The influence of climate on air and precipitation chemistry over Europe and downscaling applications to future acidic deposition

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ABSTRACT: Atmospheric circulations, and related climate conditions, exert a strong influence on the transport and ultimate deposition of atmospheric pollutants. One potential outcome of climate change is alteration of atmospheric circulation patterns. The first stage in the development of a downscaling methodology to assess the influence of these possible future atmospheric circulation changes on transport and deposition of atmospheric pollutants over Europe is described. Firstly, the main modes of regional-scale circulation over a domain covering the North Atlantic and Europe are determined using principal components analysis of sea level pressure patterns. To determine whether the principal components represent circulation types found in reality, and show realistic relationships with surface temperature and precipitation amount, they are compared to the Lamb Weather Types, Central England Temperature, and England and Wales Rainfall. There are statistically significant relationships between some of the principal components and aqueous and ambient pollutant concentrations at 5 selected stations from the European Monitoring and Evaluation Programme monitoring network. Although, for any one combination of principal component and pollutant variable, the level of explanation can be quite small, notwithstanding some levels of variance explained for individual combinations being up to 69% (45% for the winter season, described in detail here). Because the pollution climate at any one location is a function of many combinations of circulation/pollution relationships, despite the relatively small magnitude of variance explained for individual combinations in this study, the results confirm the utility of atmospheric circulation principal components in deriving downscaling relationships for surface pollution behaviour (via eventual multiple regressions, not reported in this paper), as well as giving further insight into the climatic influences on air and precipitation chemistry over Europe.

KEY WORDS: Acidic deposition · Air pollution · Downscaling · Synoptic climatology · Principal components analysis · Climate change

1. INTRODUCTION

Acidic deposition is the final stage in a chain of processes linking emission sources and atmospheric transport and transformation. Meteorology exerts a controlling influence in this chain. A number of synoptic climatological approaches have been used in attempts to embrace a range of the pertinent meteorological processes. Examples include the use of Lamb Weather Types (LWT) (Davies et al. 1986, 1991, O'Hare & Wilby 1995), statistical clustering of meteorological variables (Sanchez et al. 1990, McGregor & Bamzelis 1995), and the grouping of airmass trajectories (Dorling et al. 1992, Moody et al. 1995). Such studies have demonstrated clear links between atmospheric circulation and the transport and deposition of acidifying compounds. An important implication is that climate change, with possible circulation changes, may lead to changes in the transport and deposition patterns of acidic species (Davies et al. 1986).

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The known links between atmospheric circulation patterns and surface climate variables have been used to produce possible future climate scenarios, on the regional scale, by the synoptic climatological classification of atmospheric circulations projected by general circulation models (GCMs) (e.g. Hewitson & Crane 1992, Goodess & Palutikof 1998). Such methods are now widely used (Hewitson & Crane 1996, Wilby et al. 1998). Extension of such a 'downscaling' methodology to the issue of possible future acidic deposition is, therefore, quite logical, given the clear links between present acidic deposition patterns and atmospheric circulation. The objective of this work, as the first step in the production of possible future scenarios of acidic deposition, is to establish whether or not there are distinct relationships between pollutant concentrations and depositions and atmospheric circulation, using the downscaling techniques which have been applied to surface climate variables. If successful, this would then allow the first application of a downscaling technique to future acidic depositions, using the projections of future circulations from GCMs. This would complement the previous, different, approaches to the assessment of the possible influence of future climate change on acid deposition over Europe. Pitovranov (1988) used an analogue method to suggest changes in Grosswetterlagen frequency, and the consequent effects on pollutant transport. Smith (1992) considered the combined effects of possible future emission changes, and possible changes in precipitation regimes, and Alcamo et al. (1995) linked integrated assessment models of climate change and acid deposition.

This study uses principal components analysis (PCA) to determine the main modes of spatial variation in atmospheric circulation over the North Atlantic and Europe. Comparisons are then made with an established climatological classification (LWT), and surface observation (Central England Temperature [CET] and England and Wales Rainfall [EWR]), to confirm the physical sense of the principal components (PCs) of the circulation. Relationships are then established between the PCs and observed pollutant concentrations and depositions at 5 stations in the European Monitoring and Evaluation Programme (EMEP) monitoring network (Jones 1997, Jones & Davies 1998, unpubl.). This is an important step since it will determine if the links between the circulation PCs and the pollution are strong enough for application to future scenario studies. Moreover, the physical reality of the relationships may be assessed through comparison with circulation/pollution relationships over the same study region determined by a different approach (e.g. Dorling et al. 1992).

The length of the data series used for this work was limited by the availability of the EMEP data. At the time analysis was begun, only about 10 yr of continuous data with acceptable levels of missing data for the variables considered were available.

2. PRINCIPAL COMPONENTS ANALYSIS OF ATMOSPHERIC CIRCULATION

The United Kingdom Meteorological Office (UKMO) northern hemisphere 5° latitude by 10° longitude gridded mean sea level pressure dataset was used (Jones 1987). Analysis was undertaken on a daily basis for the 4 standard meteorological seasons, for the region 40° E–40° W, 30–80° N, and for the period 1982 to 1991. This period corresponded to the time of greatest EMEP data availability for this study. Orthogonally rotated PCA was applied to the data, using the varimax rotation (von Storch & Zwiers 1999). Opinions differ on whether or not to use rotation (Richman 1986, Jolliffe 1987). Because interpretation of patterns is regarded as more straightforward with rotation (Yarnal 1993), it was used in this study. Components with eigenvalues greater than 1 were retained for rotation (i.e. only components which explained more variance than the original variables), a criterion used by Horel (1981) and Villalba et al. (1997). This led to between 14 and 16 components in each season being retained for rotation. The retention of a relatively large number was desirable, since a characteristic of acidic deposition is that it may be episodic (i.e. a large proportion of annual deposition at a site may occur under relatively infrequent meteorological conditions; Smith & Hunt 1978). In extreme cases, in some locations, up to one-third of the total annual deposition of acidic species may occur in 1 day (Davies et al. 1992). Thus, although much deposition may be expected under frequently occurring conditions, inclusion of the more uncommon synoptic types increases the probability that a greater proportion of deposition may be accounted for by the components. Thus components which explain only a few percent of the total variance have been retained. Jolliffe (1982) suggests that PCs with smaller eigenvalues can be important in PC regression. The rationale that PCA identifies the most frequent modes of variance in a dataset naturally means that perhaps very infrequent conditions important to deposition at the stations may not be captured by the classification.

There has been some discussion as to which PC retention rules to use. North et al. (1982) suggest that if the sampling error of a particular eigenvalue is equal or larger to the spacing between it and the neighbouring eigenvalue then the sampling error of the empirical orthogonal function (EOF) is comparable to the size of the neighbouring EOF. Von Storch (1995) suggests that this approach is more objective from a

statistical point of view. Kidson (1988) uses a combination of this method and the 'scree test'. The latter involves plotting the eigenvalue against PC number. A break in slope is then identified, after which the eigenvalues of the following components, which represent uncorrelated noise, should decay exponentially. This methodology has the advantage of being easy to understand, and is relatively conservative. It involves however a subjective judgement about the location of the break in the plotted curve. The results in the following sections, of relationships between the components and CET, EWR, the LWT and with the EMEP station data, support the physical reality of the components retained.

The component loadings are shown in Figs. 1 (winter) & 2 (summer). The maps show the contribution of the variable at each grid-point to each PC. The values in parentheses are the percentage variance explained by each component. In the winter PC3, explaining 9.8% of the variance, for example, the highest variance occurs in the grid points over the southern UK. It can be seen that the majority of the patterns are common between these 2 seasons, as in spring and autumn (not shown), although there are some differences in the ordering. For example, the PC6 map for winter (Fig. 1) is clearly the equivalent to that of PC3 in summer (Fig. 2).

