Final Report
Pollution Climate Modelling Contract (EPG 1/3/128)

ON THE RECURRENCE OF AIR POLLUTION EPISODES

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ABSTRACT

The Met Office was asked by Department of the Environment, Transport and the Regions to investigate meteorological aspects of air pollution episodes. This study was designed as a broad ranging, but preliminary, exercise to see what lessons could be drawn before engaging in more detailed (but more costly) modelling simulations. This work analyses the historic record of meteorological observations and pollution monitoring in the London UK area. At the Department's request, much of the effort focussed upon the meteorological records. The first aim was to identify weather related variables that correlate with different types of pollution episode. The second aim was to estimate the frequency of such weather conditions in the meteorological record. The third aim was to provide insights into the likely recurrence or frequency of episodes occurring again. This study of historic synoptic data does not use emissions inventory data. It was intentionally designed to not require the complexities of dispersion modelling.

Hourly meteorological observations from the station at London Heathrow Airport have been analysed in terms of frequencies from 1949 to 1997. Some analyses of hourly data extend to 1998. The most useful variables in the present context were wind speed, wind direction and temperature. A limited set of data for Lamb Weather Types from 1995 to 1999 was also analysed. The role of meteorological conditions in winter episodes of primary pollutants in London was clearly seen for Black Smoke, oxides of nitrogen, and PM_{10}. A possible pointer to summer ozone episodes in the UK was also noted. Wind directions from an easterly range 060-090° proved especially relevant to the secondary particulate episodes recorded at London Bloomsbury. Scope for research projects to develop the ideas in more detail is discussed.

Key words: air pollution, particles, PM_{10}, episodes, synoptic meteorology, observations, easterly flows, wind direction, pollution rose, pollutant flux, North Atlantic Oscillation.

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1. INTRODUCTION
1.1 Past Meteorology

Natural fluctuations in climate cause significant changes in air pollutant concentrations. When analysing episodes, providing advice for policies to manage future air quality, and in seeking to avoid conditions under which pollution episodes may occur again, a number of factors must be considered. For example, the technical feasibility and economic cost of engineering changes in order to meet future air quality Objectives depend upon the magnitude of emission controls that might be required by future regulations. Even if current emissions were assumed to be maintained, neither increasing in future years nor reducing, the same pollution climate as seen currently in the monitoring may not recur in future. This study therefore sought to use the longest available computerised archive of synoptic meteorological data from a station at London Heathrow Airport and address this question:

What is the likely frequency or probability that meteorology associated with pollutant episodes seen in London in the recent past may recur in the future?

The question was posed because of its importance to current policy initiatives. These include, for example, the likelihood of future weather conditions being such that the high particulate concentrations (PM_{10}) seen in March 1996 may be repeated in some future year. The Department specifically requested that simple analyses be undertaken, as speed was of the essence in addressing these questions.

Some aspects of policy can be developed by choosing reference data, or base years, from which to extrapolate the pollution concentrations into the future under various emission scenarios. Monitoring data were from Automatic Urban Network (AUN) sites supported by the Government and Devolved Administrations and stored in the UK National Air Quality Information Archive. They reveal that average PM_{10} concentrations throughout the country were much higher in 1996 than 1998. The difference seen in these concentrations seems too large to be explained just as arising from emissions controls. Instead, a much more likely explanation seems to be that the meteorology in 1996 was some how different from 1998, so that the one year might be seen as more polluted by fine particulates than the other. Malcolm et al. (2000) have shown the role of long-range transport and chemical reactions in forming secondary sulphate aerosol during March 1996, and briefly in July 1996. With this background knowledge that particulate concentrations differed in 1996 and 1998, and a role for secondary particulates being implicated by recent research, the question then arises:

What are the quantitative differences in the meteorology of 1996 vis a vis 1998?

A contrasting of years within the Strategy merits careful scrutiny, which this report provides.

1.2. The Air Quality Strategy and Choice of Year.

At the commencement of this work, understanding of the effects of different meteorological years on policy formulation was summarised in the Air Quality Strategy for England, Scotland, Wales and Northern Ireland (AQS, 2000). On particles, Section 8 of the Technical Annexe to the Strategy notes that levels of PM_{10} were exceptionally low in 1998 compared with data from 1992-1996. The explanation given in the Strategy was that 1998 had very low levels of transboundary secondary particles (i.e. imported from Europe) and a very low contribution from UK primary particles. The low primary PM_{10} contribution illustrates that 1998 meteorology in the UK was characterised by 'effective dispersion of pollutants'. This is also illustrated by the low levels of primarily locally emitted (e.g. from transport) pollutants, such as oxides of nitrogen and carbon monoxide, during 1998 (paragraph A124, AQS 2000). Statistics for PM_{10} (with TEOM correction factor 1.3.applied) illustrate 'the exceptionally low concentrations that were measured over the UK during 1998' (paragraph A126, and Table A34, AQS 2000). This new study complements work that has been carried out by John Stedman and colleagues at NETCEN (AEA Technology), and which was documented in the Air Quality Strategy AQS (2000).

The Air Quality Strategy follows the Airborne Particles Expert Group (APEG) in citing three sources for PM_{10}: primary (directly emitted), secondary (from reactions), and coarse. It describes the future projections for PM_{10} based on work at NETCEN. The method regresses observed PM_{10} against other pollutants to develop surrogates for the three components. Estimates of future concentrations are made (paragraph A129, AQS 2000). The method of projection is not under scrutiny in the present work. However, the choice of years on which the projections were based is of great interest and forms part of the rationale for the present work, as we now explain by reference to paragraph A132 in AQS (2000):

> "The analysis has been carried out for 1995 and 1996 because 1996 was characterised by a much higher than normal frequency of easterly winds, particularly in January and March, leading to a higher frequency of elevated concentrations associated with the transport of polluted air from mainland Europe to the UK. In contrast, 1995 was a 'normal' year in this context."

In short, the present study starts from these assumptions:

1. 1995 was 'normal'. It serves as a policy starting year.
2. 1996 had high concentrations of PM_{10} with an elevated frequency of easterly flows (especially in January and March).
3. 1998 was 'low' (low primary concentrations; low European role).

In looking for meteorological factors that influence the formation of pollution episodes, the authors were asked to provide some deeper insights into these assumed comparisons of air pollution meteorology from recent years. Did these assumptions withstand detailed scrutiny, or were there new lessons to be drawn? What was the difference(s) between these years?

1.3 The Return of Secondary Episodes

Further, in order to address the likely recurrence of future secondary PM_{10} episodes, it is necessary to enquire:

What aspects of the meteorology favour secondary PM_{10} episodes and what is the likely frequency or probability of such conditions appearing again in future?

To address these questions, the study looked at the meteorological record, and sought links with the pollution record. A key aspect of the work was to establish which meteorological variables were most relevant to the problem in hand. The results are described below. First we summarise the data retrievals.

1.4 Data Retrievals

The focus has been to access meteorological archives of synoptic observations from London Heathrow Airport. Key questions were:

1. Could the archives be accessed successfully for the long period of interest?
2. Could synoptic observations that characterise pollution episodes be identified?
3. Could existing retrieval routines assist in making statistical objective comparisons of one year with another, such as 1996 versus 1998, or even one year say 1996 versus a long run of data representing typical conditions? What is a typical or atypical year of meteorology in the context of episodes?
4. Could conditions associated with secondary particulate episodes (as in March 1996) be identified in the synoptic observations, and were these observations sufficient to unequivocally characterise them? Whilst primary winter smog episodes were thought to be relatively easy to characterise, there was no clear information to say whether the secondary episodes could be unequivocally associated with synoptic data from a single station. It may turn out that more complex analyses, such as trajectories, or analysis of multi site data (e.g. geostrophic wind), might be needed.
5. Would the results of such broad ranging analyses be useful tools to apply when analysing the results of more complex modelling, such as trajectories or NAME, especially when used on numerical data from the Met Office climate model or from ECMWF Analyses.

The work followed this sequence: We retrieved hourly and daily data, confirming that retrieval was practicable (Point 1) back to 1949. Synoptic meteorological data for primary pollutant winter episodes were then studied. Once this seemed feasible (Point 2), the work turned to looking for ways of year-to-year comparison (Point 3). Finally, this preliminary study turned its attention to the real task of trying to seek features for the secondary particulate episodes (Point 4). Their frequencies and likely recurrence were then evaluated.

The question of whether the data from London Heathrow is representative of that reaching central London has been under debate for several years. For the present study, the site represented one of the longest data sets available for this sort of analysis. No allowance has been made for the heat island effect widely seen over London, resulting in temperatures that are 2-5°C higher than the surrounding areas and differing wind fields due to the buildings contribution to the surface roughness. London Heathrow is well exposed with no strong directional bias.

1.4.1 Hourly Retrievals

Hourly and daily average synoptic observations (1949-1998) of important meteorological parameters have been retrieved using the program ADMSEQRH. The variables were temperature, 10 minute spot wind speed, wind direction, Pasquill stability class, precipitation rate, cloud cover and relative humidity. These are sufficient to identify winter smogs as associated with light wind, below or near freezing temperatures, and relative humidity near 100% (near saturated). Notable episodes like 1952, 1991 etc could be seen with such conditions evident in the meteorological record. Had summer smogs been the focus of the study, then sunshine hours would also have been retrieved. Data extracted by ADMSEQRH were manipulated in a spreadsheet program (Microsoft Excel) and plotted. The hourly data are too extensive to list here, but are used in the calculation of indices, and drawing of rose plots, as discussed later from Sections 3.3 on. Daily averaged observations were also used where only daily values of pollutants were available, for example, for black smoke or daily sulphate.

1.4.2 Statistical Summaries

Data for a sample set of years have also been retrieved using a statistical analysis program BLDSTAB. This sorts the data and counts them into hours observed having each property or magnitude. The results from BLDSTAB consist of three main arrays, each showing the counted observations arranged into groups as follows: Pasquill Stability (hours), Boundary Layer (hours), and Wind Sector (frequency). The frequency arrays are too extensive to include here, but are discussed later in Sections 3.1-3.2.

1.4.2.1 Pasquill Stability Category:

The stability of the atmosphere is diagnosed for each hour from the wind speed, time of day, solar radiation, and cloud cover. A seven class scheme is used to describe the atmospheric stability: A, B, C, D, E, F and G. Class D represent neutral stability, as in cloudy, windy conditions. Unstable conditions, with enhanced vertical mixing, as in hot sunny still weather, are denoted A to C. Stable conditions, with suppressed vertical mixing, often associated with winter episodes from ground level sources in cities, are denoted F to G. The routine also allows class D to be divided by day and night; for preliminary analysis it is convenient to aggregate all D categories together. The array of Pasquill Stability Category shows how often (number of hours) each class occurred in each month, and then in the whole year. Hours are counted and sorted by time of day, so it is possible to see for any month, or for the whole year, at a glance how often say Class D was observed at 1500 hours. (All results use GMT). At the bottom of every array the final row also shows how many hours in the month or year (irrespective of time of day) was in each of the Classes A to G. The monthly arrays enable a given month, say December, in one year to be compared with another. The annual summary array allows one whole year dataset to be compared with that for a different year.

1.4.2.2 Boundary Layer Depth:

The boundary layer thickness is a measure of the height from the ground through which pollutants might potentially be mixed. With shallow vertical mixing due to inversions over a city, high pollutant concentrations can develop. Boundary layer depth is also an important consideration when modelling the long-range transport of species, as it can limit the depth of atmosphere through which the emissions are spread. Arrays of boundary layer thickness in metres are generated by BLDSTAB for every month in the year, and as an annual summary. These depths are sorted in steps or ranges every 200 metres. Again, data are sorted by hour of the day, so the number of hours in the month, or in the whole year, having a particular boundary layer depth, can be identified. Likewise, the total number of hours in the month, or the whole year, of each boundary layer depth is put at the bottom of each array, irrespective of the time of day. The monthly arrays enable the depths occurring in a given month, say December, and the frequencies or number of hours of these depths, to be compared from one year to another. In a similar fashion, the year summary array allows one year to be contrasted with another.

1.4.2.3 Wind Sector Frequency Analysis:

These arrays show the frequency of the time in which the wind blew from each of 12 sectors (30 degrees wide), for all wind speeds (Table 1). Hence the frequency of easterlies for example in a given month, say March, can be seen in a given year, like 1996. The same array also shows the frequency that the wind was in each of 5 different wind speed groups, irrespective of the direction it came from. The frequencies of calms are arrayed separately (reported as zero speed and zero direction, because once the anemometer stops turning, both speed and direction are undefined quantities). Likewise, after all the months, there is a corresponding array for the whole year. With these arrays the frequency with which a given combination of wind speed, wind direction, and Pasquill stability class, has occurred during the month, or during the whole year can be measured.

BLDSTAB is a useful tool for the comparison of one year with another. Unfortunately it cannot be run on archives dating from 1997 onwards due to changes in the archiving procedures. In principle, a further useful feature of BLDSTAB is that it should be able to accept up to a ten year run of data, although this has not been tested in this study. This could facilitate the estimation of how often typical conditions may occur over a long run of data, perhaps for use as a yardstick on which to test any year of special interest like 1996 or 1998. It may also be feasible to conduct seasonal comparisons from year to year should this be needed.

To summarise,

1. Data have been retrieved back as far as 1949 for London Heathrow Airport.
2. There is a change in archive structure from 1997.
3. Hourly data are useful for calculating indices, and drawing directional rose plots.
4. Frequency arrays suggest a useful way of comparing the same months in different years, or comparing different years as annual datasets.

2. EPISODES

2.1. Primary Pollutant Episodes

The work begins with an analysis of the frequency of calm and of light winds. They have special significance for urban air quality. Historic hourly data were therefore extracted using ADMSEQRH as noted earlier. Table 2 shows major air pollutant dates for London in the literature. The time period is so long (48 years) that the nature of the pollutants monitored has changed from Black Smoke to fine particulates as PM_{10}, also sulphate and oxides of nitrogen. There seem few, if any, pollutant measurements (for the same pollutant at a given site) that span the whole study period. It is the meteorological record which has the longer span. The main episodes and their dates are tabulated for reference later in this study. For convenience on these dates, the Table also shows the values we calculated for the temperature-wind index *Tu* as discussed below in Section 4.1.

TABLE 1 Example of output stability frequency analysis - mean percentage frequency of time from BLDSTAB for London Heathrow Airport 1952 December and Annual; 1991 December and Annual; 1996 March and Annual.

