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The influence of weather-type and long-range transport on airborne particle concentrations in Edinburgh, UK

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Abstract

This study investigated the influence of regional-scale synoptic weather type and geographical source regions of air masses on two-particle concentration metrics (Black Smoke (BS) and PM₁₀) in the city of Edinburgh, UK, between 1981 and 1996. Twenty-seven classifications of Jenkinson Daily Weather Types (JWT) were subdivided into 9 directional categories and 3 vorticity categories, and the influence of JWT category on BS and PM₁₀ determined. Four-day air mass back-trajectories for 1 July 1995–30 June 1996 were computed and grouped into 8 categories depending on the geographical route followed. Significantly elevated concentrations of BS (median values 2, 5 and 4 $\mu\text{g m}^{-3}$ greater than median for 1981–1996) and PM₁₀ (median values 3, 5.5 and 8 $\mu\text{g m}^{-3}$ greater than median for 1992–1996) were observed for anticyclonic, southerly and south-easterly weather types, respectively. These differences were not identified at conventional levels of significance for BS in 1995–1996. This may reflect a shift in more recent times to lower concentrations of predominantly locally emitted BS less affected by regional scale meteorology. Conversely, significant inter-trajectory category differences were observed for PM₁₀ during 1995–1996, with highest concentrations associated with Eastern European trajectories and south-easterly weather type categories (11.4 and 10.7 $\mu\text{g m}^{-3}$ greater than annual means, respectively). The variation in particle concentration across weather-type was a significant proportion of total median particle concentration, and of a magnitude associated with adverse health outcomes. Thus current PM₁₀ concentrations (and associated health outcomes) in Edinburgh are likely to be significantly influenced by regional-scale meteorology independent of local air quality management areas. Furthermore, changes in long-term trends in distributions of synoptic weather types indicate that future climate change may influence exposure to PM₁₀ and the PM₁₀:BS ratio in Edinburgh. Further definition of the relationships between long-range transport and particle concentration will improve classification of human exposure in epidemiological studies.

Keywords: Air mass trajectories; Synoptic weather types; Black smoke; PM₁₀

1. Introduction

Epidemiological studies have consistently shown an association between ambient concentrations of particulate matter pollution and adverse effects on human health (COMEAP, 1995; Pope and Dockery, 1999). In the UK, research on particulate matter formerly focused on the black smoke (BS) measure, but has latterly used PM10 (a particle distribution with median aerodynamic diameter $<10\ \mu\text{m}$). PM10 is currently perceived to be the best practical measure of particulate matter most likely to cause ill health, although concerns have also been expressed over smaller size fractions, PM2.5 and ultrafine ($<100\ \text{nm}$) particles (Donaldson et al., 1999). In a meta-analysis of published work from around the world, Pope (2000) suggests that for each $10\ \mu\text{g m}^{-3}$ increase in concentration of PM10, there is an associated approximate increase of 1% in daily all-cause mortality, with possibly higher associations for cardiovascular and respiratory mortality. Particulate air pollution has also been shown to be associated with increased morbidity for both respiratory and cardiovascular disease (Pope and Dockery, 1999). Epidemiological work in the city of Edinburgh (UK) supports these findings. Prescott et al. (1998) report a significant association between BS concentrations in Edinburgh from 1981 to 1995 (as a mean of the previous 3 days) and daily respiratory mortality in those aged 65 or over. They also report that increments of PM10 concentration in Edinburgh (as a previous 3-day mean) are associated with cardiovascular admission in elderly (>65 years) inhabitants. In addition to time series studies, a limited number of cohort studies (Dockery et al., 1993; Abbey et al., 1999) also show definite associations between health outcomes and particulate air pollution (relative mortality risk ratios of approximately 1.2 for PM2.5 pollutant ranges of approximately $20\ \mu\text{g m}^{-3}$) after attempts to control for individual risk factors.