For each time-step of the analysis a score for each PC is obtained, which represents the strength of a PC weight on a particular day. Interpretation of the PC patterns can be aided by the construction of composite circulation plots. Figs. 3 (winter) & 4 (summer) show the mean pressure pattern of those days with component scores of more than twice the standard deviation (positive and negative) from the mean value. A greater number of composites are shown for winter as these are referred to later. These composites reveal that high positive scores correspond to high pressure at the centre of maximum variance, and large negative scores correspond to low pressure. Taking winter PC3 as an
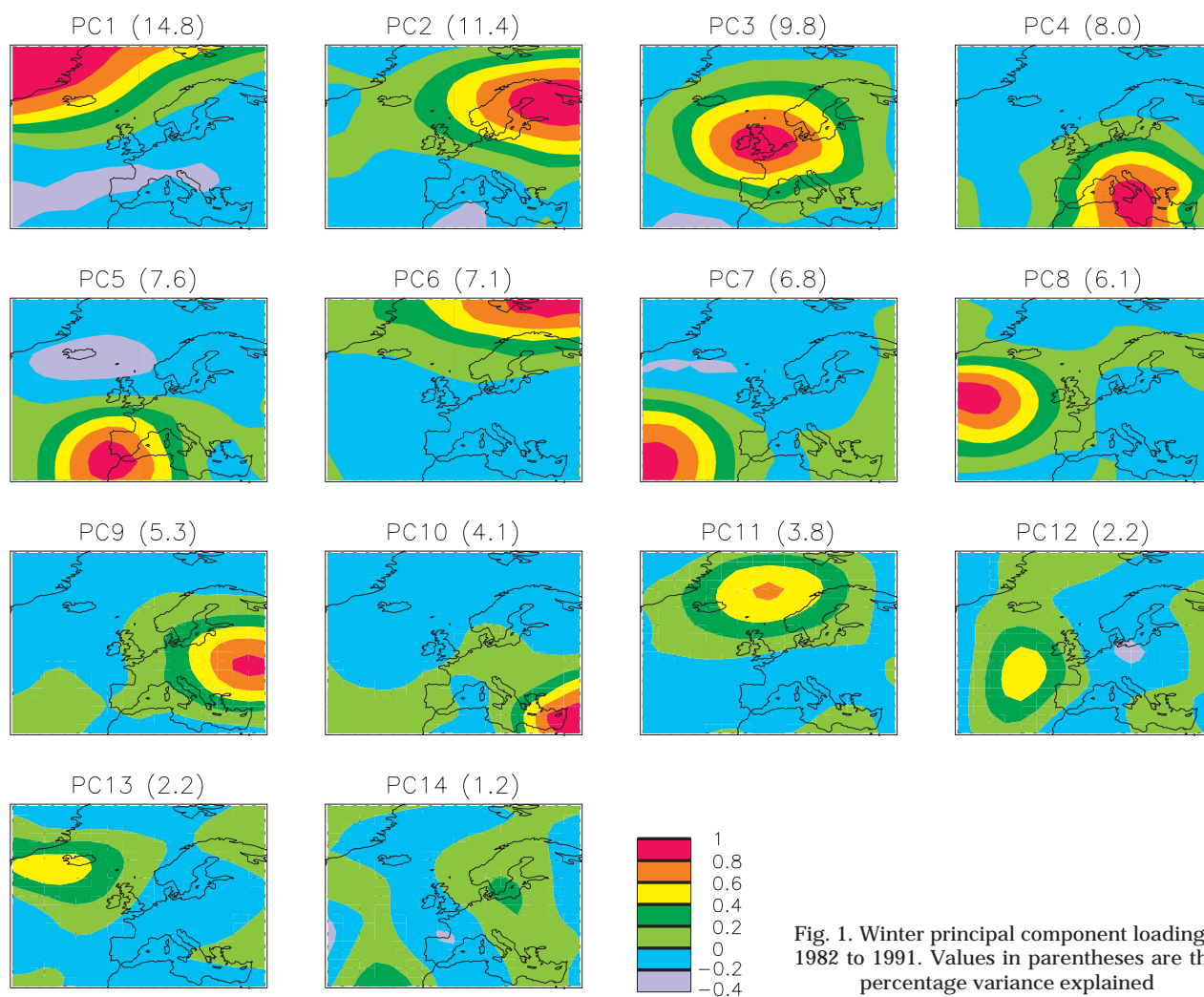

Fig. 1. Winter principal component loadings, 1982 to 1991. Values in parentheses are the percentage variance explained

PC1 (13.6) PC2 (7.6) PC3 (7.4) PC4 (6.4)

PC5 (5.8) PC6 (4.9) PC7 (4.7) PC8 (4.6)

PC9 (4.4) PC10 (4.3) PC11 (4.0) PC12 (4.0)

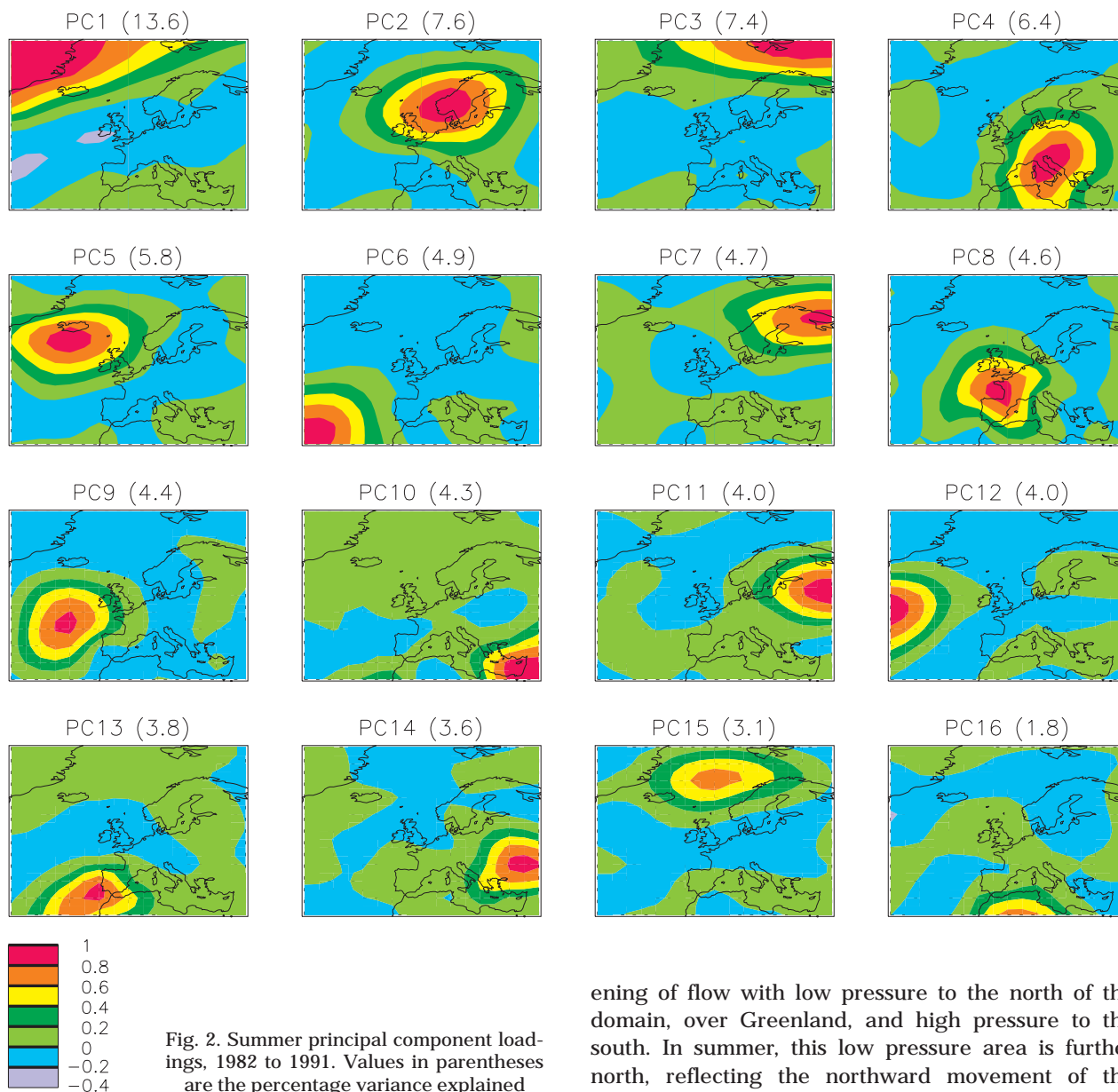PC13 (3.8) PC14 (3.6) PC15 (3.1) PC16 (1.8)

Fig. 2. Summer principal component loadings, 1982 to 1991. Values in parentheses are the percentage variance explained

example again, the composite map for high positive score days shows an anticyclone centred on the region of maximum variance (Fig. 3). Similarly, the composite for the days of large negative scores reveals low pressure in this region. For brevity, hereafter, 'high scores' refer to high positive scores, and 'low scores' to large negative scores.

As previously indicated, the majority of patterns are common to all 4 seasons. The first component in all seasons represents variation in the strength of the zonal circulation. High pressure over Greenland and low pressure to the north of Scandinavia on high score days give a weakened zonal circulation (see PC1 in Figs. 3 & 4). Conversely, low scores indicate a strength-

ening of flow with low pressure to the north of the domain, over Greenland, and high pressure to the south. In summer, this low pressure area is further north, reflecting the northward movement of the northern hemisphere storm tracks, with a ridge of high pressure extending from the southwest of the domain into northwest Europe (compare Fig. 4 with Fig. 3). Another common pattern is a centre in the northeast of the domain (winter PC6, Fig. 1; summer PC3, Fig. 2). This component represents changes in the location of the north Atlantic storm track. On high score days of winter PC6 (Fig. 3), pressure is high over the Barents Sea and depressions are deflected southwards over northern Europe. In the opposite phase, pressure is low in this region as depressions travel to the north of high pressure over Europe.