DECEMBER 1952

WIND SPEED	WIND DIRECTION (DEGREES)													ALL
(KNOTS)	CALM	346-015	016-045	046-075	076-105	106-135	136-165	166-195	196-225	226-255	256-285	286-315	316-345	DIRNS
0	20.97	0	0	0	0	0	0	0	0	0	0	0	0	**20.97**
1-3	0	1.61	0.4	0.13	0	0	0.13	2.69	0.4	1.21	1.34	0.94	0.54	**9.41**
4-6	0	3.09	0.67	0	0	0.13	0.94	2.69	2.55	3.36	3.63	1.61	1.34	**20.03**
7-10	0	0.67	3.23	0.81	0	0	0.94	2.69	4.7	3.49	5.78	0.94	1.61	**24.87**
11-16	0	0.67	1.75	1.75	0	0	0.67	2.82	3.9	3.23	3.63	1.21	0.67	**20.3**
17-98	0	0	0	0	0	0	0	0.27	0.13	0.94	1.75	1.34	0	**4.44**
ALL	**20.97**	**6.05**	**6.05**	**2.69**	**0**	**0.13**	**2.69**	**11.16**	**11.69**	**12.23**	**16.13**	**6.05**	**4.17**	**100**

ANNUAL 1952

WIND SPEED	WIND DIRECTION (DEGREES)													ALL
(KNOTS)	CALM	346-015	016-045	046-075	076-105	106-135	136-165	166-195	196-225	226-255	256-285	286-315	316-345	DIRNS
0	12.06	0	0	0	0	0	0	0	0	0	0	0	0	**12.06**
1-3	0	1.4	0.89	0.76	0.52	0.48	0.39	0.81	0.58	1.06	1.75	1.42	0.94	**11.01**
4-6	0	2.07	1.98	1.28	1.12	0.75	0.68	1.42	1.66	2.44	3.8	2.46	1.74	**21.4**
7-10	0	2.12	2.74	1.96	1.68	0.63	0.96	2.09	3.4	3.31	5.05	2.71	1.89	**28.55**
11-16	0	0.74	1.62	1.12	1.25	0.33	0.44	1.95	3.96	3.59	3.26	1.41	0.85	**20.51**
17-98	0	0.02	0.15	0.49	0.17	0.01	0.01	0.68	1.82	1.42	1.17	0.31	0.2	**6.47**
ALL	**12.06**	**6.35**	**7.38**	**5.6**	**4.75**	**2.2**	**2.48**	**6.96**	**11.43**	**11.82**	**15.04**	**8.31**	**5.64**	**100**

DECEMBER 1991

WIND SPEED	WIND DIRECTION (DEGREES)													ALL
(KNOTS)	CALM	346-015	016-045	046-075	076-105	106-135	136-165	166-195	196-225	226-255	256-285	286-315	316-345	DIRNS
0	18.41	0	0	0	0	0	0	0	0	0	0	0	0	**18.41**
1-3	0	2.82	5.38	1.75	0.94	1.61	2.42	2.02	2.42	1.21	2.02	3.23	1.61	**27.42**
4-6	0	0.4	4.3	1.61	1.75	1.88	0.54	1.21	1.88	0.94	1.75	0.27	0.81	**17.34**
7-10	0	0	2.15	2.69	4.44	0.94	0	0.4	2.15	3.09	1.88	0.81	0.27	**18.82**
11-16	0	0	0.13	0	0.67	0	0	0	0.81	7.66	4.57	0.4	0	**14.25**
17-98	0	0	0	0	0	0	0	0	0.54	2.02	1.08	0.13	0	**3.76**
ALL	**18.41**	**3.23**	**11.96**	**6.05**	**7.8**	**4.44**	**2.96**	**3.63**	**7.8**	**14.92**	**11.29**	**4.84**	**2.69**	**100**

ANNUAL 1991

WIND SPEED	WIND DIRECTION (DEGREES)													ALL
(KNOTS)	CALM	346-015	016-045	046-075	076-105	106-135	136-165	166-195	196-225	226-255	256-285	286-315	316-345	DIRNS
0	5.65	0	0	0	0	0	0	0	0	0	0	0	0	**5.65**
1-3	0	3.07	2.57	0.89	0.5	0.54	0.89	1.7	2.25	2.04	2.42	2.61	2.2	**21.69**
4-6	0	1.85	3.34	1.59	1.03	2.03	1.45	2.49	3.06	2.83	3.15	2.12	1.76	**26.7**
7-10	0	2.09	3.4	1.75	2.05	1.51	1.44	3.11	4.53	3.81	2.43	1.11	1.47	**28.7**
11-16	0	0.97	1.46	0.81	0.81	0.3	0.68	1.16	3.76	3.38	1.07	0.11	0.17	**14.69**
17-98	0	0.01	0.11	0	0.05	0.01	0.08	0.41	1.07	0.66	0.14	0.01	0.01	**2.57**
ALL	**5.65**	**7.99**	**10.89**	**5.03**	**4.44**	**4.38**	**4.54**	**8.87**	**14.67**	**12.73**	**9.21**	**5.97**	**5.62**	**100**

MARCH 1996

WIND SPEED	WIND DIRECTION (DEGREES)													ALL
(KNOTS)	CALM	346-015	016-045	046-075	076-105	106-135	136-165	166-195	196-225	226-255	256-285	286-315	316-345	DIRNS
0	1.21	0	0	0	0	0	0	0	0	0	0	0	0	**1.21**
1-3	0	3.49	4.17	0.94	0.54	0.27	0.81	1.61	1.08	1.48	2.69	3.63	3.76	**24.46**
4-6	0	4.7	3.36	5.38	1.34	1.75	1.48	0.81	0.67	0.4	1.21	1.61	3.63	**26.34**
7-10	0	3.63	4.03	8.74	5.51	2.55	0.27	0.4	0.27	0.27	0.67	0.27	2.28	**28.9**
11-16	0	0	2.42	5.11	8.06	2.55	0	0	0	0	0.13	0	0	**18.28**
17-98	0	0	0	0.13	0.67	0	0	0	0	0	0	0	0	**0.81**
ALL	**1.21**	**11.83**	**13.98**	**20.3**	**16.13**	**7.12**	**2.55**	**2.82**	**2.02**	**2.15**	**4.7**	**5.51**	**9.68**	**100**

ANNUAL 1996

WIND SPEED	WIND DIRECTION (DEGREES)													ALL
(KNOTS)	CALM	346-015	016-045	046-075	076-105	106-135	136-165	166-195	196-225	226-255	256-285	286-315	316-345	DIRNS
0	2.17	0	0	0	0	0	0	0	0	0	0	0	0	**2.17**
1-3	0	2.97	2.27	0.51	0.58	0.81	1.54	2.07	2.31	2.38	2.45	2.87	3.06	**23.82**
4-6	0	3.39	3.14	1.65	0.87	1.66	1.4	2.5	3.29	2.23	2.02	1.91	2.49	**26.56**
7-10	0	2.31	3.4	2.49	1.67	1.95	1.23	2.97	3.65	3.68	2.03	1.17	1.46	**28.02**
11-16	0	0.71	1.56	3.23	2.11	0.87	0.71	2.05	3.22	1.46	0.67	0.34	0.52	**17.44**
17-98	0	0.11	0.16	0.3	0.16	0.06	0.15	0.26	0.58	0.15	0.07	0	0	**1.99**
ALL	**2.17**	**9.49**	**10.53**	**8.19**	**5.38**	**5.34**	**5.02**	**9.86**	**13.06**	**9.89**	**7.23**	**6.3**	**7.54**	**100**

Table 2 Dates of major air pollution episodes in London during the study period (1949-98) and associated values of the Winter Index, *Tu*. Sites indicated where known.

Dates	Pollutant	Max concentration	Averaging time	Winter Index, *Tu*
5-8 Dec 1952	Black smoke; SO_2	4460 $\mu g\ m^{-3}$ 3700 $\mu g\ m^{-3}$	Daily	30.1 – 42.4
3-6 Jan 1956	Black smoke; SO_2	1700 $\mu g\ m^{-3}$ 2800 $\mu g\ m^{-3}$	Daily	19.3 – 36.8
3-5 Dec 1957	Black smoke; SO_2	3000 $\mu g\ m^{-3}$ 2800 $\mu g\ m^{-3}$	Daily	16.5 – 22.4
4-7 Dec 1962	Black smoke; SO_2	8046 $\mu g\ m^{-3}$ (Hampstead) 5873 $\mu g\ m^{-3}$ (St Marylebone)	Daily Daily	8.1 – 35.6
7-24 Jan 1963	Black smoke; SO_2	1607 $\mu g\ m^{-3}$ (Tottenham) 1481 $\mu g\ m^{-3}$ (Fulham)	Daily Daily	2.0 – 20.6
11-15 Dec 1991	NOx; NO_2; Black smoke	1567 ppb; (Bridge Place) 423 ppb (Bridge Place) 209 $\mu g\ m^{-3}$ (Westminster)	1 hour; 1 hour; daily	12.3 – 23.4
15 Jan – 1 Feb 1996	PM_{10}	105 $\mu g\ m^{-3}$ (Bloomsbury)	Running 24 hour average	1.8 – 8.1
10-26 Mar 1996	PM_{10}	99 $\mu g\ m^{-3}$ (Bloomsbury)	Running 24 hour average	1.7 – 6.3

From knowledge of pollution behaviour, the variables that are most likely to be associated with elevated concentrations of urban primary pollutants in winter are:

- Light winds,
- high frequency of calms,
- low temperature,
- high humidity and fog,
- increase in emissions associated with low temperatures,
- strong static stability (strong increase in potential temperature with height) and
- shallow or ground based inversion.

This study has not analysed the temperature lapse rate data for static stability or inversion depth however. Emission changes are only allowed for indirectly via temperature expressed as degree-hours.

Winter smog episodes are well known features of the pollution climate record. Therefore data were processed by BLDSTAB for years which the literature shows had serious winter smogs in London. These include 1952, 1956, 1957, 1962 and 1991. Other years were also retrieved, to get reference data for purposes of comparison. In addition to these statistical data month by month from BLDSTAB, hourly observations were also retrieved for London Heathrow Airport, using ADMSEQRH. The first task was simply to study the analyses manually to address the question 'was it possible to identify features of the meteorology that would be characteristic of such episodes'. These preliminary results supported the hypothesis that the key characteristics were:

- above average frequency of calm or light winds in the month,
- low temperatures, and
- often very high relative humidity i.e. conditions we might traditionally associate with winter fogs.

These characteristics led to the calculation of an index, *Tu* (Section 4.1) based on light wind and low temperature, but without using humidity.

2.2 Secondary Pollutant Episodes and PM_{10}

Monitoring of fine particles as PM_{10} dates only from recent times on the establishment of the Automatic Urban Network (AUN) of urban centre sites in the UK. Table 3 shows the dates when the 24-hour average concentration of fine particles measured as PM_{10} exceeded 50 µg m^{-3}. There is no immediately discernible seasonal pattern or trend in these exceedence days. It was noticed, however, that a significant proportion of exceedences occur in clusters of days, typically three days or more. This is discussed later (Section 6.4 Weather Types). On identifying no simple correlate for secondary episodes, we also briefly investigated the possible role of the NAME model for addressing the recurrence of secondary episodes of fine particles.

The monitoring records at 19 urban stations and 1 rural station for the March 1996 PM_{10} episode in the UK have been analysed by Stedman (1997). This episode was geographically widespread; it was also prolonged (10-26th). He concludes there was a significant long-range transport component in this episode. A characteristic of secondary particles (sulphates and nitrates formed from reactions of SO_2 and NO_x) is their slow formation, prolonged lifetime in the atmosphere, and much more uniform contribution to PM_{10} across the country than just primary particles on their own. In this episode, Stedman (1997) reported very similar concentrations at all the sites, including the rural station. Back trajectories showed European flow during this period.

King and Dorling (1997) studied the same March 1996 episode. They also investigated an earlier episode in January-February 1996 (15th Jan-1st Feb). In addition to the PM_{10} monitoring data and back trajectories, they studied the Lamb Weather types for the period from November 1993 to March 1996 inclusive. They suggested that a majority component came from outside the immediate monitoring area, had elevated values associated with air masses from mainland Europe, and exceedences could be linked to national and trans-boundary pollution. The January-February 1996 episode had ‘predominantly pure easterly flow’ and broke down ‘when anticyclonic conditions arrived’. In view of these, and the other episodes they studied, their conclusion was that

Table 3 Dates when the concentration of PM_{10} as a 24-hour average exceeded the 50 µg m^{-3} standard at London Bloomsbury during the period 1992-1998.

Month	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	Total
Jan	23, 24, 25, 30	1		3	15, 16, 19, 21, 22, 23, 24, 25, 27, 30	11, 25		**18**
Feb	1, 25	2, 5, 10, 11, 12, 14, 15	14, 18, 19, 25		1, 2, 7			**16**
March	5	2, 15		14	10, 11, 13, 14, 15, 16, 17, 18, 19, 20, 25, 26	9, 10, 11, 12	16	**21**
April	8, 9, 10, 11	14, 15, 29, 30		25	7, 10	9, 10		**13**
May		1, 11, 12, 24	11	1, 2, 3, 4, 5, 6, 23			13, 14	**14**
June	1, 9, 10, 23, 25, 26, 27, 28, 29, 30	6, 8, 9, 10	13, 24	30	6, 7, 26			**20**
July	29, 30, 31	2	1, 2, 12, 21, 24	25, 31	21			**12**
Aug	19	19		1, 11, 12, 15, 22, 23	18, 19, 20	10, 11, 19, 20		**15**
Sept	17				15	29		**3**
Oct			10, 11, 14, 15	11, 12, 13, 14, 31		2, 30		**11**
Nov		1, 2, 3, 5, 16, 19, 24, 28	22, 25, 29	7, 12	14	2, 12		**16**
Dec	17, 30, 31		1, 9, 23	17	5			**8**
Total	**29**	**32**	**22**	**27**	**37**	**17**	**3**	**167**

> "...particulate matter entering the country from the South and East form a significant contribution to, and the majority component of, breaches of the 50 µg m^{-3} limit...".

Whilst this may not be proven in their paper, King and Dorling (1997) do suggest there is 'a case to answer'.

The results of Stedman (1997) and Dorling and King (1997) are consistent with the results in Malcolm *et al* (2000), described elsewhere. All three studies, whilst limited by the relatively short historical archive of PM_{10} monitoring, point to the importance of easterly flows. It is these flows that we have used as a marker in trying to estimate the broad frequency of episodes driven by secondary aerosols. Stedman (personal communication) finds sulphate is a useful marker of secondary episodes. The complexities of analysis that arise with data on PM_{10} are due to the multiplicity of natural sources, primary emissions, and secondary formation.

3. ANALYSES OF ARCHIVED OBSERVATIONS

In this Section we report the results of simple analyses using the long run of data from London Heathrow Airport. We begin with the frequency results, Sections 3.1-3.2, and then follow with analyses using sequential hourly data, Sections 3.3 onwards.