In considering the epidemiological and toxicological evidence, the UK Expert Panel on Air Quality Standards (EPAQS), recommended an air quality standard for PM10 of $50\ \mu\text{g m}^{-3}$ as a 24 h mean (EPAQS, 1995). The UK government has since set an objective that 24 h mean PM10 concentration should not exceed $50\ \mu\text{g m}^{-3}$ more than 35 times a year by 2004, and a requirement that local authorities assess and review air quality in their area, and if necessary, set-up air quality management areas (AQMA) to reduce emissions of harmful pollutants (DETR, 2000). The efficacy of AQMAs is based on the assumption that elevated concentrations of PM10 in urban areas are mainly the result of local traffic-related emissions of primary particulate material. However, although local sources are important, rural levels of PM10 often become elevated during urban pollution episodes, suggesting that secondary particles make an important contribution to pollution concentrations at these times (King and Dorling, 1997; Salmon and Stedman). Furthermore, some pollution episodes in the UK, are associated with easterly winds, suggesting that a significant proportion of airborne PM10 responsible for exceeding air quality standards may originate from industrialised parts of mainland Europe (Stedman, 1998; King and Dorling, 1997). In addition, meteorological records have shown gradual changes in the global climate with time (IPCC, 2001). If future predictions regarding global climate change are correct, these changes are likely to become more pronounced resulting, inter alia, in alterations in rainfall and wind patterns, i.e. in changes to synoptic-scale meteorology (IPCC, 2001). The long-range transport of pollutants is significantly influenced by synoptic scale meteorological patterns. So if climate effects change the frequency of

these synoptic patterns, the frequency of pollution episodes occurring in the UK is also likely to alter (Dorling et al., 1992; Dorling and Davies, 1995).

All the above factors imply that the effectiveness of some proposed AQMAs might be compromised, as local authorities have little control over pollution from outside their area. Therefore, to determine whether targets within the NAQS are achievable, as well as the effectiveness of the proposed AQMAs, it is important to quantify the influence of long-range transport of particles on PM10 concentrations in the UK. In this study we investigated the influence of synoptic weather type on daily BS and PM10 data for Edinburgh for the periods 1981–1996 and 1992–1996, respectively. Air mass back trajectories were also computed for 1 July 1995–30 June 1996 and particle metrics classified according to source region. Finally, the influence of climate change on weather type frequency was investigated using synoptic weather type data from 1881 to 2000 as an indicator of the future influence of long-range transport for particle pollution in Edinburgh.

2. Methodology

2.1. Pollution data

The PM10 and BS monitoring sites were located within 1 km of each other central Edinburgh (Johnson Terrace and Princes Street Gardens, respectively). Both sites were free from the direct influence of local point sources and were therefore representative of background boundary layer air in central Edinburgh. The Johnson Terrace site was classified as a ‘commercial’ area (NETCEN, 2002), and had the longest near-continuous time series of BS data available in central Edinburgh. The Princes Street Gardens site was located in city-centre public parkland, and was classified as ‘urban centre’ (NETCEN, 2002). Data collected at both sites was subject to stringent quality control procedures specified by DEFRA and its predecessor government departments (NETCEN, 2002).

Three separate time periods of data were used in the present study (abbreviations for these periods are defined in Table 1, TS denotes Time Series). The dates TS (81–96) and TS (92–96) coincide with the availability in Edinburgh of BS and PM10 data, respectively. TS (95–96) was chosen as an annual period over which to compute back trajectories since it was known to contain major episodes of particulate pollution (King and Dorling, 1997; Stedman, 1998). All particle concentration data were obtained from the UK National Air Quality Information Archive (NETCEN, 2002).

Abbreviation	Time period	Data available ^a
TS (81–96)	1 January 1981–6 November 1996	BS, JWT
TS (92–96)	1 October 1992–6 November 1996	BS ^b , PM ₁₀ , JWT
TS (95–96)	1 July 1995–30 June 1996	BS ^b , PM ₁₀ , JWT, Back trajectories

Table 1. Abbreviations for the time periods investigated in this study and the corresponding available data

^aBS=Black smoke; JWT=Jenkinson weather type. ^bBS data was studied over this time period in addition to period TS (81–96) to allow direct comparison with PM10 measurements recorded over equivalent time periods.

