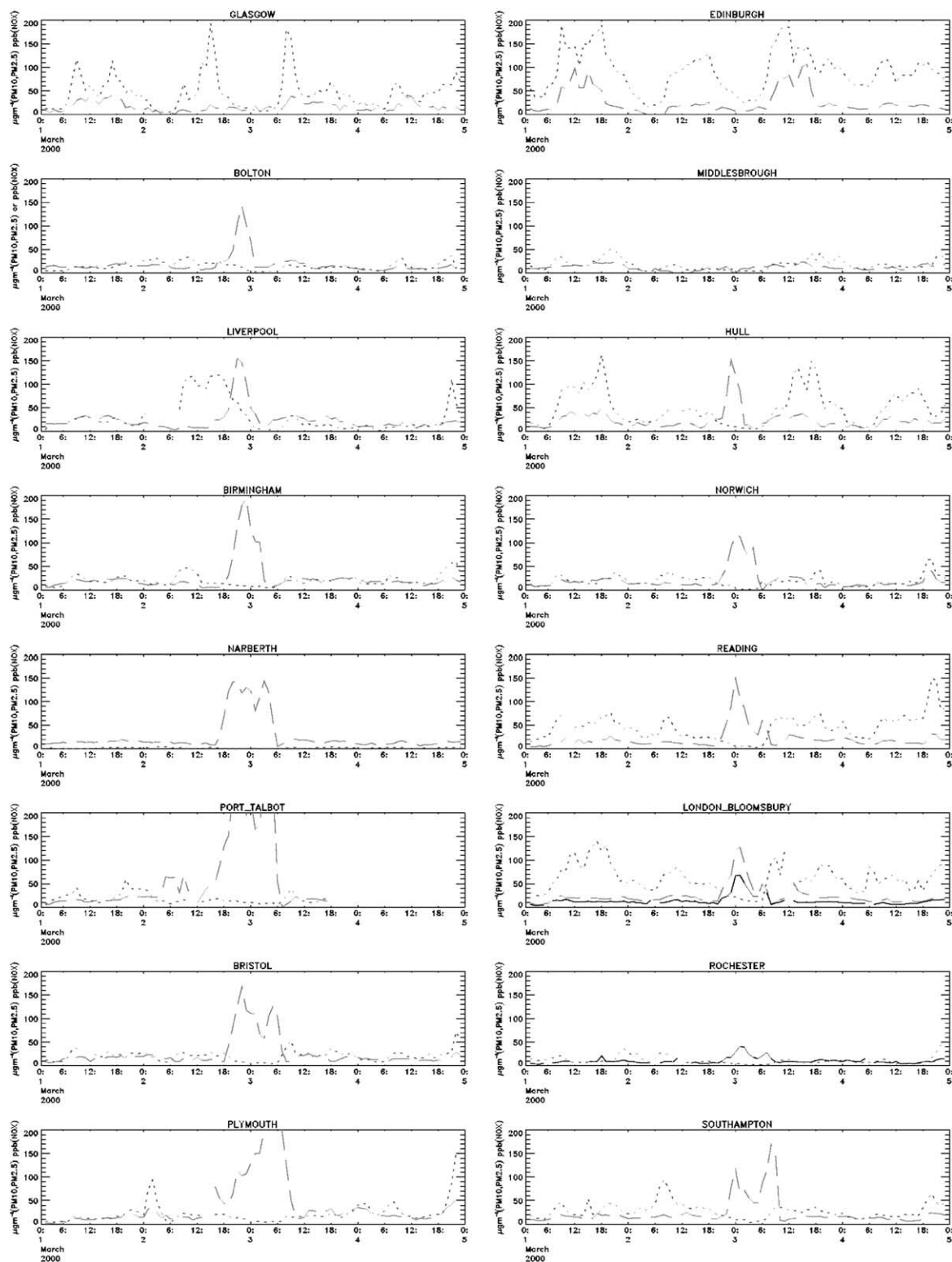


Fig. 1. Observed PM₁₀, PM_{2.5} and NO_x concentrations (PM_{2.5} in black, PM₁₀ in red and NO_x in blue). Plots on the left represent westerly locations, and plots on the right represent easterly locations.

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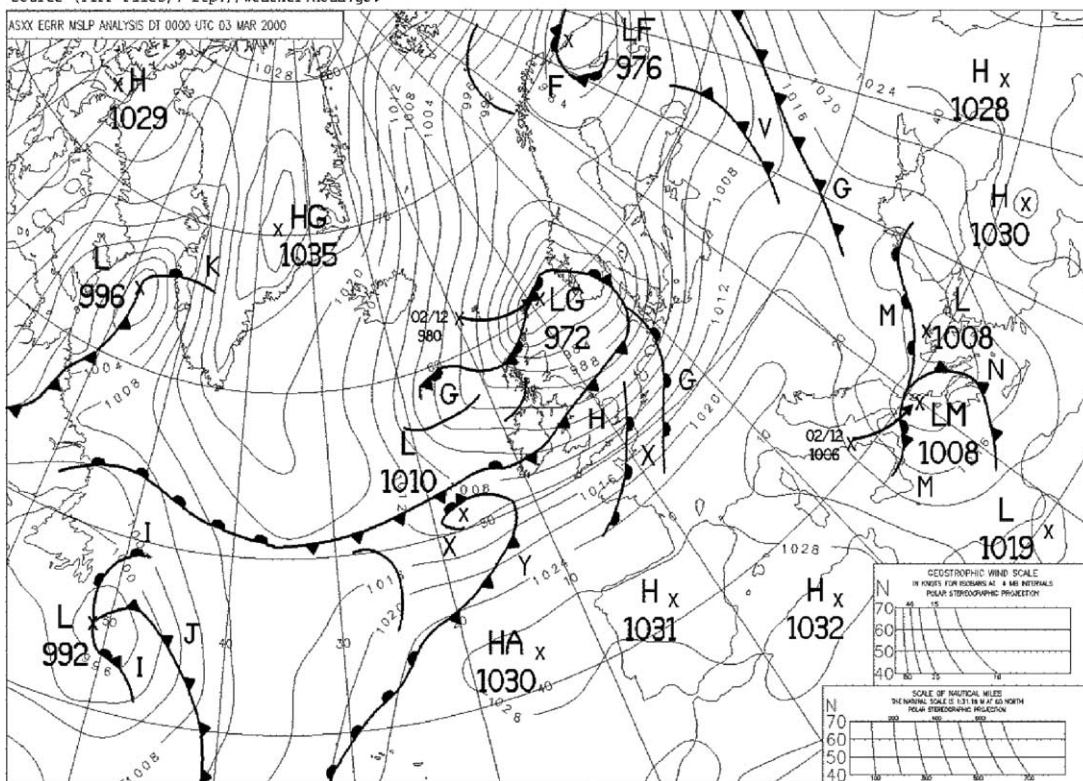


Fig. 2. Met Office North Atlantic analyses (ASXX) for 00Z 3 March 2000.

This shows that the episode is associated with the warm sector of the low pressure system, suggesting a southerly origin. The sharp fall in concentrations with the passage of the cold front suggests minimal mixing between the warm and cold sectors.

No correlation is evident between the timing of the pollution peaks and precipitation. Elevated pollution levels are evident several hours before the onset of precipitation at several locations, with high levels continuing during periods of high rainfall. For example Fig. 3 shows radar derived rainfall at 00Z on the 3 March, clearly showing a band of sometimes intense precipitation associated with the cold front lying across central parts of the UK. At this time high particulate concentrations were being measured at all sites south of the front, which include sites well ahead of the rain band as well as sites within the rain band.