3.1 Wind Speed Frequencies

The archives (1949-97) were processed as noted above by BLDSTAB to generate frequencies of different wind speeds, wind directions, Pasquill stability classes, and boundary layer depths. Frequencies of joint categories were also generated. 1998 was added by calculating the frequencies of different wind speeds and wind directions.

After studying the frequencies of wind speeds data, it was decided to plot the frequency of calms that were reported each year. The aim was first to see if the incidence of calm conditions was high in months and years in which severe episodes were recorded. The Department had asked that years like 1996 and 1998 be compared, but we found that pollution episodes are only a few days in duration, so the frequency of calms on a monthly basis is more sensitive to shorter periods of episode type conditions than it is on an annual basis. Annual summaries also transpire to be insensitive comparators of episode conditions from one year to another. Figure 1 shows the frequency of calms for *all months* in the year; Figure 2 shows the frequency of calms in *December* of each year. The notable December episodes of 1991 (NO_x) and 1952 (Black Smoke) were clearly seen, even when data are taken a whole month at a time.

More surprisingly, a general decline in the frequency of calms was seen from 1949 through to about 1975. This result was not anticipated. It could be an artefact, if the more modern anemometers have a lower stalling speed. However, this would seem unlikely since there is no step change seen in either Figures 1 or 2. Figure 3 shows Dan Hollis' study of the hourly mean wind speed at Heathrow (rather than the 10 minute spot winds used in Figures 1 and 2). There is a marked step change in the data around the middle of 1981. This seems to correlate with a change from manual to continuous automatic recording of wind speed. The step change was not recorded in the spot wind, since these continued to be produced manually from an anemograph trace. Over the period covered by this study, the anemometer at Heathrow was moved in December 1960, April 1973 and December 1982. These changes do not show up obviously in an analysis of frequency of calms, although the move in December 1960 may have contributed somewhat to the marked difference between the 1950's and the rest of the data.

The development of buildings around London Heathrow might be expected to decrease mean wind speeds, perhaps increasing the incidence of calms. However, a decrease in the number of calms, as in Figure 1, goes in the opposite trend to that expected for a general rise in surrounding surface roughness. Perhaps large buildings near the anemometers have caused some flow acceleration, but again a step change might be expected. Furthermore, a large peak in the frequency of calms is seen in December 1991, just when London had a notorious winter smog of elevated NO_2 and Black Smoke concentrations (Table 2). In the absence of other information, it was concluded that the decline in frequency of calms was a real effect, but that in occasional years, as seen in 1991, a prolonged period of calm coldness and associated high pollution occurred. Such a period of cold calm weather might occur again in the future. Quite when is unclear. The question then intrigues us; are conditions like December 1991 coupled in some way to long term slow oscillations in climate, perhaps linked to some underlying geophysical mechanism? The discussion of the North Atlantic Oscillation (Section 6) has some relevance to this.

Using the frequency arrays from BLDSTAB:

1. the wind was calm for 20.97% of the month in December 1952 (Table 1);
2. in December 1991 it was calm for 18.41% of the month;
3. but in December 1996 it was calm only for 2.55% of the month.

There were notorious winter smogs of primary pollutants in the Decembers of 1952 (Smoke/SO_2) and 1991 (NO_x), but not in 1996. Then in March 1996, it was calm for just 1.21% of the time, suggesting that the particulate episodes in March 1996 were most unlike the winter smogs of 1952 or 1991, since the incidence of calms was so much lower in March 1996 than December in 1952 or 1991. For interest, the month in 1996 with the greatest incidence of calms was October, with just 4.03% calms in the month. These results shows just how closely tied is the pollution climate month by month to the meteorology of the period.

As a further check on the wind records, the results for speeds in the ranges 1-3 knots were plotted, Figures 4 (all months) and 5 (Decembers), and proved to be somewhat similar in behaviour to the frequencies of calms. This is no doubt because both calms and light winds require similar conditions. Broadly, lighter winds were less frequent in the 1970s onwards than in the 1950s. From a policy point of view, there is nothing in these historical data to tell us whether such a trend remains, or whether calm conditions will be much more frequent in the future. Climate simulations are the main source of information for possible future wind speeds. Calm conditions are associated with certain weather types, and these are discussed later.

Finally, the frequencies for 4-6 knots, Figures 6 (all months) and 7 (Decembers), and for 7-16 knots, Figures 8 (all months) and 9 (Decembers) were plotted. The frequency for 4-6 knots has tended to rise as the frequency of calms decreased. The frequency of strong winds from 7-16 knots has been fairly steady though showing a slow decline over the decades. However, the very strong winds seen in October 1987 widely attributed to the tail-end of a hurricane, are not seen in the monthly or annual average data. Changes in wind speeds, if not due to local site developments or instrumental factors, may be linked to wider changes in large scale meteorological processes. They will have important implications for the pollution climate.

3.2 Wind Direction Frequencies

Wind speeds (above) are especially relevant to episodes due to poor dispersion of locally emitted primary pollutants, like SO_2, Black Smoke, CO, or NO_x. In studying particles we found there was an important association with wind direction. Consequently an analysis of the frequencies of wind directions was carried out. These frequencies for data observed at London Heathrow Airport were generated during the work for Section 3.1 above.

Frequencies are the percentage of observations that the wind was from the stated directions. Calms are not plotted (but their frequency is allowed for). The BLDSTAB output sorts the data to find the frequency of each direction by wind speed group (calms, 1-3 knots, 4-6 knots, 7-10 knots, 11-16 knots, 17-98 knots, or all speeds). Thus it is possible to plot the frequency from an eastern quadrant of all wind speeds, or of just moderate wind speeds. Calms and the strongest wind speeds do not seem strongly associated with elevated PM_{10} concentrations, but moderate easterlies do, as will be discussed later in this report.

Graphs of wind direction class frequency (using BLDSTAB output, see Section 1.4.2) were plotted for the years 1949 to 1997. Figure 10 shows how the proportion of time that the wind was from the east has varied over the years ranging from 8.5% in 1967 to 23.2% in 1976. In 1995, 1996 and 1997 the frequencies were 16.4, 18.9 and 20.1%, respectively. Figure 11 looks at the frequencies of moderate easterly wind speeds from 1-6 knots; this is because the pollution roses (Section 3.3) highlight the association of raised PM_{10} with such winds. They range in frequency between 2.7% and 7.7%. Frequencies in 1995, 1996 and 1997 were 6.9, 6.5, and 7.1%, respectively.

We recall that in the Air Quality Strategy, 1995 was our 'normal' year whilst 1996 was nominally a 'poor' year (see Section 1.2 above), but our analysis of the frequency with which easterlies are seen year on year does not make 1996 any more or less unusual than 1995 or 1997. In general, the occurrence of easterly flows varies over a wide range. The variations in incidence from year to year are such that no trend is immediately discernible. There is at most a rise from around 5% to 6% in the frequency of moderate easterlies seen at Heathrow Airport over the 49 year study period. However, this upward drift is less than the year-to-year variation.

The figures show the long-term changes (or fluctuations) in reported frequencies of winds from the Eastern quadrant (Figures 10 & 11). These are of especial interest for particles, which have local low level and widely distributed sources (e.g. traffic etc), elevated large point sources (e.g. incinerators, power stations, and industrial stacks), a background (natural or biological), and a variable European contribution (secondary formation). However no clear pattern was seen.

When considering the data month by month, the story is different. With regard to particulates and returning to March 1996, this showed a high frequency of 20.30% North Easterly and 16.13% Easterly winds (from 046 to 075 degrees, and 076 to 105 degrees, respectively, measured clockwise from North). This is consistent with results reported elsewhere (e.g. APEG 1999; Dorling and King, 1997; Stedman, 1997; Malcolm et al., 2000) on the trajectories during the March 1996 incidents. It is not possible at this preliminary stage however to draw any further conclusions on meteorological statistics associated with secondary pollutant episodes; much work needs to be done as this is not a simple matter. For example, visibility might seem to be a variable related to or even dependent upon secondary pollutants, as discussed briefly in section 5 below. However, later in this report, we discuss the role and relevance of air mass and weather types for this work. We shall see below that some tentative conclusions may be drawn in this fashion on the incidence of easterly air masses. Our work with pollution rose plots also sheds light on this aspect of the pollution climate.

3.3 Rose Plots

The rose diagrams are polar diagrams showing the wind directions in the usual convention, with the wind direction being the direction (degrees clockwise from North) from which the wind was blowing. The data are resolved in steps of 10 degrees, which is normal reporting convention. The plots used hourly meteorological observations from London Heathrow retrieved via ADMSEQRH. Pollution data show PM_{10} in µg m^{-3} measured hourly means from the AUN site at London Bloomsbury. All PM_{10} concentrations used in this report were the original TEOM value and were not 'corrected' to any equivalent gravimetric figure, i.e. no factor such as 1.30 has been applied. In the plots discussed below, all data reported as calm were excluded. In the following sections, the meteorological data are plotted, followed by the PM_{10} data.

3.3.1 Wind direction frequency rose

Figures 12-16 plot the frequency (%) that the wind was from each direction, obtained simply by sorting the data and counting occurrences in each direction. Their frequency is the percentage of the total observed hours. There are 36 individual wind directions at 10 degree steps and therefore uniformly distributed winds would have a frequency of 2.78%. Figures 12, 13, 14, 15 and 16 show the frequencies of wind directions recorded at London Heathrow Airport over each decade studied, from 1949 to 1998. This is a compact way of studying many years of data.

The larger radius shows the most frequent or prevailing winds: Prevailing SW winds from 230° reach 7.4% in 1994. In most years, this prevailing SW flow was present from 4-7% of the time. In contrast, winds from the opposite NE direction are less common, usually 2-4%. Easterly winds are even less common, 0.5-2.5%, of the time. In 1996 the frequency of winds from 20-50° was ~4%, but only ~1-3% for directions 60-160°. 1998 also showed a similar low incidence of winds from an easterly quadrant. Easterly types may be infrequent, but the pollution rose (Section 3.3.4, below) reveals their importance for the fine particulate pollution climate.

3.3.2 Temperature rose

Figures 17-21 plot the distribution of temperature by wind direction at London Heathrow Airport from 1949 to 1998. Most directional temperature averages lay between 9° C and 12° C. For many directions this range applies and the years studied are all fairly similar to one another. Year to year variation in temperature was greatest in the same easterly quadrant seen above in the pollution rose. In 1996, directions 10-20°, and also 30-90°, were noticeably colder, averaging ~6° C. Clearly, during 1996, some flows from near N and NE were unusually cold. By way of contrast in 1998, the wind from this quadrant was warmer, reaching a directional average of 15.5° C in flow from 80°. This would possibly indicate that during 1996 the winds from the N and NE occurred primarily during the winter months. In 1998, it seems more likely that the winds from this quadrant occurred during the spring or summer.

3.3.3 Wind speed rose

Figures 22- 26 show the average wind speed at London Heathrow Airport according to wind direction. Calms are excluded: by convention they are recorded with zero speed and zero direction. The radius shows the average wind speed (measured at a height of 10 m in m s^{-1}) associated with winds from each direction. A strong SW and NE bias can be seen with these directions attaining the higher averaged wind speeds. Going clockwise from SE round to N (i.e. 120° round to 350°) the wind speeds in all years were close together in magnitude. They rise smoothly in SE flow from ~3 m s^{-1} up to a SW maximum of ~4.5 m s^{-1} and fall smoothly again to a minimum of ~2.5 m s^{-1} near N. Winds from the NE direction have a greater year to year variation in wind speed than any other quadrant. For flow from 60°, 1993 and 1998 showed an average speed of ~3.0 m s^{-1}, whereas 1996 had a much faster average speed of 5.5 m s^{-1} for the 60° direction. Other years, and the flows around a NE direction, all fell between these limits of 3.0-5.5 m s^{-1}.

3.3.4 Pollution rose

Pollution roses are a traditional tool used to identify important directions for the pollutant contributions at a monitor. The average concentration of fine particles, is measured as PM_{10} (µg m^{-3}) at London Bloomsbury. This site has the longest readily accessible record of hourly PM_{10} data in the London area. Data from this AUN station can be accessed via the National Air Quality Information Archive. These data start in 1992. The London Bloomsbury concentrations are sorted according to the wind directions at London Heathrow Airport, the hourly PM_{10} concentrations are then averaged and used to plot the PM_{10} pollution rose, Figure 27. There are differing numbers of observations associated with each direction and their frequencies were shown in the wind frequency rose (Figures 12-16 above). As noted above, SW types are most frequently seen, whilst air from the NE and E were least frequent. The radius is proportional to the average air pollution associated with the wind from each direction. For the PM_{10} data in the years studied, these directionally averaged concentrations were always greater than 20 µg m^{-3} in all wind directions.

Relative to the other wind directions, air from the SW had the least PM$_{10}$ burden. At 230°, concentrations ranged from 19-23 μg m^{-3}. However, with winds from the ENE, E, and ESE (directions 50-110°) the pollutant burden was much larger, ranging from 27-50 μg m^{-3}. More generally, the usual range of concentrations in the other directions was 20-35 μg m^{-3}. The shape of the pollution rose is different from the wind frequency rose; the least common wind directions carried the higher pollutant concentrations. There is quite clearly a polluted easterly sector or quadrant for London Bloomsbury (directions 50-140° from N, some 90° wide) which has elevated PM$_{10}$ values. This applies even in 1998, the year in which the easterly elevation of particle concentrations was least pronounced (but still ~27-30 μg m^{-3}). It so happens that 1996 stands out in severity, reaching 47-50 μg m^{-3} for the three directions from 60°, 70° and 80° from N. However the years 1993 and 1995 were not so far behind when viewed on the pollution rose, approaching 44 and 43 μg m^{-3} for PM$_{10}$ concentrations respectively from the easterly quadrant. From this information, it would be wrong to interpret 1996 as being unique. In terms of the PM$_{10}$ rose, the year 1996 was simply worse in terms of its easterly quadrant contribution. In other wind directions it does not seem so unusual.

Summarising, in view of the number of exceedences, 1996 is an important year to investigate. The underlying meteorological and physical processes that govern the incidence and strength of easterly flows require further elucidation. Data on fine particles from other sites and years should be studied, and local versus distant causes/sources differentiated.

3.3.5 Main Features for 1996

In these rose diagrams, Figures 12-27, 1996 stands out in some respects, whilst in many ways all years have some marked similarities. They all show the prevailing winds from SW and NE, with a bias to higher pollutant levels in the easterly quadrant of 50-140°. This easterly quadrant shows the greatest year-to-year variability in speed, temperature and PM$_{10}$ burden. The year 1996 shows the following features:

1. Highest PM$_{10}$ concentrations in the easterly quadrant.
2. Coldest temperatures in the easterly quadrant.
3. Highest wind speeds in the easterly quadrant.