Prior to the mid-1980s, BS was the only method of determining concentrations of particulate matter in the UK (APEG, 1999). The reflectance (the inverse of the darkness) of the stain on the filter paper through which air has been drawn is converted to gravimetric units using the British Standard smoke stain calibration (DETR, 1997). The calibration was determined in the 1960s when smoke from the combustion of coal was the predominant source of airborne particles. Large changes to the present day sources of PM means there is now no simple relationship between BS reflectance and total PM concentration (APEG, 1999). However, since the component of PM predominantly contributing to contemporary BS "darkness" are fine carbonaceous particles from traffic sources (in particular from diesel engines), BS measurements remain a useful indicator of the contribution to PM from local primary combustion sources (Stedman, 1998; Heal et al., 2000).

The concentration of PM₁₀ is measured in Edinburgh using a Tapered Element Oscillating Microbalance (TEOM) instrument. All TEOM data presented in this paper was corrected by a factor of 1.3 to allow for loss of volatile material in the heated inlet of the device (APEG, 1999). To allow direct comparison with the BS data, PM₁₀ data until 6 November 1996 were used, the date on which the Johnson Terrace BS site was relocated.

2.2. Jenkinson weather types

A method for classifying the daily circulation patterns over the British Isles (50°–60°N, 2°E–10°W) was originally devised by Lamb (1972). His subjective classification used surface level synoptic charts and, latterly, charts describing the flow at the 500 hPa level in the atmosphere, to indicate the steering of the circulation systems from day to day. This "Lamb Weather type" classification contains eight main directional types: north, north-east, east, south-east, south, south-west, west and north-west and three main non-directional types: anticyclonic, cyclonic and an unclassified type, which represents days when patterns are weaker or chaotic (Hulme and Barrow, 1997). Nineteen hybrid types (i.e. those satisfying the definitions of two or more main types) are also recognised (Lamb, 1972).

An objective alternative to the original Lamb catalogue was developed by Jenkinson and Collinson (1977) to eliminate the dependency on the consistency of an experienced analyst. The Jenkinson classification uses an automated procedure based on daily grid-point mean sea-level pressure data over the British Isles to classify weather types into the same 27 categories recognised by Lamb. Close agreement is found between the objective and subjective schemes (Jones et al., 1993).

In this work, Jenkinson objective Daily Synoptic indices from 1 January 1880 to 31 January 2001 were obtained from the Climate Research Unit at the University of East Anglia (Salmon, 1998). It was difficult to establish criteria for dividing the 27 pure and hybrid weather types into a smaller number of main groups that could be compared against each other. Lamb (1972) has indicated that, for statistical purposes, hybrid types (i.e. days when two, or occasionally three types of main definitions are satisfied) should count equally to each of their main types. Although this sub-division is useful when analyzing the number of days associated with individual main weather types, it is not a valid means of dividing the types when considering pollution data, since pollution concentrations recorded for individual days cannot be divided between

various main types without artificially increasing the number of pollution days in the dataset.

Therefore, in this study, the Jenkinson weather types (Jenkinson Daily Weather Types (JWT)) were most appropriately grouped on the basis of either direction, coded as JWT(d), or vorticity, coded as JWT(v) (Table 2). Nine directional categories were created: anticyclonic/cyclonic(d); north-easterly; easterly; south-easterly; southerly; south-westerly; westerly; north-westerly and northerly. All directional categories, with the exception of the anticyclonic/cyclonic(d) group, encompass the main type and the anticyclonic and cyclonic hybrid types (Table 2). Three vorticity categories were created: anticyclonic(v), directional(v) and cyclonic(v). Again, all vorticity categories incorporated both main and directional hybrid types (Table 2). Unclassified days were categorised separately in both cases. The unclassified category applies only to about 5% or less of the days.

Anticyclonic(v)		Directional(v)		Cyclonic(v)		
0	Anticyclonic			20	Cyclonic	A/C(d)
1	ANE	11	NE	21	CNE	NE
2	AE	12	E	22	CE	E
3	ASE	13	SE	23	CSE	SE
4	AS	14	S	24	CS	S
5	ASW	15	SW	25	CSW	SW
6	AW	16	W	26	CW	W
7	ANW	17	NW	27	CNW	NW
8	AN	18	N	28	CN	N

Table 2. Sub-division of Jenkinson weather types (JWT) into categories based on direction and vorticity. Division on the basis of direction (JWT(d)) ——. Division on the basis of vorticity (JWT(v))— — —.