Further discussion, and interpretation, of the PC patterns, in climatological terms, will be restricted to a comparison with the LWT (below). This is because the purpose of this paper is to establish links with pollutant
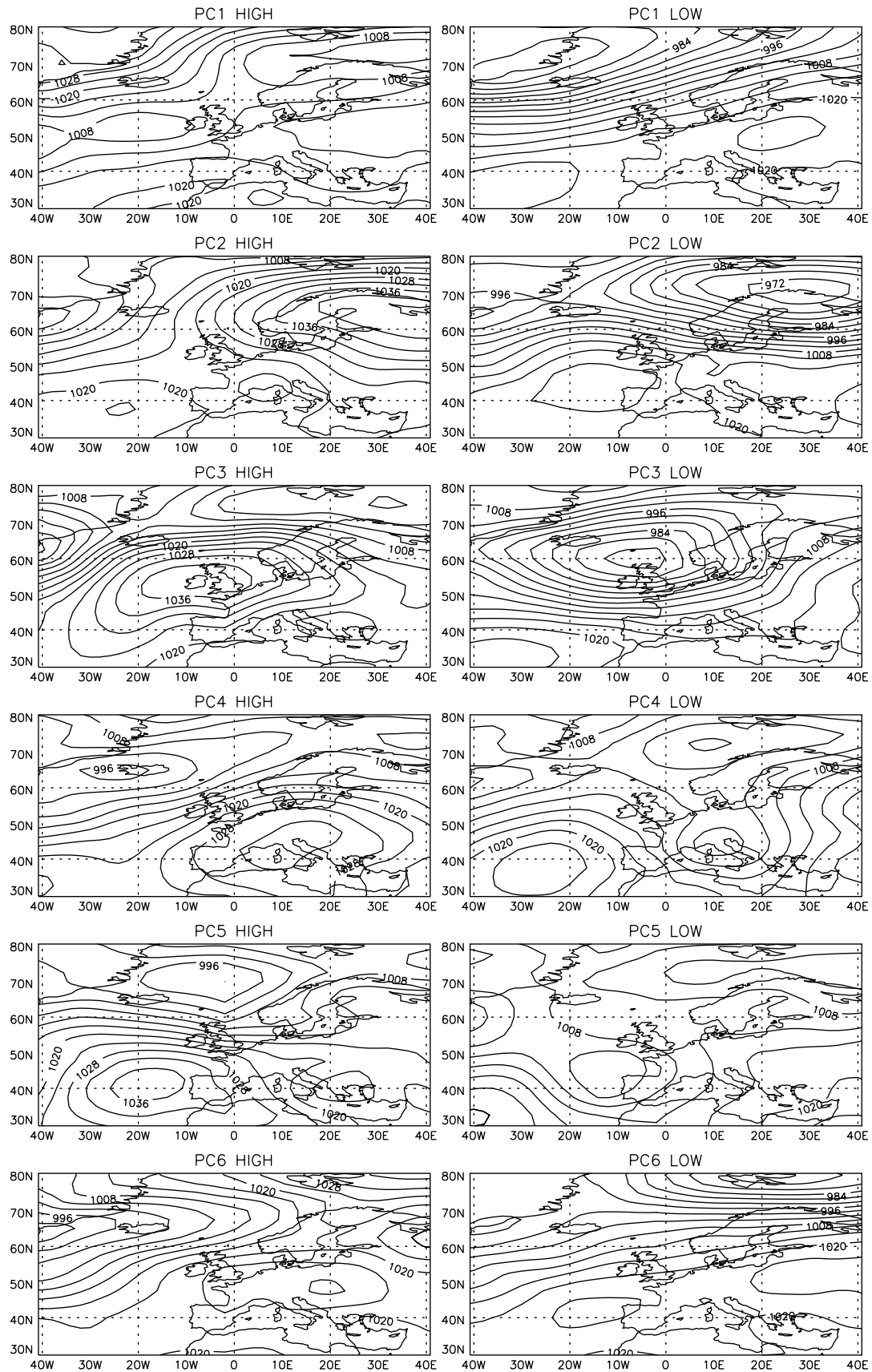
Fig. 3 (Above and on the following 2 pages). Winter mean sea level pressure composites for days with component scores >2.0 (high) and <−2.0 (low), 1982 to 1991
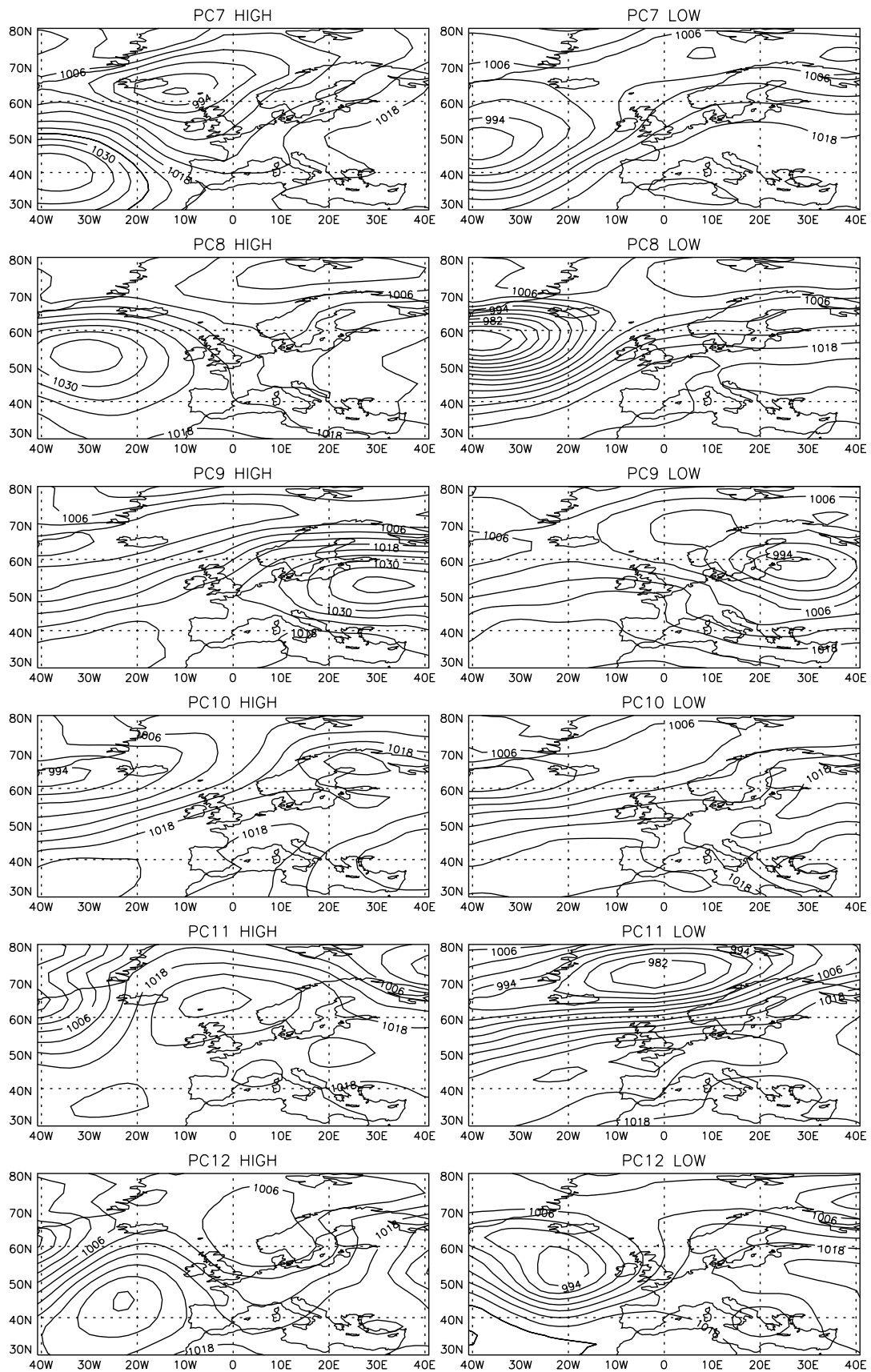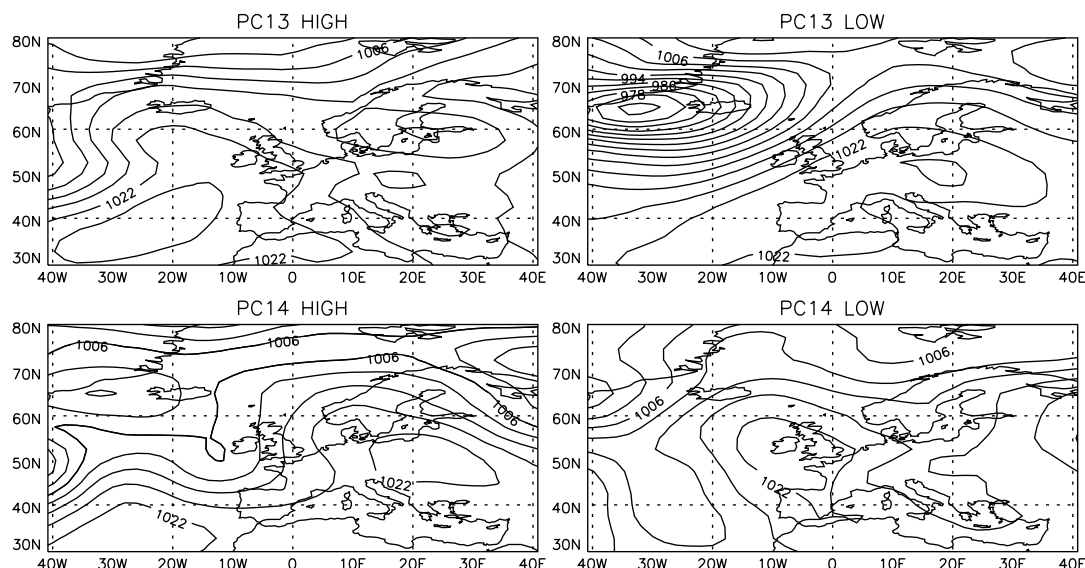
Fig. 3. (continued)

Fig. 3. (continued)

concentrations and deposition, and the sensible physical interpretation of the PCs on the European scale, in all 4 seasons, has been established elsewhere (Jones 1997). However, given the clear utility of the LWT for the study of acidic deposition in the UK (Davies et al. 1991), an examination of how clearly the PCA, at the European/North Atlantic scale, discriminates the circulation patterns on the smaller scale will be valuable.

3. RELATIONSHIPS BETWEEN PRINCIPAL COMPONENTS AND LAMB WEATHER TYPES

The LWT represent a manual daily synoptic classification for the British Isles (Lamb 1972), i.e. the judgement of the analyst is used to categorise the circulation by inspection and identification of the most frequent modes of atmospheric variability according to predetermined criteria. They are based on the surface airflow characteristics over the region 50–60° N, 10° W–2° E. There are 8 directional types, an anticyclonic type and a cyclonic type which together with hybrid types and unclassifiable situations give a total of 27 circulation types. The direction refers to the overall movement of synoptic systems and to the airflow direction over the British Isles.