These rose plots do not confirm whether these features are causally associated or not. However, they do merit some consideration, in that they may be associated with outbreaks of polar continental air masses into the UK. Such air masses may be regarded as having some characteristic properties, which, in view of the need to identify features associated with secondary PM$_{10}$ episodes, merits discussion. It is also necessary to explore the conditions under which the less frequent, but more polluted, easterly and north easterly flows can occur.

3.3.6 Pollutant Flux Rose Plot

We now introduce the use of a pollutant flux rose plot, in order to explore the directional dependence of pollutant advection towards the monitor. The flux, *F,* is the mass of pollutant borne towards the monitoring site by the wind, measured as the rate of mass passing through a unit area in the vertical plane and aligned normal to the wind. It is defined in Equation 1:

$$F = u \times C \qquad (1)$$

where the wind speed, *u,* is assumed to be uniform and representative of the mean flow, and C is the mean concentration of pollutant. The flux, *F,* of PM_{10} has units µg m^{-2} s^{-1}, representing the product of London Bloomsbury concentration, *C,* times the London Heathrow Airport wind speed, *u,* measured at 10 m above ground.

Pollutant fluxes can only be calculated for periods when the wind and pollutant data are both available. We have used hourly observations from 1992-1998. We illustrate the method using these data from all available days, then we will sort the data to select the most polluted (exceedence) days for analysis.

3.3.6.1 All days, 1992-8

Figure 27 was discussed in Section 3.3.4. It shows the easterly bias in the PM_{10} observations at London Bloomsbury. Note that PM_{10} concentrations in 1992 and 1996 approach 50 µg m^{-3}, albeit in slightly different eastern directions (60-80° and 100-110°, respectively). Figure 28 shows the frequency of each wind direction in all years; 1992-1998 had similar prevailing winds; SW being most frequent. Figure 29 shows the average wind speed from each wind direction; 1992-1998 had broadly similar wind speeds when averaged by direction, but 1996 had stronger winds from 60-80°. Figure 30 shows the cooler easterly and north easterly flows in 1996. Figure 31 shows the pollutant flux plot for PM_{10}. Here 1996 stands out quite dramatically, with a large flux component from wind directions 60-80°. This plot also shows that from 1992-1998, the flux in 1998 was the smallest. Notice also that 1994 had a somewhat elevated flux, especially from 80°, although this only reached 200 µg m^{-2} s^{-1} instead of 275 µg m^{-2} s^{-1} seen in 1996. To summarise, the flux rose plots for all available days in 1992-8 highlighted 1996 in easterly flows as the highest, with 1994 second highest, and 1998 the lowest.

3.3.6.2 Exceedence Days, 1992-8

Exceedence days were those where the 24 hour (daily) mean concentration (TEOM) of PM_{10} at London Bloomsbury was greater than 50 µg m^{-3}. The number of exceedence days varies greatly from year-to-year, for example, 1996 had 37 exceedence days whilst 1998 had only 3. Data from the entire 24 hours (00:00 – 23:00) of an exceedence day were used, sorted and plotted in the same way as before. This is really useful because it allows the analysis to focus on periods of particular concern. Figures 32-36 plot the data taken from exceedence days. Figure 32 has the PM_{10} rose, with relatively similar concentrations in all directions for all years. There is more scatter now, as the number of days in the sample is quite small compared to the full year discussed earlier. Generally, the concentrations in any direction or year were in the range 40-80 µg m^{-3}. Figure 33, the wind frequency rose, shows wide scatter due to the small number of days, e.g.1998 has just 3 exceedence days. Again, 1996 stand out with a large frequency (7.5-11.5%) of easterly winds from 50-80°, although 1994 also has around 6.0-7.5% of winds in the directions from 70-100°. Note that 1998 did not have easterly exceedence flows. Figure 34 shows the low wind speeds (0.5-2.0 m s^{-1}) from the North West, and the largest easterly wind speeds in 1996 (5.7-6.3 m s^{-1}), followed by 1994 (4.6-5.4 m s^{-1}), from the directions noted earlier for these two years. Figure 35 shows low temperatures in easterly and north easterly exceedence flows for 1996 (~2-5°C). The exceedence PM_{10} flux plot in Figure 36 is wholly consistent with the all days flux plot of Figure 31; both plots show the strong easterly contribution to the flux. Generally, the 1996 exceedence days tended to have hourly concentrations ≈ 40-80 μg m^{-3}, but the easterly flows (50-100°) had the larger fluxes, with low temperatures,≈ 5°C, moderate winds and ≈ 6 m s^{-1}

The exceedence days were then sorted into two groups: Group 1 which looks at episodes of a few days, and Group 2 which took the remaining isolated one day episodes. The idea was to see if prolonged episodes might have a different behaviour from individual days with exceedence. The Group 1 pollution rose was to test a hypothesis of a European, or more strictly any Easterly bias, in the days when prolonged exceedence occurred. Group 1 has 68 days and Group 2 has 77 days of exceedence in 1992-8.

3.3.6.3 Group 1: Prolonged Episodes

This group included all hourly observations from the following dates (inclusive):

1992: 23 Jan - 1 Feb; 8 - 11 Apr; 23 - 30 June.
1993: 10 - 15 Feb; 6 - 10 June; 1 - 5 Nov.
1994: 10 - 15 Oct.
1995: 1 - 6 May; 11 - 14 Oct
1996: 15 Jan - 2 Feb; 10 Mar - 26 Mar
1997: 9 - 12 Mar.
1998: 0 days.

Group 1 selects just those episodes that were clusters of exceedences with typically 4 or more days coming next to each other. Some discretion or subjective judgement was involved, but the sub set seemed, by their prolonged nature, most likely to have some European airflows. Confirmation of the mesoscale flow could come from a study of the meteorological charts, or trajectory modelling.

3.3.6.4 Group 2: Isolated Episodes

This includes all hourly observations from the remaining exceedence days that were left after Group 1 were extracted. These days were generally in exceedence but only on isolated days (<4) on their own. This group had the following dates:

1992: 25 Feb, 5 Mar, 1 Jun, 9-10 Jun, 29-31 Jul, 19 Aug, 17 Sep, 17 Dec, 30-31 Dec
1993: 1 Jan, 2 Feb, 5 Feb, 2 Mar, 15 Mar, 14-15 Apr, 29-30 Apr, 1 May, 11-12 May, 24 May, 2 Jul, 19 Aug, 16 Nov, 19 Nov, 24 Nov, 28 Nov
1994: 14 Feb, 18-19 Feb, 25 Feb, 11 May, 13 Jun, 24 Jun, 1-2 Jul, 12 Jul, 21 Jul, 24 Jul, 22 Nov, 25 Nov, 29 Nov, 1 Dec, 9 Dec, 23 Dec.
1995: 3 Jan, 14 Mar, 25 Apr, 23 May, 30 Jun, 25 Jul, 31 Jul, 1 Aug, 11-12 Aug, 15 Aug, 22-23 Aug, 31 Oct, 7 Nov, 12 Nov, 17 Dec.
1996: 7 Feb, 7 Apr, 10 Apr, 6-7 Jun, 26 Jun, 21 Jul, 15 Sep, 14 Nov, 5 Dec

1997: 11 Jan, 25 Jan, 9-10 Apr, 10-11 Aug, 19-20 Aug, 29 Sep, 2 Oct, 30 Oct, 2 Nov, 12 Nov
1998: 16 Mar, 13-14 May

Sorting the exceedence days from 1992-8 to select Group 1, those days which fell into prolonged episodes (≥ 4 days adjacent or only spaced by a day's interlude) then revealed that:

1. There were fairly constant concentrations of PM_{10} for all wind directions during these prolonged episodes (a somewhat unexpected result since an Easterly bias had been expected). In Figure 37, the concentrations were between 50 and 70 $\mu g\ m^{-3}$ in all wind directions.
2. Prolonged polluted periods for many directions had light winds ($< 2\ m\ s^{-1}$) associated with them, Figure 38, but for the more easterly flows there were moderate winds, ranging from 2 to 5 $m\ s^{-1}$ at winds from 60°-80°. Figure 38 also shows that for the prolonged episodes, north easterly flows were much more frequent, and had much stronger wind speeds reaching 5 $m\ s^{-1}$ in 60-80°. In fact about one third (34%) of these prolonged exceedence hourly wind directions were from directions 40-90°. Such directions encompass the areas north of the Thames, as well as flows from further a field on the continent. In short, during these prolonged episodes, the winds were more commonly from an easterly direction and were stronger, but the pollutant concentrations were not in themselves higher in any particular direction for prolonged exceedences.
3. Figure 39 emphasises clearly that the temperatures were lower in the east to north eastern prolonged exceedence flows.
4. If these prolonged episode days have the same particulate concentrations in the stronger easterly winds as the light winds from say the west, it suggests that the pollutant flux should be calculated[1].

When such a pollutant flux rose plot was drawn for the exceedence days, Figure 40, a very pronounced directional bias was seen. There was much larger flux from the North East Thames direction (60-90°) than from any other direction. It strongly suggests important PM_{10} sources could be just North of East (around 70-80°), perhaps large sources within the London conurbation. There are a number of large pollutant sources in this direction. This was an unexpected finding; a continental contribution was anticipated, even presumed, but the flux suggests a quite strongly focussed directional contribution. Further study of this finding is clearly needed; it should be regarded as tentative at this stage. Our analyses relied upon London Bloomsbury data in the centre of London, with an abundance of sources around the monitor.

To be sure that the pollutant flux rose diagram is a useful analysis tool that leads to sound conclusions, study of other sites is needed, especially to include some rural data. It would also be very interesting to use the London emission inventory data in

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[1] Other things being equal, stronger winds dilute source emissions more. Our flux plots assume inverse proportionality between surface wind speed and observed concentration, which may not always be true. The dependence of concentrations on wind speed will also vary with long-range transport, as wind varies in space and time. Formation and removal processes en route also influence concentrations.

a modelling study to see whether known sources within the conurbation are sufficient to explain such a bias in the flux of PM_{10}.

3.4 Pollutant Flux Rose Plots for Port Talbot

It was agreed with the Department that before firm conclusions are drawn using pollutant flux rose plots, other sites should be examined. In the area near the Port Talbot steel works, the local authority of Neath and Port Talbot has declared an Air Quality Management Area based upon PM_{10}. Attention has focussed upon the steel works, although the roles of other sources, such as roads or European imported pollutants, have also received some attention. Martin Hooper from Neath Port Talbot local authority very kindly provided a PM_{10} flux rose plot (not shown). The pollutant rose plot suggested a WSW direction for elevated PM_{10} concentrations at a monitor to the north of the works. Later work at the south side, showed a similar effect with a bias from directions over the works. The authority has rose diagrams showing the distribution of wind directions (wind frequency), the wind speed by direction, the PM_{10} concentration, and the distribution of PM_{10} flux by direction.

The flux rose plot at the Port Talbot AUN site has a large peak from SW and WSW. This direction is broadly from over the steel works. Interestingly there is also a lesser flux from S to SSE and a minor contribution from E. There may be significant local sources in each of these upwind directions. We are still evaluating the pollution flux rose plot as a tool for analysis of air quality data, having recently been drawing these for London Bloomsbury and ended up wondering about possible industrial sources on the North East Thames. In the context of the present study, it is noticeable that moving to a quite different location generated a very different prevailing direction for the largest flux. In Port Talbot at least, this was apparently consistent with the position of a recognised local source. There is a clear need for a more detailed study of pollutant flux rose plots for other pollutants and sites, including rural locations.

4. DEVELOPMENT OF INDICATIVE INDICES

Where climatic data are sought, long runs of data are needed if slow, perhaps irregular, processes are to be resolved e.g. tree rings have been used as a surrogate for temperature. Many pollutants have been recorded for too short a time-scale for robust climatic analysis to be carried out. Here we attempt to develop simple indices that may associate with pollution episodes; the climatology of these indices, rather than the pollutant data, is more amenable to study if they are derivable solely from archived meteorological information.

4.1 Winter Index, *WI* (or Temperature-Wind Index, *Tu*)

The box model (Equation 1 in Middleton, 1998) uses conservation of mass of pollutant to calculate a city average concentration from sources near the ground. The concentration, C, is proportional to the emission rate, q, and inversely proportional to the ventilation wind speed, u, and the vertical extent of mixing or height of the box, h:

$$C \propto \frac{q}{hu} \tag{2}$$

In cold winter episodes the emission rate, *q,* for space heating increases as it gets colder (Middleton, 1998), and here it will assumed to be proportional to the degree days:

$$q \propto \left(T_{ref} - T_{mean}\right) \tag{3}$$

where T_{ref} is a reference temperature (18°C was used here) and T_{mean} is the daily mean temperature (°C) calculated from the hourly observations. Combining the rise of emissions in cold weather with the inverse dependence upon wind speed (adding 0.5 m s^{-1} to avoid overflow in division with calm winds), and ignoring changes in the mixing height (*h* is for all practical purposes small, perhaps 100-200 m, in stable conditions during winter episodes), we have a simple weather related winter index, WI:

$$WI = Tu = \frac{T_{ref} - T_{mean}}{u + 0.5} \tag{4}$$

This will be largest in the lightest winds and coldest conditions. It isolates two very important factors in primary pollutant episodes, whilst avoiding the uncertainties of diagnosing the stability for estimating *h*. In this work, it was not feasible to include the emissions data, because the focus is on spanning as many years as possible. The index *Tu* has been calculated for every day of observations in the synoptic record. *Tu* showed large peaks that mostly coincided with the dates of well known severe episodes (Table 1, seen earlier) in London. Such an index is dependent only on meteorology and has the attraction in this study that it can also be quickly calculated from climate model output. Assuming an unchanged emission scenario, the frequency of large values of *Tu* in the climate predictions could be a pointer to likely future winter episodes. The winter index does not require wind direction; the primary pollutants to which it may relate are assumed to be widely distributed around the receptor. (*Tu* is operating like a box model, but without any dependence upon box depth.)

Figures 41-44 examine the behaviour of *Tu* in the four years of especial relevance to this study. In December 1952, London had its worst measured smoke and sulphur dioxide episode. This was a primary pollutant episode driven by low temperatures, calm conditions, and freezing fog with very strong inversion. *Tu* reaches a value of 42 at this time (Figure 41). In December 1991, London had its worst measured nitrogen oxide episode. Again, this was a primary pollutant episode in very similar conditions of suppressed dispersion and low temperatures. In Figure 42 *Tu* reached 23, coinciding with a maximum in the daily average of black smoke that attained 76 µg m^{-3}. Since *Tu* is calculated solely from the meteorological record (i.e. temperature and wind speed), these values of 42 in 1952 and 23 in 1991 suggest that for dispersion of local emissions, 1952 was perhaps nearly twice as severe as in 1991. A direct pollutant based comparison of their severity would require the same substance to have been collected and analysed in the same manner, with the same precision, over a 40 year period. *Tu* helps avoid this challenge.