2.3. Possible sources

The evidence clearly suggests that the episode was of natural origin. Had the episode been due to industrial or traffic emissions, similar peaks should have been observed in a range of other pollutants such as nitrogen

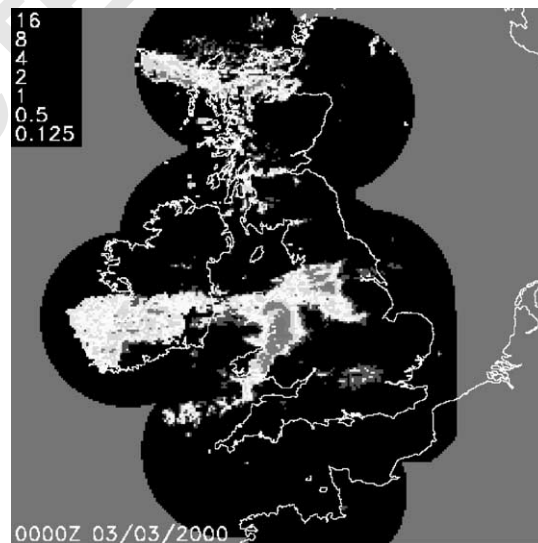


Fig. 3. Radar derived rainfall at 00Z 3 March 2000.

oxides or sulphur dioxide. In fact, observations of all other routinely monitored pollutants remained at low

levels throughout the period, consistent with relatively clean air associated with strong westerly winds.

Initially the plume was thought to be volcanic ash originating from Mount Hekla in Iceland, which erupted during the evening of the 26 February, some 5 days before the episode. Earlier in the week the Met Office had been issuing volcanic ash forecasts which indicated ash might reach the United Kingdom during the 2 and 3 March. Back trajectories (not shown) for midday on the 2 and 3 March also indicated a source to the north west of the UK. However back trajectories for the morning of the 3 March show a more southerly source, suggesting air over southern parts of the UK originated in the mid Atlantic to the west of Africa. Coincidentally there had been press reports of a large sandstorm observed by satellite moving west from the Sahara into the Atlantic, covering the Canary Islands. Given the level of interest in the episode, with enquiries originating from local and central government departments and the public, further modelling work using the Met Office's dispersion model NAME was undertaken to identify the most likely origin.

3. Name model

The Met Office's dispersion model NAME (Nuclear Accident Model) is a Lagrangian particle model used to predict the transport of airborne pollutants over ranges of a few kilometres to many 1000s of kilometres (Ryall and Maryon, 1998). It utilises three dimensional wind fields from the Met Office's numerical weather prediction model, the Unified Model (Cullen, 1993). Pollutants are represented by large numbers of particles which are released into the model atmosphere and then advected by the local mean wind, with various random walk techniques used to represent turbulent diffusion processes. Parametrisations are available for entrainment between the boundary layer and the free troposphere, for mixing by deep convection and for wet and dry deposition processes. Originally developed for emergency response purposes, NAME is implemented operationally for generating forecasts in the event of major atmospheric releases, such as from a nuclear accident or volcanic eruptions. NAME has also been applied to a range of air quality problems, such as understanding PM_{10} transport to the UK (Malcolm et al., 2000) and interpreting Mace Head observations (Derwent et al., 1998a, b; Ryall et al., 2001).

The eruption from Mount Hekla is assumed to have started at 1800Z on 26 February, lasting for 3 h with the plume extending from the surface to 45,000 ft (13700 m), as reported by pilots following the eruption. A nominal release rate of 1 g s^{-1} was used. For Saharan dust an area source covering the region 15W–5E, 15–25N was assumed, also with a nominal emission rate of 1 g s^{-1} .

Material was released continuously between the surface and 1000 m above ground. A surface release could have been used, letting the model handle vertical mixing in the boundary layer, but it was felt that a deeper release was appropriate to ensure a reasonable depth of dust near the surface. In practice the dust sources are likely to be localised, depending on the nature of the soil, rainfall history and boundary layer wind profiles. However, the aim here was not to replicate the detailed nature of the dust, rather to look at the broad scale transport to the UK.

Fig. 4 shows the predicted plume from both Mt. Hekla and the Sahara between the 2 and 4 March. Note that quantitative comparisons of predicted and observed concentrations are inappropriate as nominal release rates were used in absence of known emission rates. These simulations suggest that volcanic ash from Mount Hekla would have reached northern parts of Scotland in the boundary layer on the 28 February, before slowly moving south and clearing the South coast by 12Z on the 2 March, prior to the start of the PM_{10} and $\text{PM}_{2.5}$ episode. As particulate levels remained low during this period it is likely that the ash plume from Hekla was too dilute to be detected. In contrast material originating from the Sahara is predicted to rapidly cross the UK from the Atlantic late on 2 March, covering most of Ireland, England and Wales but not Scotland. The plume is then predicted to move south across England and Wales, clearing the south coast by 12Z on the 3 March. These predictions match the observed pattern of high PM_{10} and $\text{PM}_{2.5}$ concentrations very well, suggesting that dust from the Sahara is the most likely source. Extensive rain associated with the cold front would have resulted in dust being washed out, causing widespread deposits. As the plume moved east the rain will have depleted the plume significantly, which may in part explain why lower air concentrations were observed in eastern areas.

3.1. Comparison with AUN measurements

Spatially averaged observations of $\text{PM}_{2.5}$, PM_{10} , nitrogen oxides and sulphur dioxide from Automatic Urban Network (AUN) sites are plotted in Fig. 5. For $\text{PM}_{2.5}$ data are averaged over just three sites, whilst for the remaining species observations were averaged over more than 30 sites spread across England and Wales. Averaging observations over a number of sites helps emphasise periods of elevated concentrations that occur over a broad area from a common and distant source. In addition NAME predicted concentrations of Saharan dust and volcanic ash from Mount Hekla are shown averaged over Birmingham and London.

The spatially averaged observations clearly show the episode of the 2–3 March with a peak in both PM_{10} and $\text{PM}_{2.5}$, but no corresponding increase in nitrogen oxides