2.3. Air mass back trajectories

Air mass back trajectories provide a useful means of establishing source–receptor relationships of air pollutants (Stohl, 1998; Beverland et al., 2000). In this study, 366 4-day trajectories with a central Edinburgh arrival point (55.95°N, 3.22°W) were computed for the period TS (95–96) using the European Centre for Medium Range Weather Forecasting Reanalysis Project at the British Atmospheric Data Centre (BADC, 2000). Daily trajectories were calculated for an arrival pressure of 950 hPa, and an arrival time of 1200 GMT, the midpoint of the time periods used to collect pollution data. Three-dimensional wind fields were used in the computation of

trajectories to allow for vertical air movements of air masses in addition to horizontal displacement.

The choice of a 4-day trajectory is a compromise between the several-day atmospheric residence time of fine particles and the declining accuracy in calculation of the back trajectory (due to model assumptions and spatial and temporal resolution of the meteorological data) (Stohl, 1998). Any error associated with a single trajectory is reduced when daily trajectories are grouped together according to the common path the air masses followed (Stohl, 1998). Therefore, the 366 daily trajectories were assigned to eight categories (Fig. 1) using the criteria in Table 3: United Kingdom (UK); Atlantic (AT); Arctic (AR); Western Europe (WE); Scandinavia (SC); Maritime (MT), Eastern Europe (EE) and Unclassified (UC).

Category	Description of trajectory categories
UK	Trajectory stagnating over, or spending most of 4-day period in the vicinity of the UK, i.e. within 50–60°N and 10°W–2°E
AT	Trajectory spending most of 4-day period crossing the Atlantic Ocean (including those passing to the west of Iceland)
AR	Trajectory spending most of 4-day period in the region 60–70°N and 10°N–10°E, including those originating in or near Greenland
WE	Trajectory originating in or spending most of 4-day period crossing Western Europe, i.e. area west of the Netherlands
SC	Trajectory originating in or spending most of 4-day period crossing Scandinavia or Northern Baltic areas
MT	Trajectory spending most of 4-day period crossing Maritime regions close to the UK and then crossing England en route to Edinburgh
EE	Trajectory originating in or spending most 4-day period crossing Eastern Europe, i.e. area east of the Netherlands
UC	Trajectory unable to be unambiguously classified into any <i>one</i> trajectory category

Table 3. Criteria used to classify air mass back trajectory atmospheric transport patterns for TS (95–96)