Figures 43 and 44 show the values of *Tu*, and daily average black smoke at Westminster. In both 1996 and 1998, *Tu* did not exceed 13. Clearly these years did not have conditions conducive to severe winter primary pollutant episodes. 1996 is notable for its PM_{10} episodes, whilst 1998 is regarded as a low pollution year (Section 1.2). These results for *Tu* being below 13 are consistent with the idea that the 1996 PM_{10} episodes were not of a winter primary nature. In terms of *Tu*, 1996 and 1998 do not appear very different.

The temperature-wind index *Tu* shows that for avoiding severe winter pollution episodes in the future, it might be desirable to have emission policies in place that can address (and suppress) any increase in emission rates which may occur in response to space heating demand. Likewise policies that can suppress any tendency for motor vehicles to have increased emissions in cold weather (cold starts, and running in slow moving city traffic with catalysts below light-up temperatures) would also appear desirable. It appears that light winds alone are insufficient to cause these severest episodes, and so the index *Tu* focuses attention on the role of low temperature in conjunction with calms, and how emission sources respond to the coldest weather. With regard to vehicle emissions, engine and catalyst design can be optimised to minimise the impact of cold weather on catalyst performance.

4.2 Summer Index, *SI*, for Ozone Episodes

When comparing *Tu* with observations, we noticed that it takes larger negative values in hot still conditions that are conducive to ozone formation. Low wind and elevated temperature with trajectories of air from mainland Europe are strong indicators of elevated summer ozone. To some extent *Tu* indicates ozone episodes, though less directly than the primary winter episodes for which it was intended. Daily mean temperatures T_{mean} >18.0 cause $Tu < 0.0$. In discussion afterwards, Derwent (personal communication) pointed out that daily maximum temperatures in excess of 25°C are an indicator for elevated ozone concentrations. On this basis an ozone index might be defined as a summer index, *SI*

$$SI = \frac{T_{\max} - T_{ref}}{u + 0.5} \qquad (5)$$

where T_{max} is the daily maximum temperature, T_{ref} = 25°C, and u is the wind speed as before. This has not been tested, not least because it probably requires some sorting on the wind direction for continental air as a supplementary condition. (Notice that the signs in the two indices are reversed; in winter it is coldness that matters through increased emissions; in summer, it is warmth that elevates evaporation of precursors and accelerates the reaction rates).

Finally, since the temperatures T_{mean} and T_{max} are very closely related, both *SI* and *WI* are correlated. Consequently we only discuss the values of *Tu* here.

4.3 Results for *Tu* from Climate Model Data

The Met Office climate model run in the Hadley Centre was used to supply daily values of the minimum temperature, maximum temperature, and wind speed. Daily values of the winter index, *WI* (i.e. *Tu*) were calculated using daily mean climate modelled temperature (mean of minimum and maximum). These have been plotted as time series of 360 days (the climate model year) from 1990 through to 2020.

Figure 45 shows for 1991 how *Tu* behaves when calculated from 360 day climate model values for temperature and wind-speed. Poor correspondence was found between *Tu* from observations at London Heathrow and the climate model results, as in Figure 46. This may be attributable to several features of climate simulations, the time for the model to 'spin up' or initialise, the effects of area averaging in large model grid squares, the effects of temporal averaging, the fact that only daily maximum and minimum temperatures were used here (and averaged for daily 'mean'), and the year modelling as 360 days (whilst observations have 365). Such factors need careful investigation. The results for *Tu* in 2010 and 2020 are shown in Figures 47 and 48. Considerably more work is needed before climate runs can be used as safe predictors of the future pollution climate. Work in Section 3 reveals the importance of relatively short-term meteorological events, such as synoptic conditions leading to episodes that last for a few days, in determining the overall pollution climate of a year.

5. VISIBILITY AND PARTICLES

Visibility is a measure of the distance through the atmosphere that distant objects may be seen clearly. It decreases as the loading of droplets and particles in the air increases. Consequently a link between visibility and particle concentrations merits study; visibility is a meteorological variable which has some dependence upon pollution and which has been recorded over many years. This Section explores the idea of linking visibility and particle/aerosol loading in an inverse fashion.

5.1 A Visibility Mass Equivalent?

Visibility in the atmosphere decreases both as humidity increases, and as more droplets form on condensation nuclei; it is therefore related to both humidity and particulate concentrations. A program, PABLY, can be used to retrieve hourly observations, and since it contained the code needed to extract visibility data it has been copied and modified. The new program is called PABLYVIS because it retrieves and processes visibility. In principle, it seems that when searching for a sort-key that can be used to scan the last 50 years of synoptic meteorology for conditions associated with secondary pollutants, the visibility should be a prime candidate. PABLYVIS has been set up in this study to also calculate a nominal 'Visibility Mass Equivalent', or VME. The author defines the visibility mass equivalent to be that concentration of particles, in µg m^{-3}, which is inversely proportional to the visibility.

This was arrived at after reflecting on the idea that as the particle loading increases, the visibility can be postulated to decrease. The behaviour is similar to the Beer Lambert law, which governs the absorption of light in spectroscopy. A subsequent search of the literature based on this argument led to the following important ideas:

Charlson et al. (1968) demonstrated the link between visibility, extinction coefficient (light absorption and scattering), and particulate mass concentration (NB not the same as PM_{10}). For the observed visibility, L_v^* (which is a distance expressed in metres to the most distant easily distinguished object), and the mass concentration, C, in µg m^{-3} they found:

$$L_v^* \times C = K \tag{6}$$

where K is an empirical constant with most probable value 1.2 g m^{-2} (this being the modal value of their observed data), and ninety percent of results lying between values for K of 0.7 and 2.6 g m^{-2}. Note that in converting from g to µg a factor of 10^6 is required.

Noll et al. (1968) also found a quantitative inverse relation between visibility and mass concentration of particulate matter. Their result can be expressed in the same formula, but their Table 4 has a range of values for the empirical constant K which were 1.9, 1.6, 1.0, and 1.2 g m^{-2}. Their recommended value for K was 1.4 g m^{-2}, which combined all four values.

Using this equation and values for K, PABLYVIS is now coded to tabulate the range of values for VME alongside the hourly wind data, temperatures and relative humidity. A few data were run through the program for March 1996; unfortunately they seem less encouraging than first supposed. Frequent high humidity may become a significant constraint on the usefulness of the method. The results are tentative at this stage.

A secondary episode sort key cannot use just the visibility or VME however. We know this because winter smog episodes have often a very small visibility, which from the above implies a very high mass concentration. However these episodes also have up to 100% relative humidity, and may have temperatures at or below freezing. Consequently VME alone would not be linked unambiguously to secondary particulate. The role of relative humidity must be stressed; in the work of Charlson et al. (1968) and of Noll et al. (1968), the authors stress the importance of selecting aerosol concentrations when the humidity was not saturated i.e. up to perhaps 60% or 70% and no more. Consequently their data from American sites may differ from data at UK stations; in the UK the humidity may turn out to be too high on many occasions for this analysis to be of merit. However now that the program PABLYVIS can generate the required data it is possible now to compare temperature, humidity, wind, visibility, stability, boundary layer depths and VME data with the recent record of fine particulate concentrations.

Our use of visibility (or VME) here is in its infancy; for example we used the site at London Heathrow Airport (because we were set up to use it already) which has a long archive. However it may be better to combine visibility data from several more remote sites, if these are less sensitive to local emissions (traffic and aircraft particulate emissions may perchance affect visibility at Heathrow Airport). Such an average might reflect secondary episodes from remote sources. Several sites might be averaged, or differences between VME at different sites evaluated. In any case, humidity as a complicating factor as well as local emissions needs consideration.

5.2 Other Relationships for PM_{10}, RH and Visibility

There are well established relations which relate visibility to particle concentrations as PM_{10} and inversely to relative humidity (Helen ApSimon, personal communication) but it was not possible within the scope of the present Contract to investigate this further. This is felt to merit further work, as it could offer a route to using visibility data recorded over many decades. The data might be useable for inferring some particle loading when RH is allowed for. There is scope here for work jointly with Imperial College where these relationships have been studied previously. It would be fairly straightforward to program their relationships into PABLYVIS. The program could be run on many years' data (much longer than existing PM_{10} records could permit).

This is an area for a fuller study. It requires additional effort to apply current knowledge of RH, particulate concentration or PM_{10}, and visibility. It would make an interesting and possibly informative study. It has application in analysing historical records of observed data.

6. SYNOPTIC SCALE PROCESSES

A proper understanding of the meteorology associated with episodes of secondary pollutants must address processes over a very wide area. Synoptic processes include the movement of fronts, cyclones and anticyclones; they control the presence of strong westerly flow, or the presence of blocking systems with a tendency for easterly flow. This topic will now be discussed and its implications for the present study explored.

6.1 Easterly flow and Polar continental air masses

In seeking to explain the occurrence of episodes, especially of fine particles which depend upon remote as well as local sources, and in order to say something of their climatology and likely frequency of recurrence, it is desirable to seek some physical understanding of the physical processes which influence the movement of bodies of air to and from the continent. Plumes of reacting gaseous pollutants can be brought to these shores according to the synoptic conditions. Easterly flow for example brings continental pollutants to Britain. The behaviour of plumes, such as their chemical reactions, vertical dispersion, and deposition, will be sensitive to the meteorological conditions in the air masses that are bearing the admixture along. The manifestation or otherwise of pollutants at a local receptor or monitor may be influenced by meteorological processes which occurred far up stream of the release point, as well as during the trajectory or passage of the pollutants. The physical causes and frequencies of these conditions set some upper bound to the likely frequency of imported secondary particulates. Certain air masses may be associated with episodes, whilst others may be associated with much cleaner air, as we now investigate.

In 1928, Bergeron first proposed the idea of air masses, as widespread bodies of air with relatively uniform temperature and humidity in the horizontal plane (temperature and humidity vary in the vertical due to diurnal cycles driven by solar radiation).

Petterssen (1940) has summarised the concept which defines what is meant by an 'air mass':

> 'An air mass is defined as a huge body of air whose conservative properties, notably temperature and humidity or functions thereof, are more or less homogeneous in the horizontal direction. The horizontal and vertical dimensions of an air mass are normally of the same order of magnitude as those of the large systems of circulation, such as anticyclones and depressions. The various portions of an air mass must be of common "origin", and the "development" or "life history", of the various portions must be essentially the same. For example, a vast mass of air remaining over the snow-covered arctic regions for a sufficient length of time will acquire physical properties characteristic of that region. When such a mass moves towards warmer regions, it will change its properties and develop weather phenomena that are determined by the arctic origin of the mass and its movement from a cold toward a warm region. When classified according to origin and development, the air masses often appear as distinct types, and the recognition of this fact is of substantial assistance in the forecasting of weather.'

In his classic text, Lamb (1964) analysed the English climate in terms of weather types. The weather here is very sensitive to small changes in where the air has come from. We argue that this has significance too for the pollution climate, especially secondary pollutants. We now summarise the points from Lamb (1964) that seem directly relevant to the present investigation:

1. *Easterly type*: With anticyclones over Scandinavia, and perhaps a ridge of the high pressure extending out to Iceland or Northern Britain, flow from continental Europe can bring cold weather in autumn, winter or spring; and warm, possibly thundery weather in summer. This at once conveys the idea of continental pollutants moving along a ridge of high pressure from a region over Germany or Russia, since these could be upwind source regions in such conditions.

2. *Continental Polar*: In this weather type, cold or very cold winter or warm summer may occur. The atmosphere may be unstable, especially in the lower layers. This suggests a possibility of elevated plumes being mixed down to ground level

(fumigation). In continental polar weather, the cloud cover depends on the trajectory followed by the air mass. Air from the Baltic area or North German plain is likely to be cloudier than if from over the hills or mountains further south. Such topography can cause the air to ascend and lose moisture. With the northerly air it is commonly overcast with stratocumulus. In summer all districts experience a good deal of warm conditions with clear skies. The continental polar air is however often somewhat hazy, Lamb (1964). This no doubt reflects the presence of aerosols formed from precursor pollutants released over the continent. From Figure 12 in Lamb (1964) a typical route for continental polar type air masses is across Russia and Germany.

According to Lamb (1964), a definable weather type can be defined 'as something which often lasts for some days, whilst the weather undergoes variations typical of the succession of air masses and depression tracks etc. occurring with that prevailing wind direction and type of weather sequence or spell' (ibid: p. 54). This concept implies that the prevailing wind direction over a period of days is the one closest to the main flow of the upper winds and is related to the steering of cyclones and anticyclones. The weather type may be described as cyclonic or anticyclonic, combined with one of the direction types W or SW; NW; N; E; S. As discussed above the pollution rose revealed elevated particulate concentrations in the easterly quadrant. With weather types lasting some days, perhaps with variations within that period, we note this is quite consistent with the PM_{10} measurements for March 1996. The envelope of PM_{10} concentrations during those episodes are seen to be raised over periods of a few days, with some drops to lower values during these polluted weather types. The skill of air mass classification as a predictive tool for secondary PM_{10} will inevitably be much less than either a calculated trajectory or modelling by NAME for several reasons:

1. Air mass type indicates a broad scale classification of the situation; it cannot say in any detail where individual plumes may travel.
2. The air mass classification ignores the detailed spatial distribution of emission sources.
3. Other processes such as chemistry, deposition, and the emission of primary PM_{10} near the receptor site, are ignored.

The cyclonic type has ascending unstable air, wet and windy weather. The anticyclonic type has descending air that causes a subsidence inversion that warms the lower layers. This may trap pollutants by limiting the depth of unstable air. It can also suppress cloud formation. The state of sky depends upon the height of the subsidence inversion; if low enough sky remains clear, with haze below due to accumulated pollutants; if the air below the inversion becomes saturated then cloud is formed, causing a persistent sheet of stratus or stratocumulus. Combined with the haze, and reduced light in winter, this can make for very gloomy days. It is interesting that during March 1996, when secondary particulate pollution was judged high, anecdotal reports suggest such cloud was indeed present. Data from ascents could reveal if this month had frequent inversions suppressing vertical mixing in the continental air.

6.2 Frequency of Easterly Air Masses

The frequency with which air masses from continental Europe enter the British Isles may shed some light on how likely are the conditions of March 1996 to recur. Assuming that the elevated concentrations in 1996 can be associated with flows from our easterly quadrant, 50-140°, we look at some classical studies reported by Belasco (1952). He analysed upper air data to study the physical properties and occurrence of tropical air masses, polar air masses, and anticyclonic air masses of 'indeterminate' trajectory (being in the central region of the anticyclone). Of the 4 main weather types he identified 21 subdivisions. From Figures 1-3 in Belasco (1952) we selected the ones that seem likely to bring polluted air into the country, as follows:

1. Tropical continental air mass, subdivision T_3 and T_4.
2. Polar continental, subdivision A_1 cyclonic and A_2 anticyclonic, especially with NE, E and SE flows.
3. Anticyclonic central region with indeterminate flows, subdivisions H_{NE} and H_{SE}. Of these H_{SE} seems more likely to bring polluted air from southern Europe, whereas H_{NE} skirts the Netherlands coast and spans the North Sea passage.