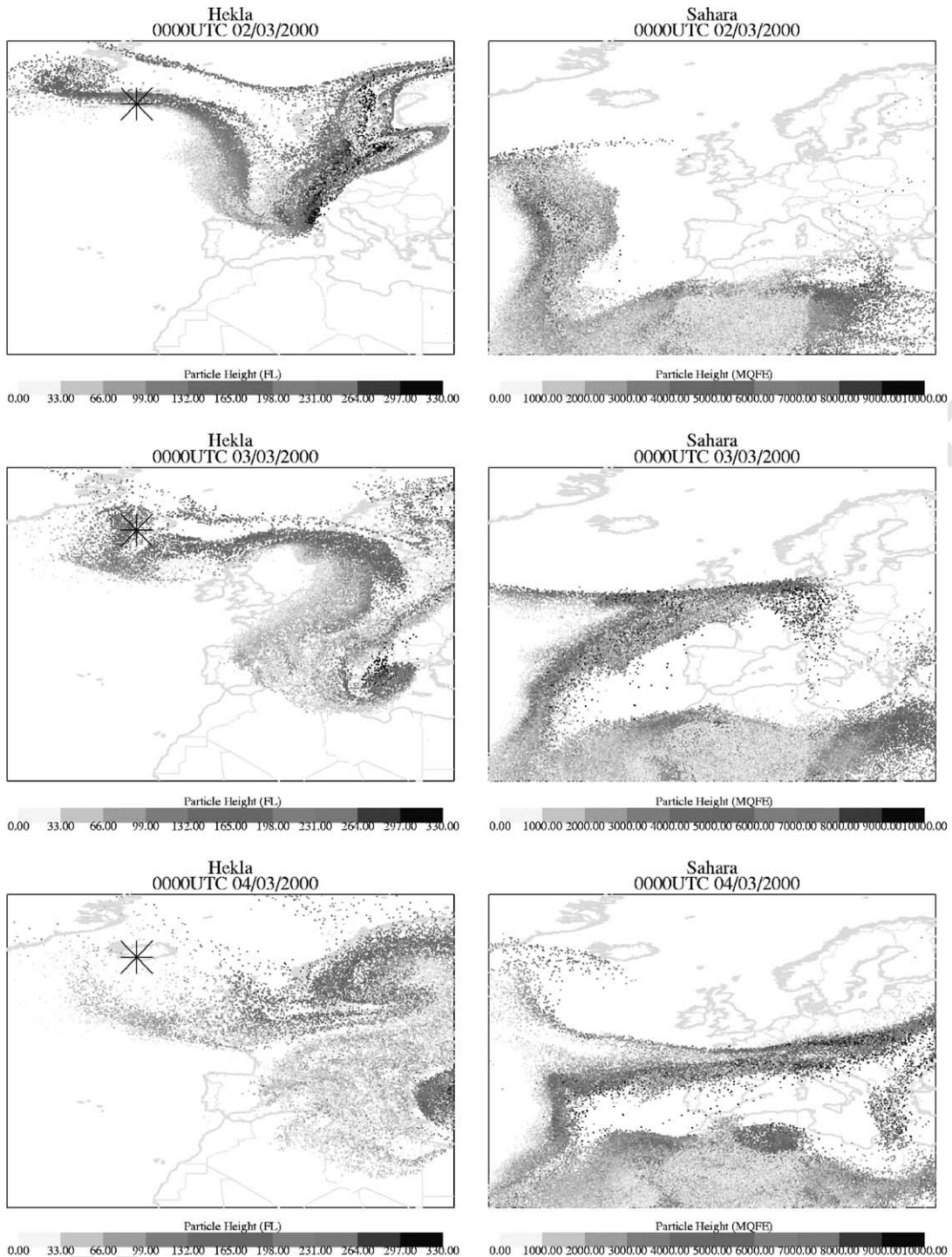


Fig. 4. NAME model predictions for volcanic ash from Mount Hekla (left) and Saharan dust (right). The plumes are coloured according to their height, Flight Levels (100s feet above sea level) for volcanic ash, metres above ground for Sahara dust.

and sulphur dioxide. In fact concentrations of both nitrogen oxides and sulphur dioxide appear particularly low during the episode, confirming that the dust arrived

in a clean airmass, unpolluted by industrial emissions. The timing of the episode corresponds well with the timing of the NAME predicted peak for Saharan dust.

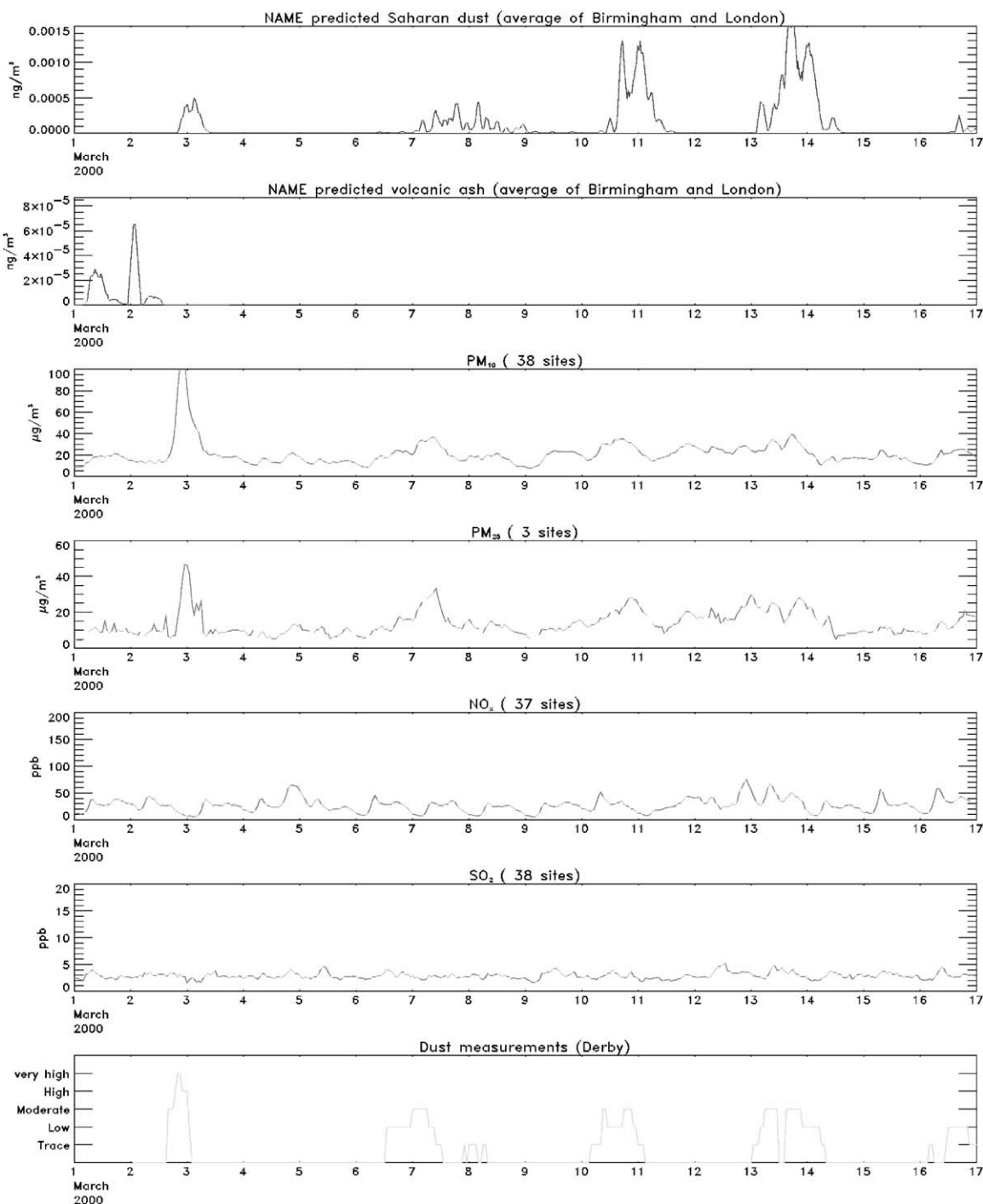


Fig. 5. NAME model predicted air concentrations for Saharan dust and volcanic ash, network averaged concentrations of $PM_{2.5}$, PM_{10} , nitrogen oxides and sulphur dioxide over England and Wales, and dust measurements from Derby. Units are $\mu g m^{-3}$ for PM_{10} and $PM_{2.5}$, ppb for SO_2 and NO_x .

3.2. Further episodes

The NAME model also predicts three further episodes of Saharan dust transport, on the 7–8, 10–11 and 13–14 of March. On the morning of the 14 March the Met Office received a number of calls from the public who reported that significant dust deposits had appeared on surfaces such as cars and windows overnight. These deposits, variously described as grey or yellow/red in colour were observed over a wide area, including Cumbria, Cornwall, Wales, Dorset and Sussex. This supports the conclusion that dust transport from the Sahara was again the source of the reported deposits. A small increase in particulate concentrations was evident during this period, but as nitrogen oxide concentrations were also elevated it is not possible to attribute elevated particulate concentrations to Saharan dust, either wholly or in part.

On both other occasions there were corresponding increases in measured $PM_{2.5}$ without associated peaks in nitrogen oxides or sulphur dioxide. On the 7 March there is also a small but significant increase in average PM_{10} levels. These observations suggest that there were two more episodes of dust transport from the Sahara. As there was no precipitation on these dates, very little dust would have been deposited, resulting in less visible evidence. The NAME predictions shown in Fig. 6 show that the dust plume reaching the United Kingdom on the 3 March took ≈ 1 week to reach the United Kingdom, whilst the plume in the subsequent three episodes took 2 weeks or more to reach the United Kingdom. This may explain why high PM_{10} values were only observed in the first episode. Larger particles will be lost more rapidly due to gravitational settling, leaving a progressively larger proportion of smaller particles as the plume ages.