Correlations were calculated between monthly frequencies of the LWT and monthly means of the component scores. This was done for the anticyclonic and cyclonic types, and for each of the 8 directional types. The cyclonic types were also summed with all the cyclonic hybrid types (e.g. cyclonic westerly + cyclonic southwesterly + ...) to give a cyclonic composite (CC)

class. Similarly, anticyclonic types were summed together with anticyclonic hybrid types to give an anticyclonic composite (AC) class. Tables 1 & 2 show the values of the correlation coefficients for winter and summer, respectively. Significant positive (negative) correlations indicate high frequencies of a particular LWT in months with a high positive (negative) score. To use the example of winter PC3 (a centre over the British Isles) again, here there are significant negative correlations with cyclonic and CC classes (Table 1), and a significant positive correlation with the AC class. The composite plots show a cyclone over this region on low score days and an anticyclone on high score days (Fig. 3).

The northward shift in the zone of westerly flow, with anticyclonic cover over the North Atlantic and Northwest Europe (compare PC1 in Figs. 3 & 4) is reflected by the strong correlations with anticyclonic and AC types (Table 2). In summer, PC3 (the component representing variations in the location of the storm track, as described above) shows a significant positive relationship with cyclonic and CC types, and a significant negative relationship with anticyclonic and AC types (Table 2). Thus, as expected, a high frequency of cyclonic LWT occurs in months with high pressure to the northeast, because of the southward deflection of Atlantic cyclones, whereas high pressure extending over Europe is reflected in the negative relationship between PC3 and anticyclonic and AC types.

Although further examples will not be discussed in this paper, inspection of Tables 1 & 2 indicates that most components are significantly correlated with at least 1 LWT. Results are similar for spring and autumn (not shown here—see Jones 1997), with correlation
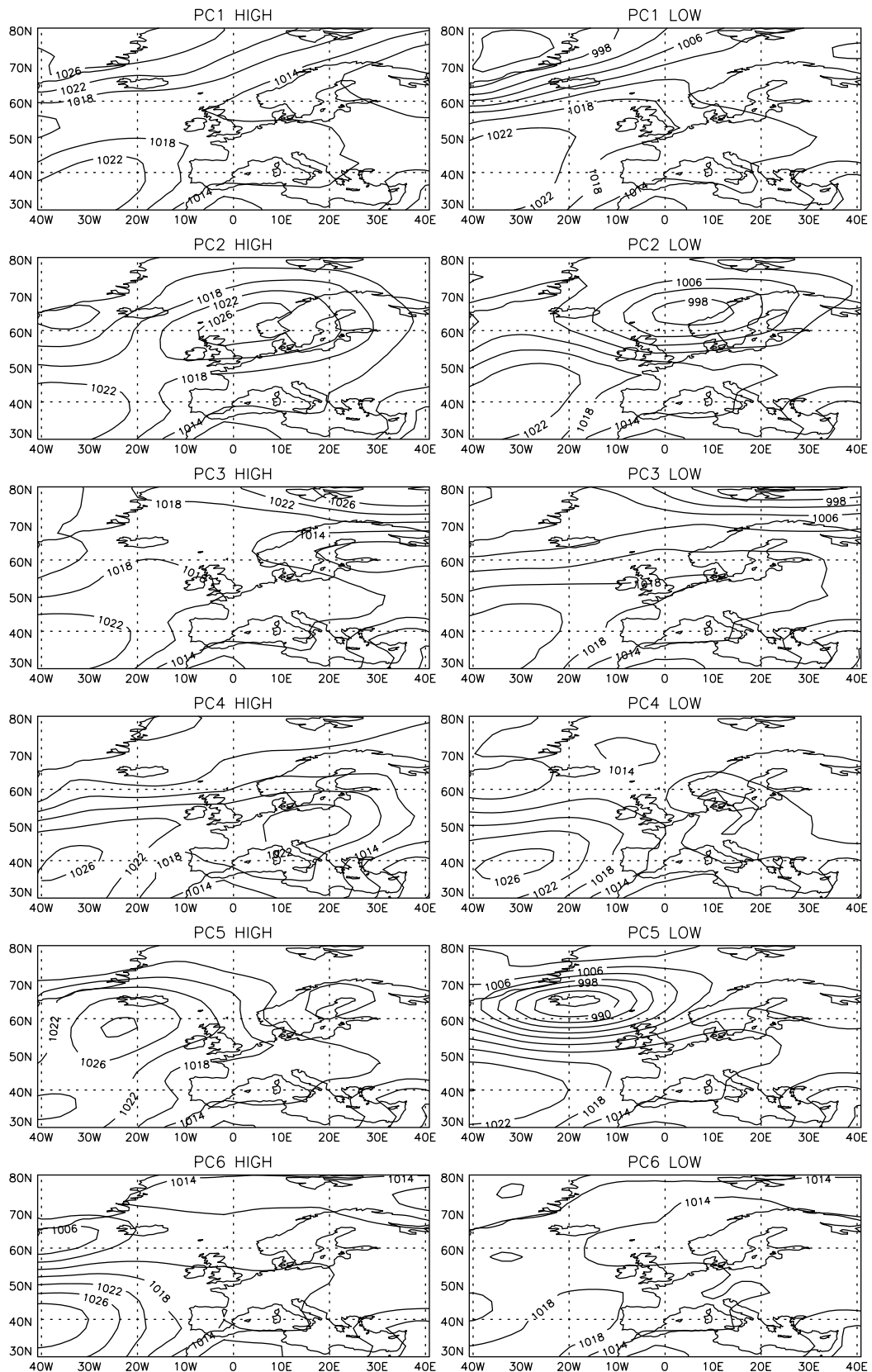
Fig. 4. Summer mean sea level pressure composites for days with component scores >2.0 (high) and <−2.0 (low), 1982 to 1991

Table 1. Winter correlations between monthly mean component scores and monthly frequencies of the Lamb Weather Types. Values in **bold** are statistically significant at the 5% level. n = 27. A = anticyclonic, C = cyclonic, NE = northeasterly, E = easterly, S = southerly, SW = southwesterly, W = westerly, NW = northwesterly, N = northerly, AC = anticyclonic composite, CC = cyclonic composite

	A	C	NE	E	SE	S	SW	W	NW	N	AC	CC
PC1	0.13	−0.12	**0.52**	**0.52**	0.37	−0.32	−0.21	**−0.49**	−0.10	0.17	0.18	−0.40
PC2	0.27	−0.08	0.06	**0.56**	−0.04	−0.03	−0.33	**−0.45**	−0.22	0.20	0.17	−0.05
PC3	0.36	**−0.81**	−0.09	0.31	0.02	−0.07	−0.20	−0.08	0.04	−0.04	**0.58**	**−0.78**
PC4	−0.20	−0.17	**−0.44**	**−0.39**	−0.07	**0.38**	**0.45**	**0.42**	0.17	−0.28	−0.23	−0.14
PC5	**−0.57**	0.06	−0.19	−0.37	**−0.59**	0.03	0.16	**0.74**	**0.53**	−0.12	**−0.45**	0.18
PC6	−0.05	−0.03	0.21	0.22	0.07	0.13	0.21	−0.30	−0.19	0.35	−0.07	−0.01
PC7	**−0.62**	0.32	−0.01	−0.06	−0.31	0.05	0.06	**0.51**	0.25	−0.05	**−0.69**	**0.47**
PC8	0.10	−0.18	−0.31	−0.13	−0.28	−0.37	**−0.53**	0.25	**0.43**	**0.44**	0.20	−0.05
PC9	−0.09	0.23	0.06	−0.16	−0.02	**0.45**	0.25	−0.23	**−0.45**	−0.04	−0.10	0.25
PC10	−0.15	0.10	−0.16	**−0.43**	−0.12	0.22	0.11	**0.40**	0.10	−0.21	−0.14	0.13
PC11	0.24	−0.07	0.37	**0.39**	0.18	−0.23	−0.28	**−0.38**	−0.10	0.04	0.26	−0.05
PC12	0.04	−0.10	−0.23	−0.38	−0.34	0.03	−0.21	0.24	0.30	0.33	0.14	−0.08
PC13	0.13	−0.11	0.33	**0.47**	0.19	−0.02	−0.08	−0.35	−0.24	0.21	0.10	−0.13
PC14	−0.06	−0.20	0.35	0.15	0.30	0.14	0.01	−0.05	−0.02	−0.21	−0.04	−0.23

coefficients attaining values of up to ±0.83. An interesting feature is that, although the LWT classification is based on the British Isles, some remote centres of variation still show associations, because they are related with distinct flow directions over the Isles. Winter PC4, for example, is a centre in the eastern Mediterranean (Fig. 1). Southwesterly flow over the British Isles, around an anticyclone over central/southern Europe on high score days (Fig. 3) is reflected in positive correlations with westerly and southwesterly LWT (Table 1). When a centre of variation is further west, as in the case of winter PC5, there is a significant correlation with southeasterly LWT (Table 1), due to flow around the northern flank of a cyclone situated to the southwest of the British Isles (Fig. 3).