Belasco analysed the incidence of these air masses and their subdivisions, based upon data recorded daily at 1800 hours at the old Kew Observatory in London. Table 4 shows the frequencies of each relevant type T_3+T_4; A_1+A_2; and H_{SE} (we neglect H_{NE}). Belasco also reported the number of years in which the subdivision of weather type was not recorded during each month; this is useful for it conveys some sense of the fact that several years may pass in which a particular weather type may not occur. Note that the frequency data in Belasco's (1952) full table add up to 100% in any one month if all 21 weather type subdivisions are considered.

The table by Belasco (1952) was compiled using the following physical variables:

1. trajectories from geostrophic wind derived from isobars on surface synoptic charts.
2. Screen temperature and humidity at Kew
3. Pressure, temperature and humidity aloft via aeroplane and radio-sonde.

In the case of just polar continental air masses, as seemed important in March 1996, the frequency is rather small, ~ 1-7% in winter (up to say ~2 days/month), and a negligible frequency in summer. In any month, the combined frequency of receiving air masses from continental Europe is therefore from the Table of the order ~ 10-15% (say ~ 3-4 days/month). These data merely reflect the observed physical properties and trajectories of the air masses studied by Belasco (1952) and do not indicate the magnitude of any elevations in pollutant concentrations that might be associated with them. For this it is necessary to analyse data taken when pollutant monitoring in the AUN network was operating. The extract from Belasco's (1952) results contained the number of years (within the 12 year study period) when no subdivision of a given weather type was observed. For example, Polar Continental types A_1+A_2 were never observed in the months April-November inclusive. In the months when they were seen, this may only have been in 3 or 4 out of the 12 years

Table 4 Frequencies of relevant air mass types arriving at Kew from 12 years' observations, 1938-1949, arranged by month and type, using Belasco, 1952.

Month	Tropical continental	Polar continental	Anticyclonic indeterminate	Total Frequency
	T_3+T_4 (%)	A_1+A_2 (%)	H_{SE} (%)	(%)
January	3	7	5	15
February	4	6	6	16
March	5	1	7	12
April	5	0	7	12
May	6	0	5	11
June	3	0	5	8
July	4	0	8	12
August	5	0	6	11
September	3	0	9	12
October	9	0	7	16
November	7	0	7	14
December	2	3	6	11

available. Such events do not recur every year; rather they are somewhat infrequent and a number of years may pass in which they do not happen. The anticyclonic indeterminate weather type with a SE trajectory appeared in every month, but is still only with a frequency of occurrence 5-9%. Note that if an Anticyclone is over the British Isles, winds may be light and inversion height small, with conditions conducive to elevated local or primary PM_{10} pollution, and this can add to the effects of European air masses bearing secondary particulates.

6.3 Mechanism of Easterly Flows: The North Atlantic Oscillation

The prevailing wind tends to be from the west, as shown by the wind roses. For European derived secondary episodes, the pollution roses suggested that atmospheric flows from the eastern side of the country are necessary. The flows associated with this behaviour require high pressure over Scandinavia, the Baltic or Russia. This is because in the Northern Hemisphere, the flow is clockwise around high pressure and this drives the air across the German plain and France towards Britain. These flows may be coupled to longer-term quasi-oscillatory behaviours in the atmosphere, of which the North Atlantic Oscillation is a leading consideration. Two articles from New Scientist gave an introduction to this Oscillation, see Morton (1998) and Walker (2000). They show the possible importance of coupled ocean-atmosphere processes.

Barry and Chorley (1998) describe how sea surface temperatures may influence climate in Europe, Africa and South America.

> The "North Atlantic Oscillation (NAO) also shows strong air-sea interactions. The NAO is an oscillation in the pressure field between Iceland (65 °N) and a zone about 40 °N across the Atlantic. Fluctuations in the NAO give rise to alternating mild/severe winters in western Greenland-Labrador and north-west Europe. Severe winters in the Greenland area also have cold northerly airflow and more extensive sea ice in the Labrador Sea. On a longer time

> scale, the NAO index (of S-N pressure difference) was generally low from 1925 to 1970, when air temperatures in the northern hemisphere were above normal and cyclones over the east coast of North America tended to be located over the ocean, thus causing longer, drier east coast summers. Prior to 1925, a regime of colder climatic conditions was associated with a higher NAO index. Since 1989, the NAO has been consistently positive."

The difference in pressure between the Azores and Iceland has been linked to the westward or eastward strength of the flow. Indices of oscillating flow were discovered early in the twentieth century: they include the Southern Oscillation SO (atmospheric component of El Nino, a sea current in the Pacific), and the NAO.

In Britain a large NAO index is associated with strong winds from the west, relative warmth in northern Europe, mild winters, and storms. A small or negative NAO index is associated with cold winters, weak westerlies (or even easterlies) and a continental climate in Britain dominated by cold air from the north and east. Monthly average values of the NAO index were provided by Mark Rodwell of the Hadley Centre (personal communication). Values are the pressure at Ponta Delgada in Azores minus pressure at Stykkisholmur in Iceland. Differences are measured in multiples of 0.1 hPa (missing data -999). Values from January 1866 to March 2000 inclusive were kindly supplied.

Table 5a shows a 10 year sample of the NAO values in multiples of 0.1 hPa (i.e. 0.1 mbar). Large values, such as 458 in January 1990 may associate with strong flow from the west (westerly) whilst small values, like -38 in March 1996 when polluted air came from Europe, may associate with flow from the east (easterly). Fine particulate monitoring data, especially for PM_{10}, do not go far enough back in time for them to provide us with a reliable measure of the frequency of secondary particulate episodes to be diagnosed (Table 5b). This is the reason why other less direct indices are studied in the current project.

The data also show that:

1. In 1996 NAO fell below zero in both March and December; the March PM_{10} was generally elevated, with some interludes in which it decreased (Malcolm et al., 2000) consistent with a sustained small or negative average NAO for the month.

2. In 1998 however the NAO tended to be noticeably larger, flow from the continent seems less likely, and PM_{10} values were not high.
3. Finally we observe there were negative means for NAO in both October and November 1995; but PM_{10} was not noticeably elevated in those months.

The NAO values here are monthly means; they may give a broad measure of tendency in the flow, but are clearly insufficient to predict PM_{10} concentrations in the UK. Whether daily values of NAO could serve as predictors of daily exceedences remains to be seen. It is usual when analysing climate records for quasi-oscillatory behaviours, patterns, or links to other processes, to first process the data. For example running means may be applied, or high pass filtering can be followed by spectral analysis, as in Currie et al. (1993).

Table 5a Values of NAO pressure difference (monthly mean NAO index: Pressure at Ponta Delgada, Azores minus Pressure at Stykkisholmur, Iceland). The units are 0.1 hPa. Data from Mark Rodwell, the Hadley Centre, Met Office.

Date	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Jan	458	273	135	397	343	325	102	45	166	318	215	200
Feb	460	202	362	171	147	364	225	462	242	326	417	112
Mar	288	127	307	225	433	252	**-38**	218	156	147	156	
Apr	252	157	193	175	157	12	75	33	123	99	**-18.2**	
May	2	90	106	**-40**	67	55	17	10	71	118	142	
Jun	157	97	117	117	209	42	149	35	75	172	103	
Jul	160	127	171	138	192	130	192	171	166	130	888	
Aug	175	212	221	137	114	120	158	170	203	75	149	
Sep	142	147	152	110	140	144	140	139	82	223	164	
Oct	192	128	80	**-21**	107	164	245	45	205	170	308	
Nov	95	263	318	315	195	**-27**	189	176	231	210	147	
Dec	282	203	290	369	322	**-21**	**-1**	188	335	297	67	
Ave	221	168	204	174	202	130	121	141	171	190	228	

Table 5b Number of daily exceedences of the particulate matter Air Quality Standard of 50 μg m^{-3} in each month and annual average concentrations (μg m^{-3}) at London Bloomsbury, UK.

Month	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Jan			4	1	0	1	10	2	0	0	0	0
Feb			2	7	4	0	3	0	0	0	0	1
Mar			1	2	0	1	12	4	1	0	0	0
Apr			4	4	0	1	2	2	0	1	0	0
May			0	4	1	7	0	0	2	1	0	0
Jun			10	4	2	1	3	0	0	0	0	1
Jul			3	1	5	2	1	0	0	0	0	0
Aug			1	1	0	6	3	4	0	1	0	1
Sep			1	0	0	0	1	1	0	1	0	0
Oct			0	0	4	5	0	2	0	0	0	1
Nov			0	8	3	2	1	2	0	0	0	
Dec			3	0	3	1	1	0	0	0	0	
Total			**29**	**32**	**22**	**27**	**37**	**17**	**3**	**4**	**0**	**4**
Ave μg m^{-3}			39	37	35	37	39	35	30	28	21	

6.4 Log of Lamb Weather Types

Duty forecasters within the Met Office are required to keep a log of the occurrence of Lamb weather types. These data are collated and stored for use in verifying the quality of the forecasting process. Data has been obtained from Rob Darvell (personal communication) who processes the forecaster log. His files contained Lamb weather types produced every 12 hours for the 5 years 1995-1999 inclusive. Every midnight and midday a symbol is stored as an alphanumeric code. There are

29 distinct codes. The 8 principal directions each have 3 codes, for example easterly has D, E, and F codes, namely Anticyclonic Easterly, Easterly, and Cyclonic Easterly respectively. The latter may coincide with primary episodes.

6.5 Frequency of Easterly Air Masses, (2000)

This Log of Lamb weather types spans 5 years, and the NAME runs span 3 years. Before attempting any statistical analysis we examine the data for one month. Table 6 compares the Lamb weather types that were reported by the forecaster log in the Met Office every 12 hours at midnight and midday for each day of March 1996. Daily mean concentrations (µg m^{-3}) of sulphate aerosol from the NAME model for the same month are also given. (Alison Malcolm, personal communication). At start of each monthly run the model takes a few days to import any European contribution, so UK starts as 100% contributor, and Europe 0% contributor, to the total modelled sulphate aerosol. These are preliminary results, because the distribution of sources given to NAME was simplified; large point sources are assigned to the 50 km EMEP grid squares in which they fall. The Table shows a tendency during this period for Easterly and South Easterly types of flow to be associated with the raised concentrations of modelled sulphate aerosol. However, according to NAME, the results in Easterlies and South Easterlies also indicate that the elevation of aerosol concentration is associated with a rise in contributions from both UK and European sources. It is not just caused by aerosol of European origin. This is an important point, and seems consistent with the pollutant rose plots shown elsewhere in this report where these directions have the higher PM_{10} concentrations and fluxes (see Section). We caution the reader that the NAME results in this Table were for sulphate aerosol produced by sulphur dioxide emitted from sources assigned to a 50 km grid for just one month, and a much more detailed study is advisable.

Table 6 Lamb Types from the Met Office forecaster log, collated by from Rob Darvell, (personal communication) for March 1996, along with calculated sulphate aerosol using NAME model from Alison Malcolm (personal communication). She did suggest there may be some undercalculation by the present model setup, but this is sensitive to model settings such as the size of the grid squares over which the modelled Lagrangian 'particles' are assigned and counted for determining concentrations. Note also that sulphate aerosol concentrations are generally smaller in magnitude than total PM_{10} concentrations.

Date	Lamb code* 00Z 12Z	Flow from	NAME UK SO_4 µg m^{-3}	NAME EU SO_4 µg m^{-3}	Total SO_4 µg m^{-3}	UK %	EU %
01/03/96	B B	NE	0.59	0.00	0.59	100	0
02/03/96	A A	NE	2.93	0.00	2.93	100	0
03/03/96	M G	N, NE	2.66	0.00	2.66	100	0
04/03/96	G M	NE, N	2.14	0.00	2.14	100	0
05/03/96	M M	N	0.73	0.00	0.73	100	0
06/03/96	A Y	NE, Ac	0.90	0.00	0.90	100	0
07/03/96	Y D	E, Ac	1.26	1.02	2.28	55.16	44.84
08/03/96	J K	SE	0.78	1.26	2.04	38.28	61.72
09/03/96	K K	SE	1.16	0.84	2.00	58.09	41.91
10/03/96	J J	SE	1.43	0.69	2.12	67.50	32.50
11/03/96	P P	SW	2.05	0.45	2.50	81.93	18.07
12/03/96	Q S	SW, S	0.48	0.37	0.84	56.31	43.69
13/03/96	J J	SE	0.38	0.65	1.03	37.09	62.91
14/03/96	J J	SE	1.11	0.79	1.90	58.57	41.43
15/03/96	J J	SE	1.73	0.84	2.57	67.19	32.81
16/03/96	K Ua	SE, Ac	3.81	1.90	5.71	66.80	33.20
17/03/96	Uc L	SE, C	5.73	0.60	6.33	90.49	9.51
18/03/96	L L	SE	3.90	1.55	5.46	71.50	28.50
19/03/96	L K	SE	5.95	2.00	7.96	74.81	25.19
20/03/96	E E	E	6.34	3.41	9.75	65.05	34.95
21/03/96	F F	E	2.14	1.27	3.40	62.81	37.19
22/03/96	Z K	SE, C	7.87	0.43	8.29	94.86	5.14
23/03/96	L Uc	SE, C	7.29	0.38	7.67	95.13	4.87
24/03/96	L L	SE	4.55	0.24	4.79	95.07	4.93
25/03/96	L K	SE	7.70	3.14	10.83	71.04	28.96
26/03/96	D D	E	3.35	1.19	4.54	73.73	26.27
27/03/96	G M	NE, N	0.11	0.00	0.11	98.48	1.52
28/03/96	B C	NW	0.75	0.00	0.75	100	0
29/03/96	O N	N	0.54	0.00	0.54	100	0
30/03/96	Y Y	Ac	0.37	0.00	0.37	100	0
31/03/96	Y Y	Ac	1.11	0.00	1.11	100	0

*Code:
Northerly Types M, N, O; North Easterly Types: G, H, I; Easterly Types: D, E, F;
South Easterly Types: J, K, L; Southerly Types R, S, T; South Westerly Types P, Q, +
Westerly Types V, W, X; North Westerly Types A, B, C;
Cyclonic Z, U_C; Anticyclonic Y, U_A.