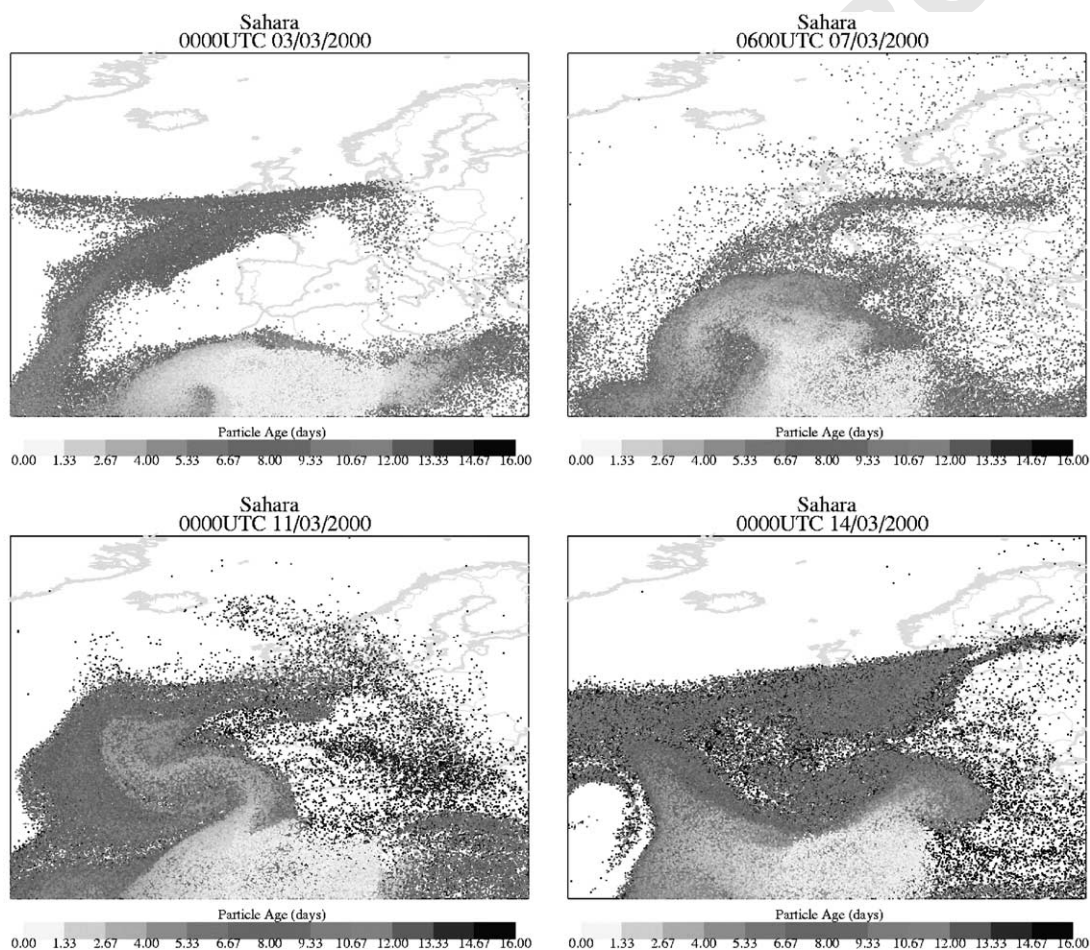


Fig. 6. NAME predicted plumes from the Sahara for 3, 7, 11 and 14 March 2000. Plume colours indicate time since release.

4. Supporting observations

4.1. Satellite imagery

Concentrating on the 2–3 March episode a range of satellite imagery and observational data have been studied to identify those products that best reveal the presence of significant dust clouds. The clearest images come from the SeaWiFS project <http://seawifs.gsfc.nasa.gov/SEAWIFS.html>. These true colour, visible images taken from low earth orbit clearly show widespread areas of dust to the west of Africa on the days before the 2 March episode. In Fig. 7 a plume of dust can clearly be seen extending towards the Southwest of the UK.

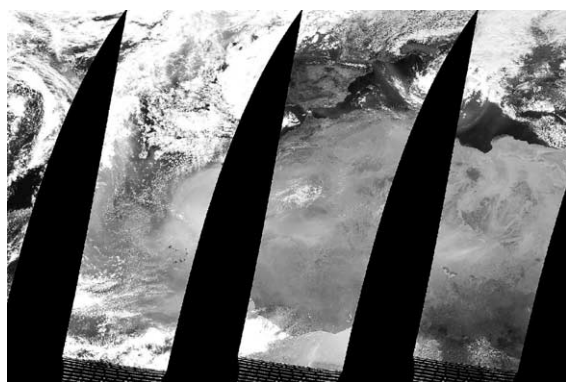


Fig. 7. SeaWiFS image from the 2 March 2000. Areas of dust can clearly be seen to the west of Africa and Spain, extending towards the UK.

Another satellite product that shows an aerosol cloud reaching the UK from Africa is the TOMS derived aerosol index (<http://jwocky.gsfc.nasa.gov/>) shown in Fig. 8. These images, which are generated from measurements of ultraviolet radiation backscatter (Torres et al., 1998), provide estimates of the aerosol content of the atmosphere. The pattern of transport is consistent between the two products and agrees well with the NAME model predictions. Visible products from polar orbiting satellites also revealed the dust cloud, especially at low sun angles. Whilst many satellite products can help track dust clouds, they do not tell the whole story. Observations are generally qualitative rather than quantitative, and give limited detail about the vertical distribution of material. In particular it is not possible to determine if the plume extends to the surface, where it may pose a risk to health.

4.2. Spore traps

The Midlands Asthma and Allergy Research Association (MAARA) in Derby has been operating volumetric spore traps (Hirst, 1952) since 1968 with the aim of monitoring and quantifying pollen and spore contents at hourly resolution. These traps are sited 10 m above ground level, on a roof site at the University of Derby's Mickleover site, ≈ 4 km south-west of the city centre. In addition to pollen and spores, the traps will capture any other particulate matter present in the atmosphere, including dust particles. Over the years elevated dust levels have been observed on a number of occasions, but levels during the 2–3 March were exceptionally high.

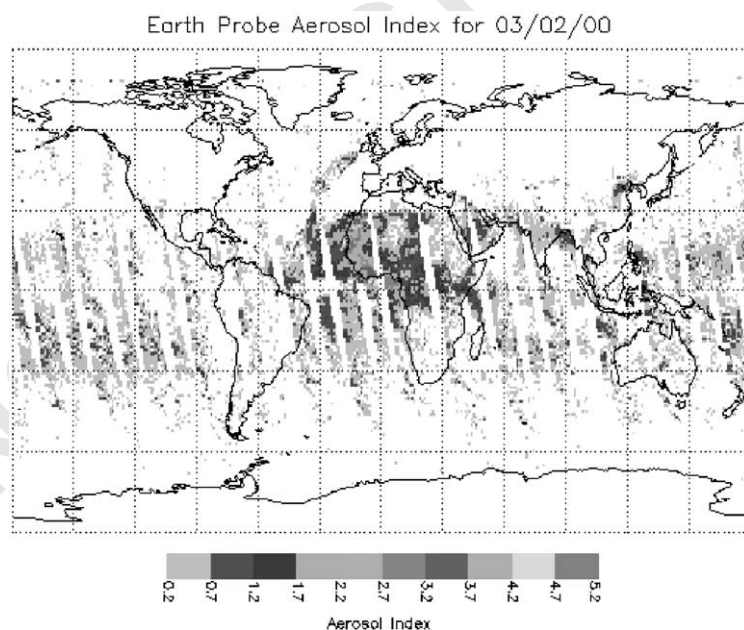


Fig. 8. TOMS derived aerosol index for 2 March 2000.