4. RELATIONSHIPS BETWEEN PRINCIPAL COMPONENTS AND THE CENTRAL ENGLAND TEMPERATURE AND ENGLAND AND WALES RAINFALL SERIES

Another way of confirming the physical sense of the components is to examine their relationships with climatic variables. The datasets chosen for this analysis are the CET and EWR. The latter series is particularly pertinent because of the importance of precipitation as a deposition mechanism for acidic species. The CET was constructed on a monthly time scale by Manley (1974) for the period 1659 to 1973. This was updated to 1992 and constructed on a daily time scale by Parker et al. (1992). The EWR is also an areal-average of a network of sites

Table 2. Summer correlations between monthly mean component scores and monthly frequencies of the Lamb Weather Types (abbreviations explained in Table 1). Values in **bold** are statistically significant at the 5% level. n = 30

	A	C	NE	E	SE	S	SW	W	NW	N	AC	CC
PC1	**−0.67**	**0.58**	0.20	−0.01	0.24	0.05	−0.01	−0.03	−0.12	**0.47**	**−0.70**	**0.57**
PC2	**0.74**	**−0.59**	−0.17	0.45	−0.02	0.09	**−0.50**	**−0.40**	0.21	−0.28	**0.75**	**−0.62**
PC3	**−0.47**	**0.48**	−0.08	−0.13	−0.10	−0.06	0.21	0.18	−0.15	0.25	**−0.50**	**0.42**
PC4	0.07	−0.14	−0.31	−0.25	−0.07	0.03	−0.17	0.16	**0.45**	−0.19	0.13	−0.17
PC5	0.27	0.17	0.32	0.13	−0.08	−0.04	**−0.50**	**−0.72**	0.13	**0.46**	0.16	0.06
PC6	0.15	0.07	0.10	0.06	0.01	−0.28	−0.24	−0.11	0.19	−0.36	0.13	0.08
PC7	−0.25	0.31	0.10	0.04	**−0.39**	0.13	−0.16	0.02	−0.24	0.31	−0.24	0.27
PC8	0.14	−0.13	−0.14	−0.21	0.20	0.25	−0.26	0.17	**0.46**	−0.19	0.11	−0.21
PC9	**0.37**	**−0.57**	0.12	−0.35	**−0.48**	−0.08	−0.09	0.16	**0.51**	−0.05	**0.43**	**−0.48**
PC10	−0.12	0.11	0.30	0.04	−0.15	0.20	−0.25	−0.36	0.03	**0.47**	−0.15	0.09
PC11	−0.13	0.18	−0.35	0.14	0.20	−0.02	−0.04	0.08	−0.04	−0.02	−0.22	0.21
PC12	0.21	−0.09	−0.11	−0.15	−0.05	−0.13	−0.19	−0.13	0.10	0.02	0.19	−0.06
PC13	−0.27	**0.51**	−0.27	0.13	0.22	−0.15	−0.15	−0.22	0.10	−0.04	**−0.37**	**0.49**
PC14	0.03	−0.09	−0.27	−0.30	0.22	−0.02	−0.01	0.28	0.34	−0.10	0.02	−0.17
PC15	−0.15	0.13	0.31	−0.10	−0.04	0.16	0.03	−0.32	0.03	**0.56**	−0.25	0.19
PC16	0.14	−0.35	**−0.52**	−0.02	0.17	0.15	0.04	0.02	0.03	−0.05	0.30	**−0.40**

Table 3. Correlation coefficients between the observed components and the Central England Temperature (CET) and England and Wales Rainfall (EWR). Values in **bold** are statistically significant at the 5% level. n = number of cases

	Winter		Summer	
	CET	EWR	CET	EWR
PC1	**–0.47**	–0.31	**–0.52**	**0.68**
PC2	**–0.62**	**–0.40**	**0.56**	**–0.64**
PC3	–0.26	**–0.79**	–0.35	**0.47**
PC4	**0.57**	0.22	0.28	–0.24
PC5	**0.39**	0.22	–0.19	–0.09
PC6	–0.21	–0.16	0.10	–0.26
PC7	0.14	0.35	–0.27	0.12
PC8	–0.09	–0.25	0.08	–0.26
PC9	0.04	–0.03	0.28	**–0.60**
PC10	0.21	0.28	**–0.53**	0.01
PC11	**–0.49**	–0.26	0.01	0.20
PC12	0.11	–0.17	0.12	–0.01
PC13	**–0.47**	–0.21	–0.21	0.35
PC14	–0.22	–0.01	0.26	0.01
PC15			**–0.43**	0.34
PC16			**0.37**	–0.33
n	27	27	30	30

over England and Wales, and covers the period 1766 to present (Wigley & Jones 1987).

Correlation coefficients have been calculated between monthly mean component scores and monthly values for the CET and EWR. Table 3 shows the correlation coefficients for winter and summer. Significant positive (negative) values indicate low (high) temperature/precipitation amounts with high (low) scores. The relationships with CET in winter are physically sensible, reflecting the land/sea temperature contrast present in this season. Higher temperatures are related to zonal flow, because of the relatively warm Atlantic Ocean, whereas more meridional flow is associated with lower temperature. PC1, for example is negatively correlated with CET, indicating warmer temperatures with low scores (westerly flow) and cooler conditions with high scores (northerly/northeasterly flow) (Fig. 3). The negative relationships between CET and PC2, PC11 and PC13 (Table 3) identify the distinction between cool easterly flow from the relatively cold continent and warmer westerly flow from the Atlantic (Fig. 3). The components exhibiting significant correlations (both negative) with EWR in winter are PC2 and PC3 (Table 3). The composite pressure patterns for low loadings (i.e. the pressure patterns associated with higher precipitation) indicate westerly flow, and a cyclone, over the British Isles, for these 2 PCs, respectively (Fig. 3). Both of these synoptic situations are well known for the occurrence of precipitation in this region (Barrow & Hulme 1997).

During summer, higher temperatures under anticyclonic conditions are reflected in the negative relationship between CET and PC1, and the positive relationship with PC2 (Table 3). PC15, which is an Icelandic centre north of the British Isles (Fig. 2) is negatively correlated with CET (Table 3); high score days are associated with northerly flow, and thus with lower temperatures. Summer precipitation is also correlated with a number of components. The significant correlations with PC2, PC3 and PC9 are related to the location of cyclones. The negative correlation with PC2, for example, reflects the cyclone present during low score days. The positive correlation between PC3 and EWR, for example, reflects the more southerly storm track associated with high loadings (Fig. 4). On low score days of this PC, pressure is low in the northeast of the domain due to cyclones taking a more northerly path around high pressure extending over Europe. On high score days, pressure in the northeast is high, thus cyclones would be expected to take a more southerly path through the Baltic. This PC, as explained above, is positively correlated with cyclonic and cyclonic composite LWT. There is also a strong positive correlation between PC1 and EWR. The LWT analysis above revealed that high scores are associated with cyclonic LWT, as the composite map for low scores (Fig. 4) indicates that depressions would be steered on a more southerly track than under high score conditions.

These links between the PCs and CET and EWR values are adequately interpretable in physical terms, and so impart a reasonable level of confidence to embark on the next step: linkages with EMEP station observations.

5. RELATIONSHIPS BETWEEN PRINCIPAL COMPONENTS AND EMEP STATION DATA

The locations of the chosen stations are shown in Fig. 5: Eskdalemuir, Scotland (55° 19' N, 3° 12' W); Birkenes, Norway (58° 23' N, 8° 15' E); Karvatn, Norway (62° 47' N, 8° 53' E); La Hague, France (49° 37' N, 1° 50' W); and K-puszta, Hungary (46° 58' N, 19° 35' E). The aim was to choose sites with a wide geographical dispersion, although choice was limited by data availability, quality and completeness. A choice from southern European sites was not possible. An additional reason for the choice of the first 3 sites was to allow comparison of this method with previous work, as these were used in the study by Dorling (1992).

To assess the influence of precipitation amount and transport direction on air and precipitation chemistry at the sites, a correlation analysis was undertaken. Firstly, correlations were determined between compo-

nent scores and daily and monthly values of the aqueous and gaseous variables, using data for the period 1982 to 1991. The variables considered were aqueous non-sea salt sulphate ($SO_{4(aq)}$), aqueous ammonium ($NH_{4(aq)}$), aqueous nitrate ($NO_{3(aq)}$), sulphur dioxide gas ($SO_{2(g)}$) and sulphate aerosol ($SO_{4(a)}$), and also aqueous hydrogen ion ($H^+_{(aq)}$) for La Hague and K-puszta. Concentrations of aqueous and gaseous pollutants are inversely correlated with precipitation amount (Davies et al. 1992); in order to account for this, partial correlation analysis was undertaken, controlling for variations in the amount of precipitation.

Daily concentrations of acidic species in rainfall are known to be approximately log-normally distributed (RGAR 1990). To confirm this, when the analysis is stratified by season, and by month, the Lilliefor's version of the Kolmogorov-Smirnov test (von Storch & Zwiers 1999) was applied to the original data, and to log-transformed data. This test compares the goodness of fit to a hypothetical distribution, in this case the normal distribution, adjusted for use when the distributions of the data are not known and must be estimated from the data. The test confirmed that the daily concentration and deposition data were most definitely log-normally distributed (Jones 1997). For the monthly data, for each season, the majority of variables approximated more to a log-normal distribution than to a normal distribution, with the exception of a small number of cases (Jones 1997). Thus, most of the correlations were calculated using log-transformed data.