6.6 Plumes from the UK Crossing the North Sea

Lamb Weather Types and NAO provide us with a long historical archive that sheds light on the factors influencing the flow of continental air *into* the UK from the east. This air may bring pollutants to the UK, acting as a raised background onto which local UK emissions are then dispersed. They can of course be applied in the reverse sense, to see how the weather may bring pollutants *from* the UK to Europe. In the 1970s and early 1980s, a number of studies examined the long range transport of acidic pollutants towards Europe, particularly Scandinavia. Clark et al (1984) used the Meteorological Office research aircraft to measure plumes from Eggborough power station travelling in a variety of different weather conditions. In January 1981 they tracked a plume across the North Sea. The dispersion of the plume was measured, with the aid of SF_6 tracer gas. The plume was carried across the sea in conditions somewhat the opposite to those that may cause European contributions to elevated fine particle concentrations in the UK. The paper notes the importance of the inversion trapping the plume. These conditions seem reminiscent of the March 1996 episode, except with flow going towards instead of from the continent. Crabtree (1982) has noted the tendency for reduced lateral dispersion over the sea, because of the lack of topographic interference to cause meandering in the wind direction. This, along with an inversion above the plume(s), provides a mechanism whereby the plumes may travel long distances. Chemical processes in the polluted air generate secondary aerosols that can be measured as part of the PM_{10}. In terms of the export of pollutants to Europe, a westerly flow is required. The Lamb Weather types for this would be described as westerly, north westerly and south westerly. Using Lamb's seven basic types, the percentage over many decades were NW ~5% and W ~25%, a combined frequency of ~30% (see Lamb (1972), pp. 14-15). In contrast the percentage for easterly flow was ~8%. In the UK, flows toward the continent are nearly four times more likely than flows from it. It would be interesting to see if modelling predicts any UK contributions to background PM_{10} on mainland Europe; the above data suggest this is possible.

6.7 Sustained Secondary Aerosol Episodes

An elevated incidence of PM_{10} exceedences attributable to distant sources requires special conditions to be fulfilled, such that:

1. The meteorological (and emission source) conditions leading to high concentrations last for many hours, because the 24 hour average concentration must exceed the limit if an exceedence is to occur.
2. The same general wind direction should be maintained for a long period; the orientation of the isobars should remain relatively unchanged. Since the wind direction is very variable in light winds, and is undefined in calm conditions, it also suggests that a moderate wind speed is a requirement for PM_{10} exceedences with a significant secondary contribution. In moderate or strong winds, the direction of flow is well defined.
3. At some point downwind of the source, and in an area encompassing the monitoring station, the plume(s) must reach the ground. If the source is elevated then the ground level maximum concentration occurs at quite some distance

from the release point. This would be the case for the Eggborough plume in Clark et al. (1984). They found reasonably adjacent and parallel isobars, so the flow was fairly straight and well defined in direction. Moderate wind speeds would assist vertical mixing by generating turbulence through drag at the surface, bringing pollutants to the ground. The drag increases when the flow leaves the sea and passes over land.

4. The plume(s) should have reduced horizontal meandering; sideways motions of the plume will take the material away from the monitor and reduce the 24 hour average concentration that is measured. Travel over the sea tends to have less horizontal dispersion, other factors being the same.
5. The plume(s) should probably suffer a restricting inversion layer above them, so that concentrations remain high during plume travel and when the plume reaches ground level.
6. The possibility of several large plumes overlapping or combining may also be a factor.

Secondary particulates are aerosols formed via reactions of gaseous effluents. If we are most concerned with these aerosols, then we may enquire into how the UK stations may receive elevated aerosol contributions. This is conditional upon the above conditions being met such that distant elevated large point sources are able to contribute to the aerosol burden at ground level. This is quite a demanding multiple condition, one which one may suppose is fairly infrequent. Its frequency will increase with the larger the number of elevated sources that are located to the east; more easterly sources mean more easterly wind directions can bring material to the monitor. We suggest that research is needed to explore the relative importance for UK secondary aerosols of distant elevated point sources versus the larger number of smaller ground level sources. This may be better understood, perhaps through modelling or field studies, and might assist policy formulation.

In this work the data on Lamb Weather Types and on North Atlantic Oscillation has been used to shed some light on the frequency of easterly flows that might bring secondary aerosols into the UK. The data show that if these larger features of the general circulation are important, then they are by no means ‘as regular as clockwork’. Values of NAO have varied in a somewhat erratic manner and it is only through diligent study using time series and spectral techniques that meteorologists have discovered its possible importance. (El Nino and its associated Southern Oscillation is a better publicised large scale switching of states in the circulation.) It is clear that whilst large scale meteorological processes may govern the occurrence of pollution episodes (primary or secondary), they are not so regular or repeatable that robust predictions of the recurrence of pollution episodes can be made at the present time. The current evidence suggests that easterly weather types may occur approximately 8% of the time. If the above multiple condition is required for distant elevated plumes to cause raised and sustained aerosol concentrations at ground level in the UK, then their frequency is perhaps significantly less than this figure of 8%.

It is important to stress that this was a study of limited resources, designed to take a broad perspective to gain some understanding of the factors that influence pollutant episode recurrence. It was never designed to be a detailed modelling study.

In future work it is suggested that the monitoring data at other sites should be studied, and that long term records from remote rural sites are needed. Perhaps the reverse flow case of UK contributions to European PM_{10} episodes should be examined since many studies of UK power station plumes were carried out a number of years ago. If modelling of European contributions are sought, the role of source height should be studied. It should not be assumed without evidence that all continental sources are equally important when the UK experiences elevated secondary PM_{10} exceedences in easterly air masses. Elevated point sources within the UK may also play a part in this.

7. MODEL APPLICATIONS

There are several powerful modelling tools that can be applied to the investigation of factors causing air pollution episodes to come and go in line with the vagaries of the weather. These tools all rely upon using data generated by numerical weather prediction models.

7.1 Trajectories

Distance is the integral of velocity. Forward or back trajectories from some point A to another B are calculated by standard procedures that integrate the velocity vectors derived from numerical weather prediction. Air leaving emission sources may arrive at some receptor point of interest; trajectories enable the path of the air to be tracked and analysed. Data retrievals and trajectories may be useful in comparing recent trajectories from recent years (to which available trajectories are readily available as they need detailed meteorological fields, not just single point data) to the wind direction distribution at a single site. Whilst an exact correlation between trajectories and the wind rose may not be seen, broad features of synoptic flow will nevertheless appear at the 10 metre high synoptic anemometer. Some statistical correlation here may be useful in this study. The changes of wind speed and direction at different heights must be taken into consideration when selecting which trajectories to calculate. (The NAME model does this automatically; as particles are dispersed in the vertical they follow a changing wind field.)

7.2 Trials using NAME

A trial run of NAME was carried out, to get a feel for how the model might be applied to this study. Strictly, investigation of results produced by NAME falls outside this Contract. NAME can be run for 1 month intervals. Each run takes a few simulated days for the model to 'run up'; i.e. for the Lagrangian particles to be spread across Europe and meaningful simulations of secondary pollutants to be produced. We anticipate more detailed runs to include large point sources as stacks in the UK and Europe. It is important to be able to explain the easterly bias in PM_{10} fluxes that was seen at London Bloomsbury.

7.3. Climate Simulations of future conditions

In order to address the future behaviour of primary pollutants, it was planned to use data from the Met Office climate simulations. These provide data for nominal days up to many decades into the future. However the level of detail of information in the archives is much less than for numerical weather forecasts; for example wind speeds and temperatures may be available just once per day. Furthermore there is the (unexpected) complication that climate simulated years contain 360 days; comparison of the most recent past decade with actual pollutant observations will thus prove curious (if not impracticable): how are model dates to match monitoring (real) dates? Although Dr Dick Derwent has climate data that can be used, they have not been used here; this preliminary investigation was needed first to be sure if the meteorological indicators of pollution episodes would tie in with observed episodes. The present study has sought to establish simple verified indicators, and then to assess their frequency.

7.4 Automated Objective Weather Typing

Spellman (2000) provides a short review and brings the topic of weather typing up to date. After mentioning the work of several authors, including Lamb (1972), he says that most synoptic techniques classify each day of weather according to the direction of the flow. They describe the conditions associated with each type. He describes how Jenkinson and Collison (1977) at UEA extended Lamb's method by developing an objective scheme. The appendix to Spellman (2000) gives the algebra and coefficients used to calculate the flow directions, the total geostrophic flow (i.e. based on the isobars assuming a balance between pressure gradient force and the Coriolis acceleration that is associated with the earth's rotation), and the vorticity (curvature, angular velocity and sense of rotation) of the flow. Whereas Lamb and workers of his time used manual analysis of the synoptic charts, the objective scheme can be automated and run from a small cluster of grid points in the Analysis fields in numerical weather prediction. These are the fields that have assimilated the latest observations and represent the starting conditions from which the weather forecast will be calculated. Spellman (2000) used just 9 grid points, so any calculations as in his Appendix will be very rapid. Once the indices are calculated, they are sorted into weather types according to the rules cited. Jenkinson and Collinson (1977) have first applied the objective method over the British Isles; its attraction in the present context is that it could be fairly easily coded to run from climate simulation model products from the Hadley Centre. This would be much more rapid than running the NAME model, or even calculating trajectories, for extended periods of climate simulation. It might for example show something of future trends in easterly versus westerly weather types in the climate runs. Spellman (2000) concludes by discussing the most appropriate pressure level (e.g. 500 mbar perhaps at lower latitudes) at which to calculate the indices before typing. A future project might for instance explore the relative frequencies of weather types as diagnosed by such an automated scheme using climate simulations; this would complement climate model studies of low level winds and temperatures which we saw elsewhere are important for primary episodes.

7.5 ECMWF Archives of 'Past' Analysis Fields

The European Centre has begun to assemble a remarkable data base of 'past' meteorological fields. These are generated from the ECMWF weather forecast model that is initialised from the old meteorological observations. This work should generate Analysis Fields that date back many decades. It is in principle possible to run the Met Office NAME model from these archives. In this way the dispersion of primary emissions, and formation of secondary pollutants, from a reference emission scenario, may be simulated. Such a study is mentioned for completeness but falls outside the scope of the present Contract. In terms of analysing the frequency of episode meteorology, these data may be very useful. Significant effort may be required in order to use them however.

8 FUTURE RESEARCH OPPORTUNITIES

The Department asked that areas of work that were explored here but which could be the basis for future research projects should be identified. Topics that have been identified in this study include:

1. Investigation into pollutant flux rose diagrams: their applicability as a tool for the analysis of air quality monitoring data with reference to possible source attribution. To take the work from this report forward, work is needed to study other monitoring sites, both near identified sources, and remote rural sites. The relevance of the technique to a wider range of pollutants should also be investigated. This report briefly examined an urban site, London Bloomsbury, and an industrial setting, Port Talbot.
2. More rigorous review of large scale climate cycles or quasi periodic oscillations, to evaluate their significance for the pollution climate. Underlying geophysical (meteorological/ solar/ oceanographic/ stratospheric) mechanisms that influence climate in the UK should be considered: a review of the literature with reference to current Hadley Centre expertise in the topic. The present work only had scope to provide a fairly superficial scrutiny of such geophysical processes. This work would complement studies using climate model simulations (below).
3. Studies using the climate model data:
   - A rigorous study of whether climate model data are produced in a form that is suitable for pollution climate studies.
   - The simple indices like *Tu* and the automated Lamb Weather Types can be calculated from climate model outputs.
   - The NAME model can be used for simulations of periods having especial interest, and to investigate easterly causes of elevated PM_{10} flux.
   - The 360 day climate model year might also be examined; does it materially affect any conclusions that might be elucidated regarding likely future pollutant trends?
4. Model studies using ECMWF Analysis fields together with NAME model simulations of particular periods or episodes.

5. Evaluation of UK contributions to background PM_{10} concentrations on mainland Europe.

9 CONCLUSIONS

This research has investigated the evidence from the available meteorological and pollution archives for as long a period as was possible. It develops an understanding of the irregularly recurring nature of primary and secondary pollution episodes. A number of very interesting themes emerged and some of these could lead to new research questions. Over the period of study, 1949 till almost the present day, pollutants have changed, sites have changed, and ways of monitoring have changed. The philosophy adopted here was to seek ways whereby the ephemeral nature of past pollution records might be circumvented. This report is an introduction to episode recurrence. It poses new questions. They merit study in order to provide further technical input to the development of policy that recognises the meteorological controls that determine whether or not pollution episodes are experienced in one year or another. It is recommended that natural meteorological variability should continue to be a factor for consideration when revising policies on emissions controls. Such variability is noticed over periods spanning several decades. Further investigation of the underlying geophysical processes is required in order to explain this variability in the pollution climate.

The present work found that:

1. Frequency analysis of meteorological variables can be used to compare years.
2. Visibility in its simplest inverse relation to aerosol loading cannot be used in this work, owing to frequent high humidity. More complex analyses which also allow for Relative Humidity would be needed to explore this idea further. Secondary particulate episodes may be much more elusive in terms of defining a sort key or identifier; the role of visibility was investigated because in the absence of high humidity it was likely to be linked inversely to particulate concentrations. A multi variable key seems most likely.
3. Primary winter smog episodes can be distinguished. The major winter primary episodes for Black Smoke, Oxides of Nitrogen, and fine particulate matter as PM_{10}, in London between 1949 and 1997 were identified using the synoptic meteorological archive from London Heathrow Airport. The temperature-wind index WI = Tu defined here shows large values in these episodes, typically in the range $Tu>10$. The index is applied under the assumption that no changes of emissions control policy are operating, but that emissions do respond to severe winters. The magnitudes of the index Tu may be ranked in order to compare on a common numerical scale the potential magnitudes of air pollution severity in different winters.
4. The index Tu also showed a modest skill in pointing to some summer ozone (i.e. secondary) episodes. High temperatures and light winds are characteristic of summer anticyclones, when if the air follows a suitable trajectory into the UK, high ozone may be observed. A summer index SI was defined and is closely related to Tu.

5. It would be interesting to test the index WI in other countries whose cities suffer from winter primary episodes.
6. Secondary episodes proved much more elusive. Long-range transport plays an important role, so the hourly wind directions at 10 metres were unable in the present study to distinguish periods of high secondary pollution. However when used in conjunction with PM_{10} data there was a strong directional character to the pollutant flux. North Thames directions featured in the flux rose plot.
7. A preliminary study using NAME sought to explore the relative contributions of UK and European secondary aerosols to UK particulate episodes.
8. Some parts of this work were presented at the Third International Conference on Urban Air Pollution Measurement, Monitoring and Modelling, at Loutraki (Greece), in March 2001: see Dixon and Middleton (2001; Annex 1); Dixon and Middleton (in preparation).