For comparison with model predictions and other observations a detailed visual inspection was made of the dust deposited in the spore traps between 2 and 17 March 2000. Dust levels were reported as trace, low, moderate, high or very high at hourly intervals and are plotted in Fig. 5. The correlation for the 2–3 March is excellent, with very high dust levels observed during the episode. Good correlation is also found with the three subsequent episodes predicted by NAME. The observed particles on the 2–3 March were different in nature to the dust observed on later days, containing mainly clear or ochre particles with significant amounts of red, brown and yellow particles and a greater proportion of larger particles. This is consistent with the 2–3 March episode being of an exceptional nature, with very high particle concentrations with a broad particle size distribution. Transport to the UK was fairly rapid, which would have allowed many of the larger particles to remain in suspension.

It is interesting to note that a small number of particles found in samples from 2 to 3 March were considered to be of volcanic in origin, suggesting some of the dust may have originated from Hekla. The NAME model predictions show volcanic ash transport from Hekla to the UK both before and after the passage of the dust. There are three possible explanations for volcanic ash being present in some of the samples containing Saharan dust: (i) particles deposited before the passage of the warm front may have been resuspended by rain or strong winds; (ii) some mixing may have occurred between the air masses; or (iii) precipitation may have washed out ash particles from higher levels.

4.3. Mace head data

Since 1994, high frequency (40 min interval) real-time gas chromatographic measurements of the principal halocarbons and radiatively active trace gases have been made as part of the Global Gases Experiment (GAGE/AGAGE) at Mace Head, Co. Galway, Ireland (Simmonds et al., 1996a; Cunlold et al., 1997). In addition a fully automated gas chromatograph-mass spectrometer (GC-MS) has been used to monitor a range of additional species, typically at 4 hourly resolution, including many HCFCs and HFCs (Simmonds et al., 1996b). In recent years the NAME model has been used to help interpret measurements from Mace Head, identifying source regions and strengths for many of the monitored species (Ryall et al., 1998, 2001; Derwent et al., 1998a, b). The site is situated on the West Coast of Ireland, with few local man-made pollution sources.

Previous studies (Simmonds et al., 1997; O'Doherty et al., 2001) have shown that transport from southerly latitudes in tropical maritime air masses result in observed concentrations dropping below baseline levels

for a number of species, including carbon monoxide, methane, methyl chloroform and chloroform. This is due to air being transported from near equatorial regions, which are characterised by lower atmospheric concentrations. In Fig. 9 NAME model predicted concentrations of Saharan dust at Mace Head are plotted together with PM₁₀ observations from Lough Navar, and Mace Head observations for carbon monoxide, methane, chloroform and methyl chloroform. Lough Navar is a rural monitoring site in Northern Ireland some 180 km to the northeast of Mace Head, and is part of the AUN with hourly PM₁₀ data available.

The PM₁₀ data clearly show a peak reaching 100 µg m⁻³ during the afternoon of the 2 March, coinciding with the NAME predicted peak. Each of the four species monitored at Mace Head show a corresponding drop in concentrations, showing that the dust plume was associated with air originating in tropical latitudes. Reduced concentrations of these species have not been observed in air masses reaching Mace Head from west or northwesterly directions. It is not clear whether the dust was generated within air of tropical maritime origin, or if the plume was mixed with tropical maritime air during transport to the UK. Further trajectory or transport modelling would be required to determine the history of the air in which the dust was first suspended.

Reduced concentrations also coincide with some of the subsequent transport events during March 2000, however on the 10 March levels remain at baseline levels. Either the dust was well mixed with mid-latitude air during transport to the UK, or the air into which the dust was incorporated over the Sahara was not of southerly or tropical origin. So while the Mace Head data can confirm that the air during a given episode is of southerly origin, absence of southerly transport does not exclude dust transport from the Sahara.

5. Frequency of episodes

Having established that an episode of Saharan dust transport has resulted in air quality standards being exceeded over many parts of the UK, it is important to establish whether this is a one-off event or whether Saharan dust transport is a more frequent event than currently thought. Whilst the unusual nature of the 2–3 March episode could be readily identified, the situation is not usually so obvious. A wide range of source types with high temporal and spatial variability contribute to particulate concentrations, and the chemistry behind the production of secondary particulates is far from complete (APEG, 1999; Malcolm et al., 1999). Simply attempting to isolate PM₁₀ peaks which do not have corresponding peaks in other 'industrial' pollutants such

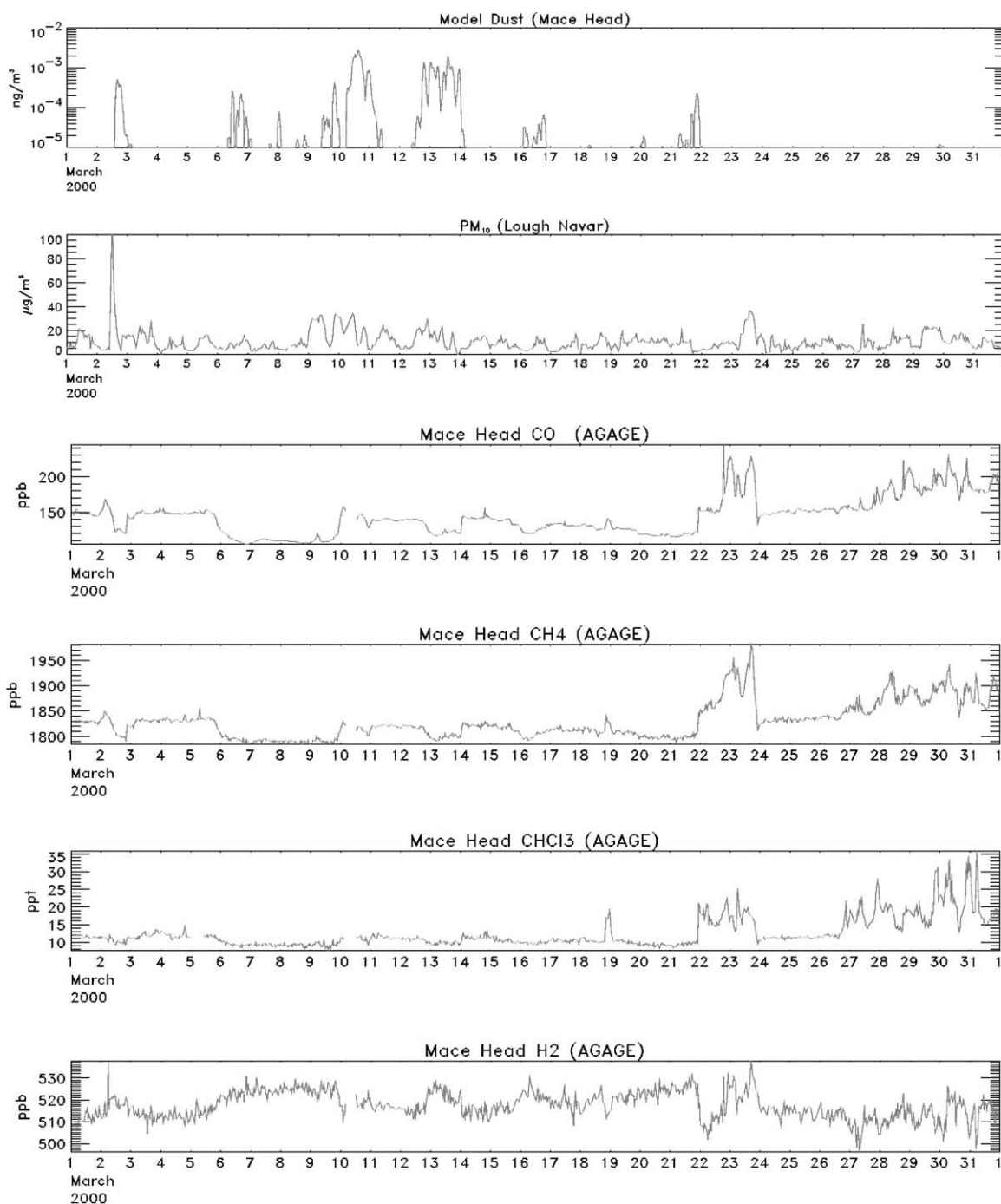


Fig. 9. NAME model predictions for Mace Head (top), PM_{10} observations at Lough Navar and carbon monoxide, methane, chloroform and methyl chloroform at Mace Head.

as carbon monoxide or nitrogen oxides is generally inconclusive. Such events may also be due to other natural sources, including sea salt, volcanic ash or other poorly understood natural sources.