The daily data were corrected for autocorrelation by adjusting the sample size based on the method of Wigley (1983):

$$n = (\mathbf{n}-2) \cdot \frac{1-(\mathbf{CD} \cdot \mathbf{CF})}{1+(\mathbf{CD} \cdot \mathbf{CF})}$$

where $\mathbf{n}$ is the original number of points correlated, $\mathbf{CD}$ is the lag one autocorrelation coefficient for one series, and $\mathbf{CF}$ the same for the other, and n the corrected number of points. The monthly averaging of data removed the effect of serial correlation. To simplify the tables, the number of cases given for the daily data in Tables 4, 6, 8 & 10 is an average for all the PCs for that variable, as the autocorrelation of each PC is slightly different. However, the individual corrected number of cases was considered when marking significance levels.

Strong correlations of pollution data with PCs will occur only where the extremes of the components are associated with conditions which lead to differing impacts on pollutant concentrations. For example, where
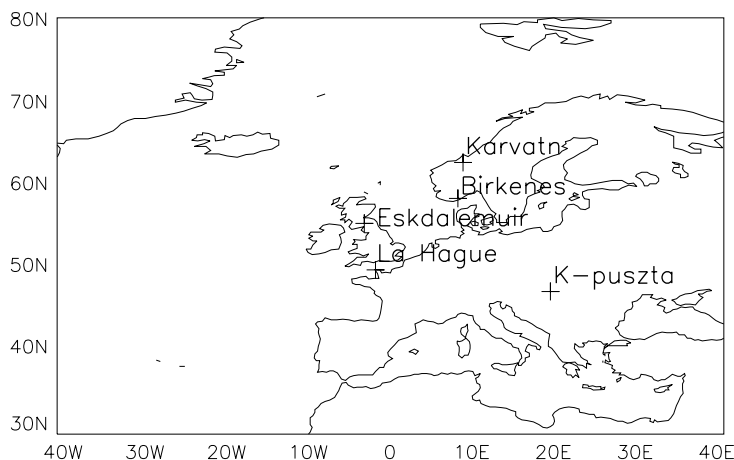

Fig. 5. Locations of the European Monitoring and Evaluation Programme (EMEP) stations used in this study

the flow direction for a particular component for both high score days and low score days comes from a strong pollution source in both cases, then a low correlation would be expected. Highest correlations would be expected when a component contrasts flow from 'clean' regions with flow from pollutant source regions, although other atmospheric conditions could also be important.

For each EMEP station, the characteristics of the pollution climate are described, and the relationships between the PCs and air and precipitation chemistry discussed for each season. A brief summary of the important relationships is also given. Due to space limitations, only winter results shall be discussed here. The results in the other 3 seasons are also robust, and reveal the same linkages between circulation and pollution climate at each of the stations (Jones 1997).

5.1. Eskdalemuir

Eskdalemuir is an upland station located in southern Scotland. Westerly flow from the North Atlantic therefore understandably exerts a strong influence on the pollution climate of the site. For example, Davies et al. (1992) examined the links between the zonal pressure gradient over the Northeast Atlantic and concentrations of aqueous sulphate, nitrate, ammonium and hydrogen ions over Europe. They detected a zone covering the UK, extending into northwestern Europe, where ion concentrations showed a strong inverse relationship with the pressure gradient. Transport from source regions of the southern UK and western Europe has also been found to affect concentrations and deposition at the site. Weakening of westerly airflow, and

therefore enhanced southerly and/or easterly transport, was found to be related to high monthly aqueous sulphate concentrations at Eskdalemuir (Farmer et al. 1987).

The majority of the partial correlation coefficients between the PCs and daily station concentration data for winter (Table 4) are consistent with the known links, described above, contrasting the polluted flow from the pollution source regions of central and eastern Europe with the cleaner flow from the North Atlantic. For example, the transport direction to Eskdalemuir on low score days for PC2, centred over northern Eurasia, is westerly, and on high score days it is easterly (Fig. 3). Correlation coefficients between PC2 and all the pollution variables are positive, implying increased concentrations with easterly flow from source regions of central Europe. Similarly, the positive correlations with PC1 (significant for only aqueous sulphate and nitrate) imply increased concentrations with weakened zonal flow.

An interesting relationship is the negative correlation between all variables and PC5, centred in the western Mediterranean (Fig. 1). It may appear that the centre is too far south to exert an influence on the pollution climate of Eskdalemuir, but the low score (high concentrations) composite map (Fig. 3) shows southeasterly flow to the UK around the eastern flank of low pressure centred off the northwest coast of Spain (Fig. 3).

A smaller number of significant relationships exist for the monthly correlations (Table 5). The positive relationship with PC2 remains for excess sulphate and nitrate concentrations (i.e. high concentrations with easterly flow).

5.2. Birkenes

Birkenes is located in southern Norway, to the southeast of the Scandinavian mountain chain. Thus when flow is from the west and northwest much moisture has been removed over the mountains before the air reaches the site. The southwest tends to be the direction associated with the most precipitation at the site (Dorling & Davies 1995), although easterly flow also brings precipitation. A frequent precipitation-bearing situation in autumn and winter is when the Atlantic Polar Front is displaced southwards, and a trough extends from Greenland to central Europe. The resulting southerly/southeasterly flow of Atlantic air to Scandinavia also picks up further moisture over the Baltic Sea, and is subjected to orographic uplifting when it reaches the Scandinavian mountains (Johannessen 1970). Due to the location of the site away from local pollution sources, long range transport exerts a large influence on the pollution climatology of the site. The UK and eastern and central Europe have all been identified as dominant source regions (Berge 1988, Amundsen et al. 1992).

The positive daily correlation coefficients between all variables and PC2 and PC9 (Table 6) show that southerly and southeasterly flow to the site, around the southern edge of the Eurasian winter anticyclone (Fig. 3), is related to high concentrations for all variables. The negative correlations between concentrations of the aqueous variables and PC13, centred over Iceland, imply that the UK is a source region of pollutants to the site, as the low score composite in Fig. 3 shows southwesterly flow from the UK, around low pressure to the west of Iceland. Likewise, southwest-

Table 4. Eskdalemuir winter daily partial correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases. Values adjusted for autocorrelation

	$\ln SO_{4(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	**0.10**	0.06	**0.14**	0.09	−0.02
PC2	**0.26**	**0.16**	**0.33**	**0.25**	**0.30**
PC3	**0.21**	**0.38**	**0.19**	0.09	**0.27**
PC4	−0.07	−0.03	−0.09	**−0.12**	0.05
PC5	**−0.13**	**−0.11**	**−0.20**	**−0.11**	**−0.17**
PC6	0.02	−0.03	−0.01	0.01	−0.02
PC7	0.02	0.00	−0.05	−0.04	−0.09
PC8	−0.02	0.01	**−0.11**	−0.07	**−0.21**
PC9	**0.14**	**0.14**	**0.12**	0.00	0.10
PC10	0.03	−0.02	0.06	0.02	0.03
PC11	**0.23**	**0.13**	**0.24**	**0.18**	**0.13**
PC12	**−0.13**	−0.07	**−0.18**	**−0.16**	**−0.17**
PC13	−0.02	−0.07	0.00	−0.06	−0.09
PC14	0.08	**0.11**	0.09	**0.14**	0.09
n	270	270	260	190	180

Table 5. Eskdalemuir winter monthly partial correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases

	$\ln SO_{4(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	0.22	0.06	0.10	0.26	−0.05
PC2	**0.40**	0.28	**0.49**	0.36	0.18
PC3	0.04	0.28	−0.01	−0.09	0.05
PC4	−0.31	−0.14	−0.24	−0.27	0.25
PC5	0.31	0.26	0.11	0.22	−0.24
PC6	0.13	−0.05	0.00	0.02	0.01
PC7	0.34	0.37	0.37	0.25	0.26
PC8	−0.04	−0.15	−0.22	0.09	**−0.51**
PC9	0.13	0.15	0.35	−0.01	0.21
PC10	−0.08	−0.09	−0.01	0.20	−0.10
PC11	0.27	−0.04	0.15	0.19	−0.08
PC12	−0.17	−0.13	−0.23	−0.05	−0.37
PC13	0.20	0.01	0.10	−0.09	0.04
PC14	0.14	0.18	0.19	0.22	−0.05
n	27	27	27	27	27

Table 6. Birkenes winter daily partial correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases. Values adjusted for autocorrelation

	$\ln SO_{4(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	0.01	−0.03	0.05	−0.07	**−0.25**
PC2	**0.49**	**0.35**	**0.45**	**0.42**	0.15
PC3	−0.02	0.05	−0.05	**−0.18**	−0.12
PC4	0.01	0.04	−0.03	0.01	**0.21**
PC5	**−0.19**	**−0.12**	**−0.21**	−0.08	0.03
PC6	0.01	0.03	0.05	0.07	−0.07
PC7	0.02	0.10	0.03	−0.04	0.00
PC8	−0.09	−0.03	**−0.11**	−0.02	−0.01
PC9	**0.22**	**0.22**	**0.23**	**0.19**	**0.23**
PC10	−0.05	−0.04	−0.06	0.02	−0.09
PC11	**0.20**	0.04	**0.24**	0.07	−0.08
PC12	0.00	−0.04	−0.07	0.02	−0.07
PC13	**−0.13**	**−0.16**	−0.06	−0.07	**−0.17**
PC14	**0.16**	**0.18**	**0.17**	0.05	**0.18**
n	240	230	240	175	155

erly flow around the southern edge of a cyclone centred to the north of Scotland brings southwesterly flow to Birkenes in the low score phase of PC3 (Fig. 3), and is related to high concentrations of gas and aerosol.