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ANNEX 1

AN ANALYSIS OF THE METEOROLOGY ASSOCIATED WITH AIR POLLUTION EPISODES IN THE UK

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ABSTRACT

This work analyses the historical record of meteorological observations and ambient air pollution monitoring in the London UK area. We identify meteorological related variables that correlate with different types of pollution episode, estimate the frequency of such weather conditions in the meteorological record and provide insights into the likely frequency of episodes occurring again. This study of historical synoptic data does not use emissions inventory data. It was intentionally designed to not require the complexities of dispersion modelling. Hourly meteorological observations from the station at London Heathrow airport have been analysed from 1949 to 1998. Parameters included: temperature, wind speed and direction, relative humidity, Pasquill Stability, visibility, boundary layer depth and precipitation rate. Analyses of the type used here have not revealed secondary particulate episodes, whether studied in the form of sulphate or secondary PM_{10} episodes. An assessment of the meteorology in 1996 and 1998 is undertaken here.

1. Introduction

Natural fluctuations in climate cause significant changes in air pollutant concentrations. When analysing episodes, providing policy advice to manage future air quality and seeking to avoid future pollution episodes, meteorology should be considered. The technical feasibility and economic cost of engineering changes in order to meet future AQ objectives depend on the magnitude of emission controls that might be required by future regulations. Regulations respond to perceived understanding of air pollution. There have been widespread discussions of future air quality and the choice of meteorology that underpins modelling. The current Air Quality Strategy for England, Scotland, Wales and Northern Ireland (AQS, 2000) includes maps produced using meteorology from 1996 and 1998. Analysis of the pollutant concentrations and exceedences of current standards indicate that 1996 was the 'worst' year (37 exceedences of the PM_{10} standard/objective $50ug/m^3$) and 1998 was the 'cleanest' year (3 exceedences) in recent years. A key question to be asked is 'What is so unusual or different about the meteorology in 1996 and 1998?' The aim of this paper is to investigate this question.

2. Methodology

Meteorological Office archives of synoptic observations were retrieved for London Heathrow (site 5113, latitude 51.48N, longitude 0.45W) from 1949 to 1998. The anemometer height was 10m and roughness length = 0.2 m. The practice at Heathrow is for 24 hourly observations per day. Wind direction and wind speed are the 10-minute mean winds reported at the time of observation. Parameters were recorded to the following accuracy: temperature: recorded to nearest whole degree centigrade; wind speed recorded to nearest whole knot and converted to meters per second; wind direction recorded in degrees true from which wind blows to the nearest 10 degrees. Each of the 48 years were analysed by other parameters averaged by wind speeds and also by wind direction.

3. Results and Discussion

3.1 Wind Speed

The frequency of each wind speed was plotted as a monthly average and hourly average for each year. Initially it was considered whether the frequency of calms in any month was associated with episode conditions. Plots were also derived for increasing wind speeds: 1-3 knots; 4-6 knots; and 7-16 knots. Figure 1a shows the frequency of calms for all months in the year for 1949-1998; Figure 1b shows the frequency of calms in the month of December in each year. These figures show that the incidence of calm conditions was high in months and years in which severe episodes were recorded. The notable December episodes of 1991 (NOx) and 1952 (Black Smoke) were clearly seen, even when data are taken a whole month or year at a time.

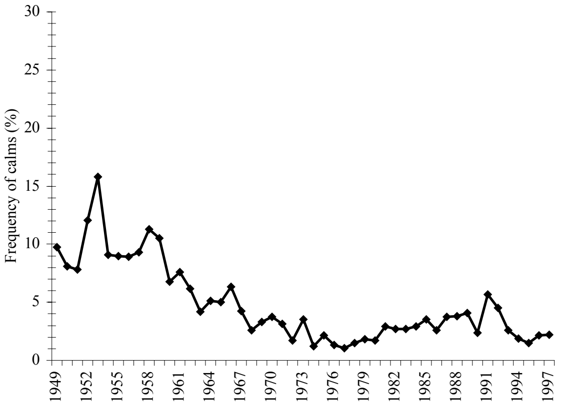


Figure 1a. Frequency of calms 1949-1997

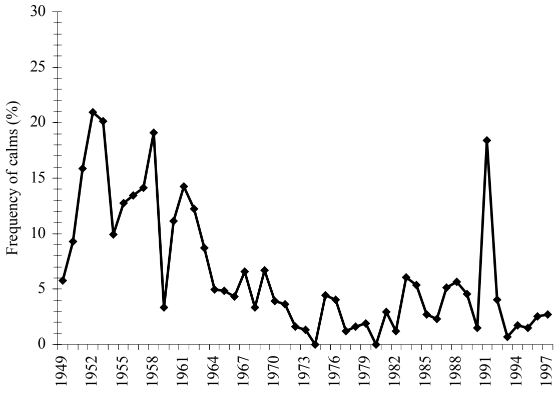


Figure 1b. Frequency of calms in December 1949-1997.

More surprisingly, a general decline in the frequency of calms was seen from 1949 through to 1997. This result was not anticipated: it could be an artefact Furthermore, there is seen a large peak in the frequency of calms in December 1991, just when London had a notorious winter smog of elevated NO_2 concentrations. In the absence of other information it was concluded that the decline in frequency of calms was a real effect, but that in occasional years, as seen in 1991, a prolonged calm period of very cold weather and associated high pollution occurred. There seems no clear nor regular pattern in the incidence of calms in different years. Light winds (1-3 knots) were somewhat similar in behaviour

In December 1952, the wind was calm for 20.97% of the month; in December 1991 it was calm for 18.41% of the month; but in December 1996 it was calm only for 2.55% of the month. There were notorious winter smogs of primary pollutants then in 1952 (Smoke/SO_2) and 1991 (NO_x), but not in 1996. In March 1996 it was calm for just 1.21% of the time, suggesting any particulate episodes in March 1996 were most unlike those winter smogs of 1952 or 1991. For interest, the month in 1996 with the greatest incidence of calms was October, with just 4.03% calms in the month and no recorded particles exceedences. These results shows just how closely the pollution climate is tied month by month to the meteorology of the period. Broadly, lighter winds were less frequent in the 1970s onwards than in the 1950s. From a policy point of view, there is nothing in these historical data to tell us whether such a trend remains, or whether calm conditions will be much more frequent in the future. Climate simulations are the main source of information for possible future wind speeds.

3.2 Wind sector analysis

Rose diagrams show the wind directions in the usual convention, in degrees clockwise from North from which the wind was blowing. In the plots discussed below, we excluded calms. We show the frequency of wind from each 10-degree sector. In addition, each of the other parameters was sorted by wind sector and averaged. This information was calculated for each year using hourly data. An average for each wind sector over the years 1949-1998 is also shown.

3.3.1 Wind frequency rose

Plots the frequency (%) that the wind was from each direction, obtained simply by sorting the data and counting occurrences in each direction. Their frequency is the percentage of the total observed hours. There are 36 individual wind directions at 10 degree steps and therefore uniformly distributed winds would have a frequency of 2.78%.A larger radius shows the most frequent or prevailing winds: Prevailing SW winds from 230° reach 7.4% in 1994 (Figure 2a). In most years this prevailing SW flow was present from 4-7% of the time. In contrast, winds from the opposite NE direction are less common, usually 2-4%. E winds are even less common, 0.5-2.5%, of the time. In 1996 the frequency of winds from 20-50° was ~4%, but only ~1-3% for directions 60-160°. The year 1998 also showed a similar low incidence of winds from an easterly quadrant. Easterly Types may be infrequent, but the pollution rose below reveals their importance for the fine particulate pollution climate.

3.3.2 Pollution rose

Pollution data shown as PM_{10} in µg m^{-3} measured hourly means from the AUN site at London Bloomsbury. Note that all PM_{10} concentrations used in this report were the original TEOM value (and were not corrected to any equivalent gravimetric figure). Pollution data are sorted on the basis of their direction into groups and the hourly values of PM_{10} concentration are averaged. In the pollution rose, the radius is proportional to the average air pollution associated with the wind from each direction. For the PM_{10} data in the years studied, these directionally averaged concentrations were always greater than 20 µg m^{-3} in all wind directions (Figure 2b).

As expected, air from the SW had the least PM_{10} burden. At 230°, concentrations ranged from 19-23 µg m^{-3}. However, with winds from the ENE, E, and ESE (directions 50-110°) the pollutant burden was much larger, ranging from 27-50 µg m^{-3}. More generally, the usual range of concentrations in the other directions was 20-35 µg m^{-3}. The shape of the pollution rose is different from the wind frequency rose; the least common wind directions carried the higher pollutant concentrations. There is quite clearly a polluted easterly sector or quadrant (directions 50-140° from N, some 90° wide), which has elevated PM_{10} values. This applies even in 1998, the year in which the easterly elevation of particle concentrations was least pronounced (but still ~27-30 µg m^{-3}). It so happens that 1996 stands out in severity, reaching 47-50 µg m^{-3} for the three directions from 60°, 70° and 80° from N.

The years 1993 and 1995 were also greater than the average PM_{10} in each sector, approaching 44 and 43 µg m^{-3} for PM_{10} concentrations respectively from the easterly quadrant. It would be wrong to interpret 1996 as being unique; in terms of the PM_{10} rose it is simply worse in its easterly quadrant. In other directions it seems not so unusual. Nevertheless, in view of the number of exceedences seen during 1996, it is an important year to investigate.

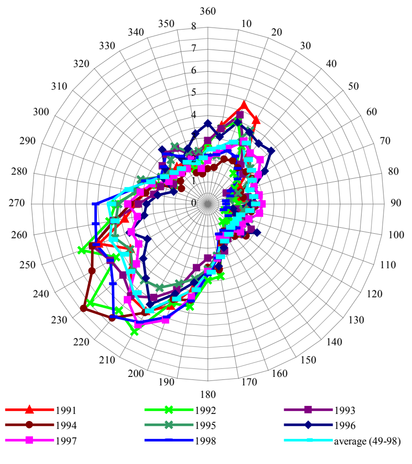
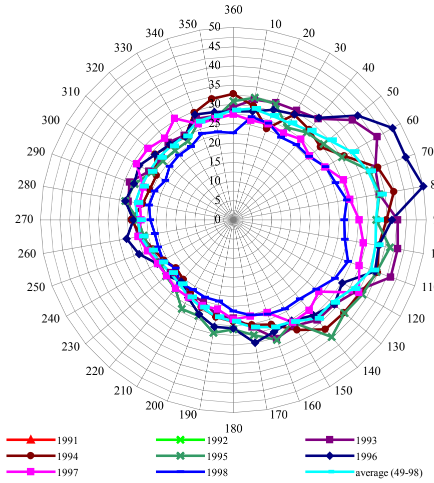


Figure 2a Frequency of winds from each sector (1991-1998).

Figure 2b Average PM_{10} concentration (µg/m^3) from each sector (1993-1998).



3.3.2 Pollution Flux rose

Rose plots of the flux of pollutant (PM_{10}) to the monitor reinforced the importance of easterly flows.

3.3.3 Temperature rose

Most directional temperature averages lay between 9° C and 12° C (Figure 3a). For many directions this range applies and the years studied are all fairly similar to one another. Year to year variation in temperature was greatest in the same easterly quadrant seen above in the pollution rose. In 1996, for directions 10-20°, and also 30-90°, were noticeably colder than average, averaging ~6° C. Clearly, during 1996, some flows from near N and NE were unusually cold. By way of contrast in 1998, the wind from this quadrant was warmer, reaching a directional average of 15.5° C in flow from 80°.

3.3.4 Wind speed rose

The radius shows the average wind speed associated with winds from each direction (Figure 3b). A strong south-westerly and north-easterly bias can be seen with these directions attaining the higher averaged wind speeds. Going clockwise from SE round to N (i.e. 120° round to 350°) the winds speeds in all years were close together in magnitude. They rise smoothly in SE flow from ~3 ms^{-1} up to a SW maximum of ~4.5 ms^{-1} and fall smoothly again to a minimum of ~2.5 ms^{-1} near N. Winds from the NE direction have a greater year to year variation in wind speed. For flow from 60°, 1993 and 1998 showed an average speed ~3.0 ms^{-1}, whereas 1996 had a much faster average speed of 5.5.ms^{-1} for the 60° direction. Other years, and the flows around a NE direction, all fell between these limits of 3.0-5.5 ms^{-1}. It can also be seen that the years 1991-8 were generally calmer that the average (1949-98) for each wind sector.

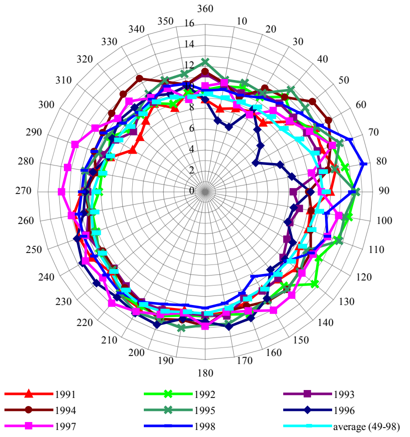


Figure 3a Average temperature (^{o}C) for each wind sector (1991-1998).

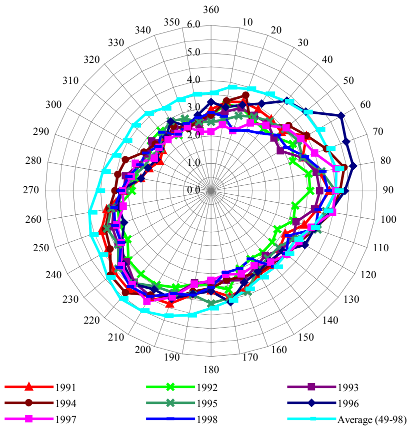


Figure 3b Average wind speed (m/s) for each wind sector (1991-1998).

3.3.5 Features of 1996

In these rose diagrams, 1996 stands out in some respects, whilst in many ways all years have some marked similarities. They all show the prevailing winds from SW and NE, with a bias to higher pollutant levels in the easterly quadrant of 50-140°. This easterly quadrant shows the greatest year to year variability in speed, temperature and PM_{10} burden. The year 1996 shows the following features in this easterly quadrant: Highest PM_{10} concentration, Coldest temperature, Highest wind speed.

The rose plots do not confirm whether these features are causally associated. However they do merit consideration in that they may be associated with outbreaks of polar continental air masses into the UK. Such air masses have some characteristic properties, which, in view of the need to identify features associated with secondary PM_{10} episodes, merit discussion. It is also necessary to explore the wider scale meteorological conditions under which the less frequent but more polluted easterly and north easterly flows can occur. The pollutant rose plots for particles point to an important easterly contribution, but cannot reveal whether this is of UK or European contribution. In conclusion, 1996 and 1998 do not seem as different as might be supposed, but the difference in their number of PM_{10} exceedences seems related to easterly flow.

4. References

AQS (2000) *The Air Quality Strategy for England, Scotland, Wales and Northern Ireland.* Cm 4548. SE 2000/3. NIA 7. The Stationery Office, London.

5. Acknowledgements

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