In order to identify the periods most likely to be associated with Saharan dust events, and to assess how frequently air from the Sahara might reach the UK, an extended five and a half year NAME simulation of an

idealised continuous Saharan dust source has been carried out. An area source covering the region 10W–20E, 15–30N was used, with a tracer species being released continuously at 1 g s^{-1} between the surface and 500 m above ground level. Fig. 10 shows the predicted concentrations at London from January 1995 to July

2000. These results show that transport of near-surface air from the Sahara to the UK is relatively common, with more events during the spring and fewer events during the summer months. Over the whole period the model predicts that air from the Sahara reaches southern UK $\approx 10\%$ of the time, with transport predicted

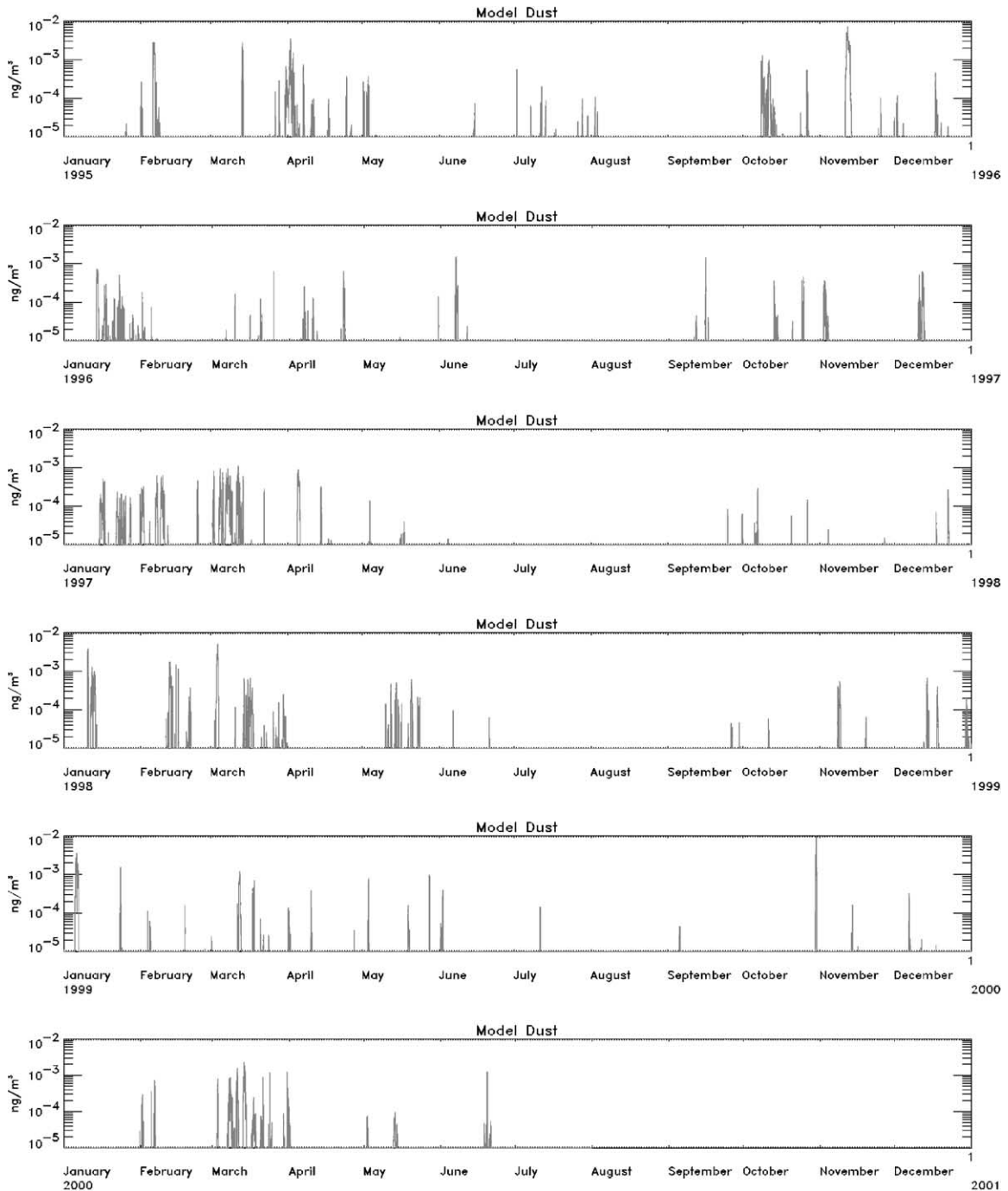


Fig. 10. NAME predicted concentrations at London due to an idealised dust source over the Sahara.

during nearly 1 in 7 days. Individual transport events range in duration from an hour up to several days. Whether air reaching the UK from the Sahara contains significant dust will depend on a number of factors. Sufficient dust concentrations need to be generated at source, then subsequent transport must occur without

excessive losses from wet and dry deposition processes, or by mixing with clean air. In addition larger particles will also be rapidly lost due to sedimentation.

In an attempt to isolate further dust transport events the NAME model predictions were again compared with Lough Navar observations of PM_{10} , and Mace Head