On a monthly basis, Table 7 shows that southeasterly flow associated with high scores for the Eurasian centres of variation, PC2 and PC9, is the dominant control for aqueous sulphate, nitrate and $SO_{2(g)}$ concentrations. The flow patterns leading to high sulphate aerosol concentrations are those with southwesterly flow from the UK. For example, PC4, in the eastern Mediterranean (Fig. 1), is positively correlated with both monthly and daily sulphate aerosol concentrations. This PC contrasts southwesterly flow to Norway around the northern edge of high pressure over central Europe (high

Table 7. Birkenes winter monthly partial correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases

	$\ln SO_{4(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	0.01	−0.06	0.09	0.01	**−0.51**
PC2	**0.57**	0.38	**0.54**	**0.47**	−0.22
PC3	−0.21	−0.07	−0.20	−0.02	0.27
PC4	0.04	0.15	−0.02	0.00	**0.48**
PC5	0.02	0.03	−0.19	−0.03	0.07
PC6	−0.07	−0.01	0.17	0.18	−0.33
PC7	0.23	0.28	0.33	0.11	−0.04
PC8	0.03	−0.06	−0.15	−0.10	0.01
PC9	0.30	0.23	**0.41**	**0.41**	0.06
PC10	0.02	0.06	−0.08	0.04	0.19
PC11	−0.17	−0.10	−0.14	0.07	−0.09
PC12	0.02	−0.05	−0.12	0.06	0.07
PC13	−0.29	−0.21	−0.07	0.05	0.02
PC14	−0.24	−0.28	−0.17	−0.21	0.31
n	24	24	24	24	24

scores and high concentrations) with northerly flow to the site (low scores and low concentrations). This is entirely consistent with an analysis of aerosol data at Birkenes by Pakkanen et al. (1996), who also identified the UK as an important source.

5.3 Karvatn

This station is the most remote of those considered in this study, situated on the west coast of Norway, to the northwest of the Scandinavian mountain chain.

Pollutants contained in flow from the southeast, an important source direction for the pollution climatology of Birkenes, are often removed when passing over the Scandinavian mountains (Dorling & Davies 1995). Consequently, Karvatn experiences the lowest concentrations for all variables of the sites considered in this study, although pollutant concentrations at the site may still be relatively high from this direction when compared with concentrations associated with other flow directions. For example, PC2 is positively correlated with all pollutant concentrations (not statistically significant for $SO_{2(g)}$) (Table 8), reflecting transport from major pollution source regions of central and eastern Europe. Gas and aerosol concentrations are positively related to PC12, a centre to the southwest of the UK (Fig. 1). The composite plot for high scores (Fig. 3) reveals a high pressure centre west of the Bay of Biscay, with a depression located over the North Sea, leading to southeasterly flow to Karvatn, without a long passage over land, which has a higher deposition velocity for gases and aerosols than does the sea surface. This circulation pattern was identified as being

Table 8. Karvatn winter daily partial concentration correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases. Values adjusted for autocorrelation

	$\ln SO_{4(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	**0.15**	**−0.12**	**−0.13**	−0.09	**0.17**
PC2	**0.20**	**0.13**	**0.24**	0.20	**0.33**
PC3	**−0.11**	0.03	**−0.20**	**−0.40**	**−0.19**
PC4	**−0.11**	0.06	0.04	−0.05	−0.06
PC5	−0.01	0.05	0.02	0.05	0.00
PC6	**0.14**	0.09	0.04	**0.18**	**0.17**
PC7	**−0.17**	0.05	0.00	0.00	−0.07
PC8	−0.03	−0.06	**−0.13**	0.08	0.04
PC9	−0.07	0.07	0.08	0.10	0.04
PC10	−0.08	0.11	0.01	0.07	0.03
PC11	0.02	−0.11	**−0.18**	0.00	**0.17**
PC12	0.10	0.01	0.02	**0.16**	**0.12**
PC13	**0.14**	−0.07	−0.03	−0.05	0.04
PC14	−0.01	0.06	−0.04	−0.11	**−0.11**
n	270	215	238	85	160

associated with above average pollutant concentrations at Karvatn by Dorling & Davies (1995).

Similarly, typical low score days for PC3 (Figs. 1 & 3), which was negatively related to all variables except aqueous ammonium, show southeasterly flow to Karvatn, around a cyclone to the north of the UK. Thus increased concentrations are still contained in flow from source regions arising from this circulation type, in comparison to northwest flow from regions with few pollution sources. The precise location of a station in relation to fronts within such a system will also influence both where wet deposition will occur and acidic concentrations therein (Haagenson et al. 1985), although this cannot be determined from the current analysis.

Different source regions are implicated as important for nitrogen species and sulphur species. Flow from the UK is related to aqueous ammonium and nitrate (negative correlation with PC1), and aqueous nitrate (negative correlation with PC11). Conversely, sulphate aerosol concentrations are positively correlated with PC11, and aqueous sulphate and sulphate aerosol positively with PC1, indicating increased concentrations with weakened westerly/strengthened easterly flow.

Significant monthly correlations occur only with aqueous sulphate and sulphate aerosol (Table 9). The stronger relationships with these variables may be due to the remote location of the station from major pollution sources. Sulphate aerosol is a secondary pollutant, hence concentrations increase with distance from sources, and those of the primary pollutant, sulphur dioxide gas, decrease (RGAR 1990). The ratio of sulphate to nitrate in precipitation has been found to

increase with increasing travel time of polluted air, as nitric acid is removed before sulphuric acid (Davies et al. 1988).

5.4. K-puszta

Pollutant concentrations are considerably higher at this site than the others, as it is located in a more densely populated and industrialised region. K-puszta is situated approximately 80 km southwest (downwind of the most common flow) of Budapest, the largest pollution source in the region. The city of Kecskemet is located 20 km from the site in a SSE direction, but there are significant local pollution sources in all directions. The region is also climatologically different from the western European sites. The station is located in the Hungarian Great Plain, in the Carpathian Basin, bounded to the northeast and east by the Carpathian Mountains, to the south by the Balkans, to the southwest by the Dinaric Alps and to the west by the Alps. This mountain barrier impedes flow of air into the region.

There is a positive daily correlation between concentrations of a number of pollution variables and PC2 and PC9 (Table 10), which represent northern and southern Eurasian centres of variation, respectively (Fig. 1). This implies southeasterly flow from Romania, Bulgaria and Yugoslavia to the site results in enhanced concentrations, as can be seen from the high score composites for these components shown in Fig. 3. PC2 also has significant monthly correlations with sulphate aerosol and sulphur dioxide gas (Table 11). Similarly,

Table 9. Karvatn monthly partial concentration correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases

	$\ln SO_{4(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	**0.52**	−0.04	0.12	−0.34	**0.47**
PC2	**0.54**	−0.35	0.05	−0.07	**0.67**
PC3	0.19	0.18	−0.12	−0.34	0.15
PC4	−0.34	0.21	0.24	−0.12	−0.29
PC5	−0.17	−0.08	0.17	0.12	−0.01
PC6	0.20	0.01	−0.15	0.14	**0.51**
PC7	−0.25	0.26	0.28	0.03	−0.22
PC8	−0.07	−0.33	−0.22	0.08	0.08
PC9	−0.35	−0.06	−0.16	0.11	−0.08
PC10	−0.19	−0.18	0.03	0.18	0.06
PC11	0.21	−0.22	−0.05	−0.08	0.18
PC12	−0.14	−0.31	−0.30	0.35	0.08
PC13	0.17	0.33	−0.18	−0.03	0.21
PC14	0.07	0.15	−0.11	−0.29	−0.17
n	27	27	27	27	27

Table 10. K-puszta winter daily partial correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases. Values adjusted for autocorrelation

	$\ln SO_{4(aq)}$	$\ln H^+_{(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	0.04	0.00	−0.23	**−0.28**	0.07	0.13
PC2	**0.24**	−0.03	−0.03	**0.27**	**0.28**	0.07
PC3	**0.18**	−0.01	0.11	0.09	**0.28**	0.14
PC4	−0.03	−0.04	**0.19**	**0.20**	−0.14	0.08
PC5	**−0.19**	−0.05	−0.06	−0.01	−0.11	−0.14
PC6	−0.06	−0.07	0.02	**0.18**	0.19	−0.06
PC7	0.09	0.01	0.07	−0.06	−0.04	−0.06
PC8	0.02	0.06	0.05	−0.04	−0.13	0.05
PC9	**0.18**	**0.47**	0.06	−0.10	0.10	0.12
PC10	**0.23**	−0.06	**0.20**	0.03	−0.02	0.04
PC11	**0.16**	0.17	**0.17**	0.09	0.11	0.01
PC12	**−0.17**	−0.03	0.09	−0.13	−0.08	0.08
PC13	0.09	−0.18	−0.07	−0.12	0.01	−0.04
PC14	**0.18**	−0.06	−0.11	−0.03	0.05	0.12
n	110	50	110	115	50	47

Table 11. K-puszta winter monthly partial correlations. Values in **bold** are statistically significant at the 5% level. n = number of cases

	$\ln SO_{4(aq)}$	$\ln H^+_{(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	0.27	−0.28	−0.27	0.12	**0.42**	**0.42**
PC2	0.25	−0.05	0.01	0.13	**0.49**	**0.51**
PC3	0.21	−0.23	−0.14	0.00	0.36	**0.49**
PC4	**−0.40**	−0.28	0.03	−0.13	−0.27	−0.26
PC5	0.16	0.26	0.36	0.34	−0.25	−0.24
PC6	−0.14	−0.31	−0.29	−0.11	**0.48**	0.01
PC7	0.07	0.13	0.05	0.11	−0.13	−0.14
PC8	0.30	0.37	0.25	0.31	−0.39	0.23
PC9	0.22	0.38	0.20	−0.15	−0.05	−0.06
PC10	−0.10	0.03	−0.07	−0.29	−0.17	−0.04
PC11	0.18	0.36	0.06	0.09	0.29	0.14
PC12	0.26	**0.51**	0.21	0.12	−0.28	−0.01
PC13	−0.09	−0.23	**−0.59**	−0.29	**0.51**	0.17
PC14	0.19	−0.17	−0.11	0.18	0.08	0.13
n	23	15	23	23	22	22