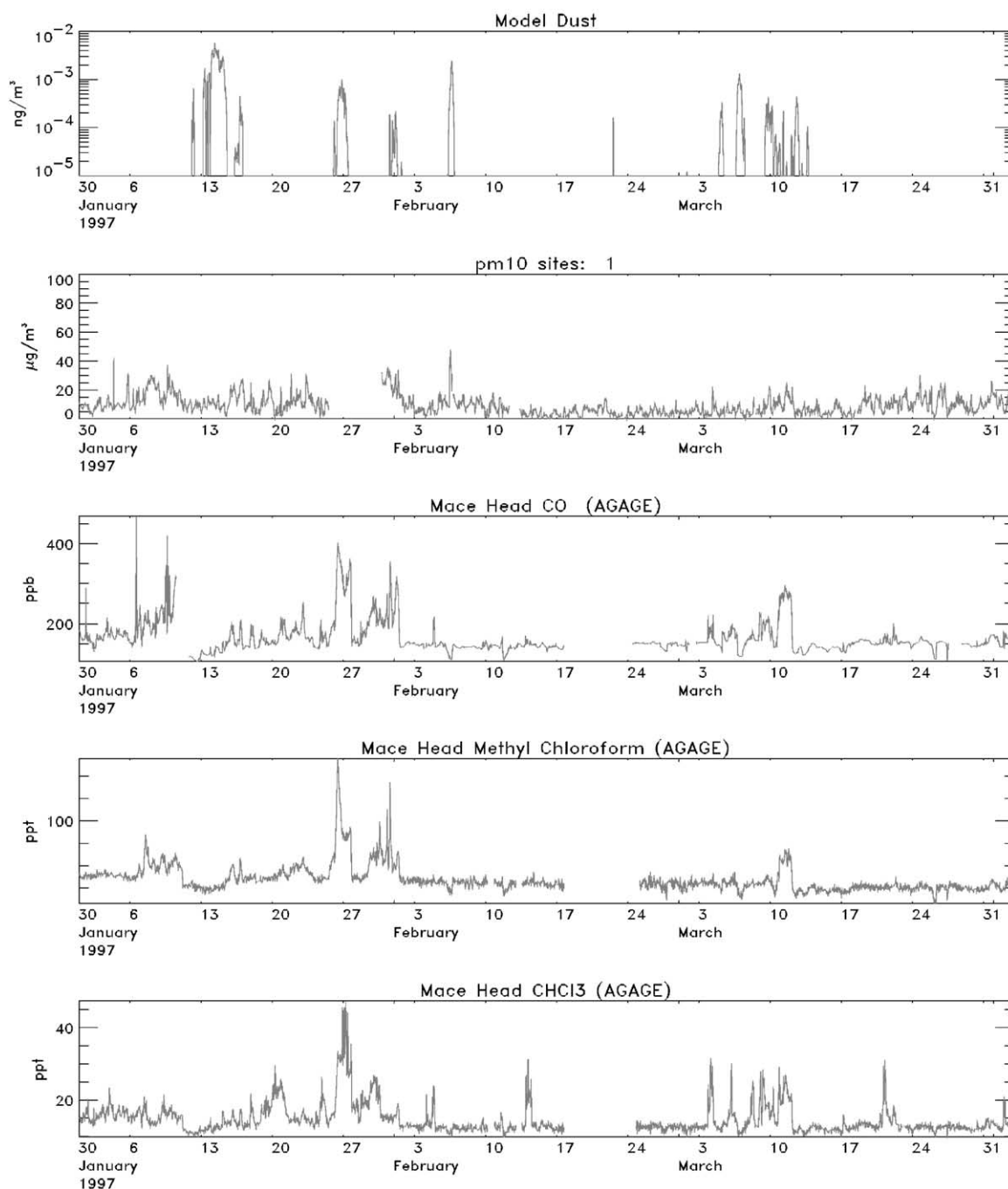


Fig. 11. NAME predicted dust at Mace Head, PM_{10} observations at Lough Navar, and carbon monoxide, methyl chloroform and chloroform at Mace Head.

observations of carbon monoxide, methyl chloroform and chloroform, a period of three months from this data is shown in Fig. 11. Unfortunately NO_x is not monitored at Lough Navar, so we use carbon monoxide as an indicator of industrial pollution. As discussed in Section 4 low concentrations of carbon monoxide, methyl chloroform and chloroform are good indicators of southerly transport, and chloroform is useful in eliminating events due to local sources. O'Doherty et al. (2001) have identified widespread natural sources of chloroform at Mace Head, which result in elevated chloroform concentrations during periods of low wind and stable conditions. This can help identify periods of high particulate concentrations that are due to local sources such as fires or peat burning, that might otherwise have the characteristics of a dust event.

On many occasions where NAME predicts possible Saharan transport PM_{10} levels remain low, showing minimal dust transport. On many other occasions PM_{10} is elevated, but so are the Mace Head concentrations suggesting the presence of man-made pollutants due to transport over populated regions. In these situations dust transport cannot be eliminated or confirmed. In a limited number of cases NAME predicted transport events correlate well with a PM_{10} peak and southerly transport, suggesting Saharan dust. A good example can be seen in Fig. 11 on the 6 February 1997, when observed PM_{10} levels reached $40 \mu\text{g m}^{-3}$. A total of eight such events could be identified during the five and a half year period, with PM_{10} concentrations rising at least $20 \mu\text{g m}^{-3}$. With the exception of the 2–3 March 2000 episode none of these transport events resulted in air quality standards being exceeded. Whilst all the events detected occurred between January and June, insufficient events have been identified to determine any seasonal patterns.

6. Discussion and conclusions

The episode of 2–3 March has been demonstrated to originate from the Saharan region of North Africa. NAME model predictions correlate well with observations, including ground based PM_{10} and $\text{PM}_{2.5}$ observations, dust analyses and satellite imagery. Analyses of Mace Head data also shows that the dust episode was associated with southerly transport from tropical latitudes. Three further transport events, albeit at much reduced dust levels, were identified in the 3 weeks following the episode, also by a combination of modelling and observational analyses.

A 5 yr NAME simulation has shown that transport of air from the Sahara is relatively common, with air originating over the Sahara reaching the UK some 10% of the time. On most of these occasions dust transport is minimal, as PM_{10} levels remain low. However analyses

of PM_{10} measurements at the rural site of Lough Navar, together with Mace Head observations has revealed a number of significant episodes between 1995 and 2000 resulting in elevated PM_{10} concentrations. On many other occasions dust may have contributed to observed levels, but positive attribution is not possible. This is likely to be due to possible contamination with particles from other sources both natural and man-made. Compared to southern European countries Saharan dust transport to the UK is much less common. We predict just one or two events per year over the UK, with daily mean concentrations rarely exceeding $50 \mu\text{g m}^{-3}$. In contrast Rodriguez et al. (1997) identify up to 23 events resulting in daily mean concentrations in excess of $50 \mu\text{g m}^{-3}$.

Prior to the availability of continuous particulate and pollution measurements and high resolution satellite imagery, the primary evidence for Saharan dust transport to the UK was deposited dust, washed out from the atmosphere by precipitation. For example on the 14 March 2000 dust transport was readily identified through deposits on windows and vehicles, but concentrations were too low to be seen in particulate measurements. This could be one reason why so few episodes have been reported over the years, as many transport events are likely to have occurred during periods of no rain.

The coarser fraction of PM_{10} will undergo significant loss due to sedimentation under gravity, reducing concentrations over long range transport of a few days or more. In contrast the $\text{PM}_{2.5}$ fraction will stay suspended for much longer periods. This is evident on the 7 and 11 March 2000, where dust is more easily identified in the $\text{PM}_{2.5}$ data shown in Fig. 5. It is possible that the contribution of Saharan dust to particulate concentrations over the UK is more significant for $\text{PM}_{2.5}$ than for PM_{10} .

Whilst some specific events have been identified, and show that dust transport is relatively common, it is not possible at this stage to quantify the dust proportion of observed PM_{10} or $\text{PM}_{2.5}$ at a particular location. There is perhaps a need to analyse particulate samples over an extended period of a few years to better characterise and quantify natural dust components. As well as Saharan dust there may also be contributions from mainland Europe, especially after prolonged dry weather, or from local sources in the UK. Significant dust storms are occasionally observed during spring in the Fens region of East Anglia, and are sufficiently well known to have a local name—the Fen Blow.

Major dust events may cause exceedances on a small number of occasions, but in contrast to southern European countries the overall impact on air quality standards as they currently stand is thought to be minimal. The impact of increased desertification and climate change are not easy to assess, given the complex

nature of dust transport, though it is possible that the frequency and strength of future events could increase over the next few decades. As air quality standards become tighter, and standards for PM_{2.5} are introduced, a better understanding of natural dust sources is required.

7. Uncited references

Savoie and Prospero, 1977; Simmonds et al., 2000.

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References

APEG, 1999. Source apportionment of airborne particulate matter in the United Kingdom. Report of the Airborne Particles Expert Group, Department of the Environment, Transport and the Regions, London.

Avila, A., Peñuelas, J., 1999. Increasing frequency of Saharan rains over northeastern Spain and its ecological consequences. *The Science of the Total Environment* 228, 153–156.

Carlson, T.N., Prospero, J.M., 1972. The large-scale movement of Saharan air outbreaks over the northern equatorial Atlantic. *Journal of Applied Meteorology* 11, 283–297.