Table 12. La Hague winter daily partial correlation coefficients. Values in **bold** are statistically significant at the 5% level. n = number of cases. Values adjusted for autocorrelation

	$\ln H^+_{(aq)}$	$\ln NH_{4(aq)}$	$\ln NO_{3(aq)}$	$\ln SO_{2(g)}$	$\ln SO_{4(a)}$
PC1	**0.20**	0.09	**0.22**	0.09	0.08
PC2	**0.17**	**0.19**	**0.25**	**0.21**	**0.39**
PC3	**0.19**	**0.50**	**0.43**	0.18	**0.28**
PC4	0.11	0.12	0.04	**−0.23**	−0.12
PC5	**−0.31**	**−0.25**	**−0.34**	**−0.24**	−0.16
PC6	0.00	0.08	−0.08	−0.09	−0.12
PC7	**−0.18**	**−0.25**	**−0.18**	**−0.25**	**−0.25**
PC8	−0.04	**−0.12**	−0.03	0.09	0.03
PC9	**0.16**	0.08	0.12	−0.06	0.05
PC10	−0.03	0.10	−0.04	0.09	0.05
PC11	**0.13**	0.11	0.08	0.13	0.08
PC12	0.02	−0.08	−0.05	**−0.19**	**−0.24**
PC13	0.08	0.06	0.03	0.09	0.07
PC14	0.00	0.11	−0.02	0.06	0.02
n	160	170	160	70	72

the monthly positive relationship between PC3 and sulphate aerosol (Table 11), and the daily positive relationship between this component and aqueous sulphate and sulphur dioxide gas (Table 10), signifies increased pollutant concentrations with transport from the northeast, from Russia, Poland and the Czech Republic (Figs. 1 & 3). The physical interpretation behind other statistical associations is not readily apparent; this is probably due to the frequent high pressure and slack pressure gradients over the region (Jones 1997). For example, PC11, centred north of the UK, shows positive daily correlations with aqueous sulphate and ammonium, yet there appears little difference in the pressure patterns around Hungary for high and low score composites (Fig. 3).

5.5. La Hague

The only possible pollution sources to the west of this station are the Channel Islands and, to the southwest, Brittany. No data are available for aqueous sulphate concentrations for this site, due to problems of measurements being highly influenced by sea salt concentrations in precipitation (J. Schaug pers. comm.). A change of laboratory occurred in 1989, and as discontinuities in the data were apparent at this point, only data from pre-1989 were used in this analysis. La Hague has the highest concentrations of all the sites, except for K-puszta.

Table 12 indicates that, in common with the other stations, southeasterly flow around the Eurasian winter anticyclone (PC2) (Figs. 1 & 3) leads to high concentrations of all pollution variables. Other important condi-

tions are southerly flow due to low pressure off the northwest coast of Spain (PC5; Figs. 1 & 3), and northeasterly flow/anticyclonic cover, due to the eastern edge of an anticyclone centred over the UK (PC3; Figs. 1 & 3). As at Eskdalemuir, sulphur dioxide gas is not significantly related to this component, possibly due to the proximity of both stations to the centre of an anticyclone, thus perhaps with higher reaction rates leading to fast transformation to aqueous and aerosol species.

Two surprising relationships are the negative correlation between PC7 and all pollution variables, and that between PC12 and gas and aerosol data. The low score composites for both (Fig. 3) show westerly and southwesterly flow to the site, a direction which might be expected to be associated with low concentrations.

There are few significant monthly relationships (Table 13). PC2 is related to aqueous hydrogen ion and sulphate aerosol (enhanced southeasterly flow; see above). The relatively strong positive relationship between aqueous nitrate concentrations and PC14 (Fig. 1) is, again, rather surprising since the composite map (Fig. 3) implies the southwest to be a source direction.

6. DISCUSSION AND CONCLUSIONS

The work presented in this paper represents the first stage of the development of pollution scenarios for 5 European stations using a climate downscaling methodology. It is necessary firstly to determine the main modes of climate variability over the region under consideration. This has been accomplished using PCA of sea level pressure, for the period 1982 to 1991. The

Table 13. La Hague winter monthly partial correlation coeffi-
cients. Values in **bold** are statistically significant at the 5%
level. n = number of cases.

	$lnH^+_{(aq)}$	$lnNH_{4(aq)}$	$lnNO_{3(aq)}$	$lnSO_{2(g)}$	$lnSO_{4(a)}$
PC1	0.37	0.13	0.19	−0.03	−0.19
PC2	**0.56**	0.15	0.18	0.14	**0.59**
PC3	−0.25	0.23	0.22	0.25	0.24
PC4	−0.08	0.12	0.08	−0.01	−0.03
PC5	−0.33	−0.19	−0.12	−0.05	−0.03
PC6	−0.07	−0.16	−0.44	−0.33	−0.42
PC7	0.20	−0.35	−0.28	0.05	−0.06
PC8	−0.11	−0.46	−0.20	0.20	0.20
PC9	0.39	0.12	0.03	−0.28	−0.21
PC10	−0.15	0.35	0.24	0.18	−0.12
PC11	0.08	0.27	0.36	0.22	−0.11
PC12	−0.27	−0.24	−0.25	−0.24	0.00
PC13	−0.07	−0.01	−0.06	−0.09	−0.22
PC14	−0.12	0.22	**0.48**	0.40	0.12
n	16	16	16	16	16

construction of composite pressure plots of days with
high and low scores for each component allowed in-
vestigation of the associated mean pressure patterns.
The components were found to adequately represent
the regional-scale circulation, since they were inter-
pretable in terms of the known broad-scale circulation
features. For example, the first component in all sea-
sons represented the strength of the zonal circulation.
Comparisons of the PCA classification with the estab-
lished manual classification of the LWT, and associa-
tions with the 2 climate series of CET and EWR, were
physically reasonable. This gave confidence that the
PCA classification was an acceptable representation of
the regional-scale atmospheric circulation patterns.

The second stage of the analysis was to determine
the nature of the relationships between the PCs and
the adopted pollution variables at the 5 EMEP monitor-
ing stations. This work confirms the versatility of syn-
optic climatological methods for the study of pollution,
in general, since the links with the PCs were consistent
with those in earlier studies and, indeed, gave further
insight into the climatological influences on air and
precipitation composition at the stations. It should be
borne in mind that the pollution climate at a particular
station is a function of an assemblage of atmospheric
circulations, and so when the combined effect is con-
sidered, then the combined level of explanation can be
high. Naturally, synoptic climatology will never cap-
ture all aspects of transport, transformation and depo-
sition. Reasons for this include, for example, that trans-
formation will partially depend on the particular mix
of emissions, affecting factors such as oxidant avail-
ability. This in turn may influence factors such as
aerosol size, rate of transformation to secondary spe-
cies, and thus transport.

Regressions have been derived using combinations
of PCs, pollution concentrations and precipitation, and
the resultant explained variance is up to 80% (Jones
1997). These relationships are stronger for monthly
data, at which time scale the regressions were derived.
The UK and central and eastern Europe were found to
be pollutant sources for both the Norwegian sites and
La Hague. Sources to Eskdalemuir are England and
central and eastern Europe. The relationships are less
easily interpretable for the Hungarian station, K-
puszta. This is partially due to its less peripheral loca-
tion, being surrounded by pollution sources. This also
highlights a drawback of this methodology: particu-
larly in regions of the domain frequently under the
influence of weak pressure gradients, it is relatively
difficult to discriminate clear relationships between
pollution and particular circulation characteristics. The
use of airmass trajectories is a method frequently used
to investigate the influence of atmospheric transport on
air and precipitation, and this can allow the identifica-
tion of more definite transport directions (Moody &
Sampson 1989, Dorling et al. 1992). However, weather
type or PCA classifications do embrace a much wider
selection of the atmospheric conditions which ulti-
mately influence ambient pollution levels (Davies et al.
1991). Whatever classification method is used, a prob-
lem associated with using any one particular atmos-
pheric level to determine transport directions is that
wind shear and vertical motion cause variations in flow
at different atmospheric levels, particularly in the pres-
ence of fronts and during precipitation (Kahl 1993).
Although results have only been shown for winter
here, those for the other 3 seasons show very similar
links between particular circulation types and pollu-
tion concentrations at the 5 stations (Jones 1997).

An advantage of the method using PCA to investi-
gate links between circulation and air and precipita-
tion chemistry is that the synoptic situation causing
a particular flow direction is usually immediately
identifiable (with the described occasional exceptions
related to anticyclonic conditions in the case of K-
puszta, in the south of the domain). The influence of
pressure variations in the Mediterranean on the pollu-
tion climatology of stations as far away as Scandinavia,
for example, is one relationship that may not have
been expected. This may possibly not have been iden-
tified from a smaller domain size which did not include
the Mediterranean.

These PC/pollutant relationships have been used to
estimate future deposition using regression relation-
ships (Jones 1997, Jones & Davies 1998), and global
climate model data from the transient integration of the
United Kingdom Meteorological Office coupled ocean-
atmosphere model (UKTR), and will be described in
full in a forthcoming paper (Jones & Davies unpubl.).

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