Chester, R., Sharples, E.J., Sanders, G.S., Saydam, A.C., 1984. Saharan dust incursion over the Tyrrhenian Sea. *Atmospheric Environment* 18, 929–935.

Cullen, M.J.P., 1993. The unified forecast/climate model. *Meteorological Magazine (UK)* 1449, 81–94.

Cunnold, D.M., Weiss, R.F., Prinn, R.G., Hartley, D., Simmonds, P.G., Fraser, P.J., Miller, B., Alyea, F.N., Porter, L., 1997. GAGE/AGAGE measurements indicating reductions in global emissions of CCl₃F and CCl₂F₂ in

1992–1994. *Journal of Geophysical Research* 102, 1259–1269.

Derwent, R.G., Simmonds, P.G., O'Doherty, S., Ciais, P., Ryall, D.B., 1998a. European source strengths and northern Hemisphere baseline concentrations of radiatively active trace gases at Mace Head, Ireland. *Atmospheric Environment* 32, 3703–3715.

Derwent, R.G., Simmonds, P.G., O'Doherty, S., Ryall, D.B., 1998b. The impact of the Montreal Protocol on halocarbon concentrations in northern Hemisphere baseline and European air masses at Mace Head, Ireland over a ten year period from 1987–1996. *Atmospheric Environment* 32, 3689–3702.

Dockery, D.W., Pope, C.A., Xu, X., Spengler, J.D., Ware, J., Fay, M.A., Ferris, B.G., Speizer, F.E., 1993. An association between air pollution and mortality in six US cities. *New England Journal of Medicine* 329, 1753–1759.

EPAQS, 1995. Particles. Report of the Expert Panel on Air Quality Standards, HMSO, London.

Harrison, R.H., Deacon, A.R., Jones, M.R., 1997. Sources and processes affecting concentrations of PM₁₀ and PM_{2.5} particulate matter in Birmingham (UK). *Atmospheric Environment* 31 (24), 4103–4117.

Hirst, J.M., 1952. An automatic volumetric spore-trap. *Annals of Applied Biology* 39, 257–265.

Malcolm, A.L., Derwent, R.G., Maryon, R.H., 2000. Modelling the long range transport of secondary PM₁₀ to the UK. *Atmospheric Environment* 34, 881–894.

O'Doherty, S., Cunnold, D., Sturrock, G.A., Ryall, D.B., Derwent, R.G., Wang, R.H.J., Simmonds, P.G., Fraser, P.J., Weiss, R.F., Salameh, P., Miller, B.R., Prinn, R.G., 2001. In-Situ Chloroform measurements at AGAGE atmospheric research stations from 1994–1998. *Journal of Geophysical Research* (in press).

Pope, C.A., Thun, M.J., Namboodiri, M.M., Dockery, D.W., Evans, J.S., Speizer, F.E., Heath, C.W., 1995. Particulate air pollution as a predictor of mortality in a prospective study of US adults. *American Journal of Respiratory Critical and Care Medicine* 151, 669–674.

Prodi, F., Fea, G., 1979. A case of transport and deposition of Saharan dust over the Italian peninsular and Southern Europe. *Journal of Geophysical Research* 84, 6951–6960.

Prospero, J.M., 1999. Long-range transport of mineral dust in the global atmosphere: impact of African dust on the environment of the southeastern United States. *Proceedings of the National Academy of Sciences, USA* 96, 3396–3403.

Prospero, J.M., Glaccum, R.A., Nees, R.T., 1981. Atmospheric transport of soil dust from Africa to South America. *Nature* 289, 570–572.

QUARG, 1996. Airborne particulate matter in the United Kingdom. Third Report of the Quality of Urban Air Group, Department of the Environment, London.

Rajkumar, W.S., Siung Chang, A., 2000. Suspended particulate matter concentrations along the East West Corridor, Trinidad, West Indies. *Atmospheric Environment* 34, 1181–1187.

Reiff, J., Forbes, G.S., Spiessma, F.Th.M., Reynders, J.J., 1986. African dust reaching Northwestern Europe: a case study to verify trajectory calculations. *Journal of Climatology and Applied Meteorology* 25, 1543–1567.

- 1 Rodríguez, S., Querol, X., Alastuey, A., Kallos, G., Kakaliagou, O., 2001. Saharan dust contributions to PM₁₀ and TSP
3 levels in Southern and Eastern Spain. *Atmospheric Environment* 35, 2433–2447.
- 5 Ryall, D.B., Maryon, R.H., 1998. Validation of the UK Met. Office's NAME model against the ETEX dataset (1998). *Atmospheric Environment* 32 (24), 4265–4276.
- 7 Ryall, D.B., Maryon, R.H., Derwent, R.G., Simmonds, P.G., 1998. Modelling long-range transport of CFC's to Mace
9 Head, Ireland. *Quarterly Journal of the Royal Meteorological Society* 124, 417–446.
- 11 Ryall, D.B., Derwent, R.G., Manning, A.J., Simmonds, P.G., O'Doherty, S., 2001. Estimating source regions of European
13 emissions of trace gases from observations at Mace Head. *Atmospheric Environment* 35, 2507–2523.
- 15 Savoie, D.L., Prospero, J.M., 1977. Aerosol statistics for the northern tropical Atlantic. *Journal of Geophysical Research* 82, 5954–5964.
- 17 Schwartz, J., Marcus, A., 1990. Mortality and air pollution in London: a time series analysis. *American Journal of Epidemiology* 131, 185–194.
- 19 Schwartz, J., Dockery, D.W., Neas, L.M., 1996a. Is daily mortality associated specifically with fine particles? *Journal of the Air and Waste Management Association* 46, 927–939.
- 21 Schwikowski, M., Seibert, P., Baltensperger, U., Gäggeler, H.W., 1995b. A study of an outstanding Saharan dust event at the high-Alpine site Jungfraujoch, Switzerland. *Atmospheric Environment* 29, 1829–1842.
- 23 Simmonds, P.G., Derwent, R.G., McCulloch, A., O'Doherty, S., Gaudry, A., 1996. Long-term trends in concentrations of halocarbons and radiatively active trace gases in Atlantic
25 and European air masses monitored at Mace Head, Ireland from 1987 to 1994. *Atmospheric Environment* 30, 4041–4063.
- 27 Simmonds, P.G., O'Doherty, S., Nickless, G., Sturrock, G.A., Swaby, R., Knight, P., Ricketts, J., Woffendin, G., Smith, R., 1996. Automated gas chromatograph/mass spectrometer for routine atmospheric field measurements of the CFC replacement compounds, the hydrofluorocarbons and hydrochlorofluorocarbons. *Analytical Chemistry* 67, 717–723.
- 29 Simmonds, P.G., Seuring, S., Nickless, G., Derwent, R.G., 1997. Segregation and interpretation of ozone and carbon monoxide measurements by air mass origin at the TOR Station Head, Ireland from 1987–1995. *Journal of Atmospheric Chemistry* 28, 45–59.
- 31 Simmonds, P.G., Derwent, R.G., O'Doherty, S., Ryall, D.B., Steele, L.P., Langenfelds, R.L., Salameh, P., Wang, H.J., Dimmer, C.H., Hudson, L.E., 2000. Continuous high-frequency observations of hydrogen at the Mace Head baseline atmospheric monitoring station over the 1994–1998 period. *Journal of Geophysical Research* 105 (D10), 12105–12121.
- 33 Stevenson, C.M., 1969. The dust fall and severe storms of 1968. *Weather*, 126–132.
- 35 Torres, O., Bhartia, P.K., Herman, J.R., Ahmad, Z., Gleason, J., 1998. Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radiation: theoretical basis. *Journal of Geophysical Research* 103, 17099–17110.
- 37 WHO, 1995. Update and revision of the air quality guidelines for Europe. WHO Regional Office for Europe, Copenhagen, Denmark.