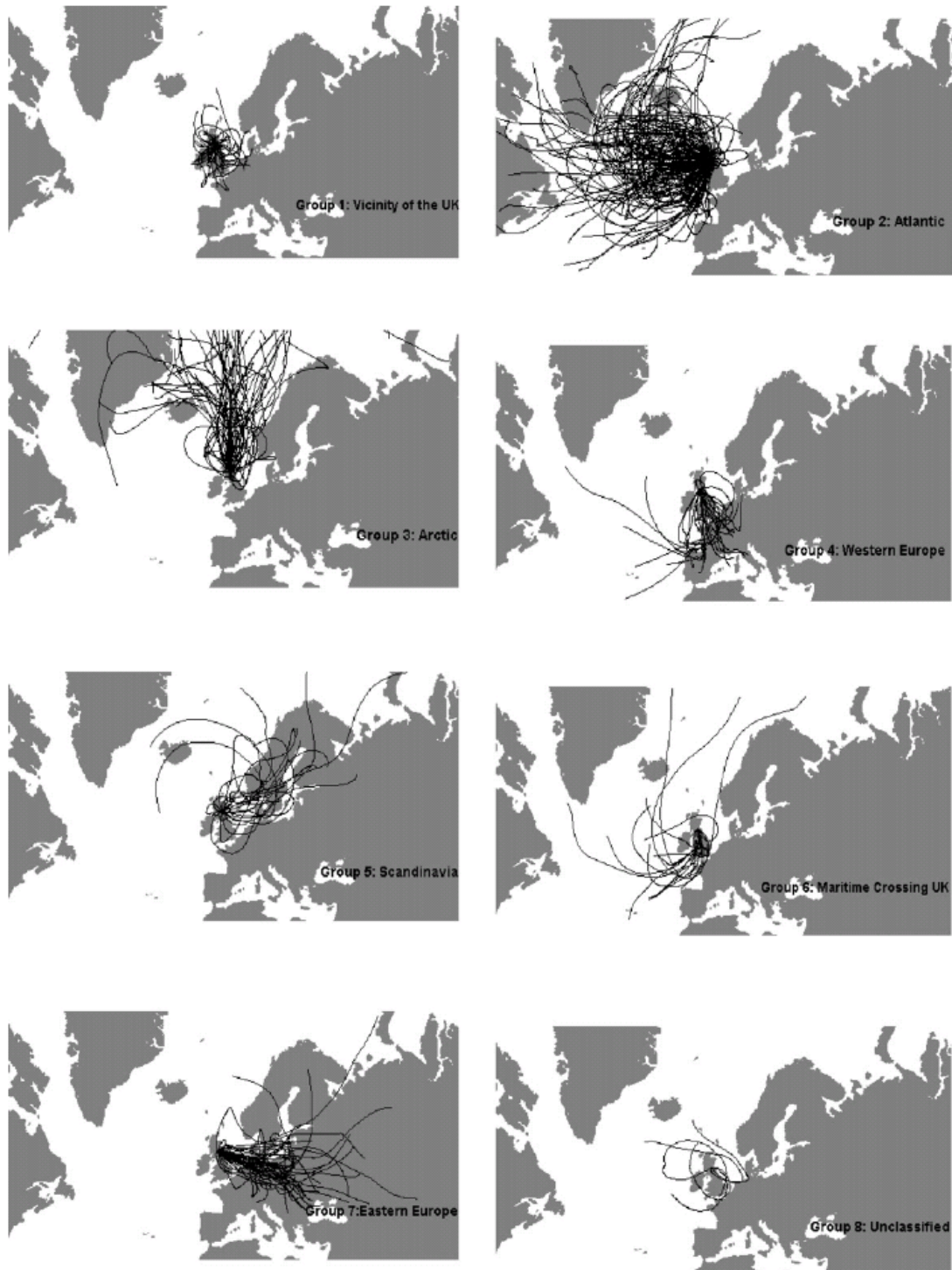


Fig. 1. Four-day back trajectories arriving in Edinburgh at 1200 GMT and 950 hPa for study period TS (95–96). Back trajectories were classified according to the criteria in Table 3.

2.4. Statistical analyses

The Kolmogorov–Sminov test confirmed that both raw and log-transformed datasets were non-normally distributed, so non-parametric tests were used in all analyses. The Kruskal–Wallis (K–W) test was used to test for the presence of significant differences in median BS and PM10 concentrations categorised by weather types (JWT(d) and JWT(v)), and by trajectory category for TS (95–96). Where the K–W test was significant, Mann–Whitney multiple comparison tests were applied to identify those categories differing significantly from others. A Bonferroni correction was applied to the critical value in each test to control for the increased probability of Type I errors (rejection of a true null hypothesis) caused by multiple testing. (The Bonferroni-corrected critical value corresponds to $p=0.05/(0.5k(k-1))$, where k is the number of categories compared). Since this approach increases Type II errors (retention of a false null hypothesis), significance was also assessed at the standard pairwise critical value (corresponding to $p=0.05$), ignoring the issue of multiple testing. These two extremes should bracket the correct statistical inferences (Levy et al., 2000).

3. Results and discussion

3.1. Summary statistics of pollution data sub-divided by all weather types

BS and PM10 concentrations, categorised according to the twenty-seven JWT, were examined over the full periods for which data were available (TS (81–96) and TS (92–96), respectively). The daily BS and PM10 observations discussed here represented 93% and 90% of the total numbers of days during TS (81–96) and TS (92–96), respectively. There was a larger proportion of missing BS data (24% missing) in TS (95–96). (The missing data were during the periods 20 July 1995–11 October 1995 and 21–26 December 1995 and did not affect our conclusions on long-range transport episodes which mostly occurred in early 1996.) Missing data were assumed to be the result of equipment malfunction or non-compliance of data with DEFRA quality control criteria (NETCEN, 2002).

Median BS concentrations for each weather type ranged from 5 to 14.5 $\mu\text{g m}^{-3}$ for the westerly and cyclonic southerly types, respectively, and median PM10 concentrations ranged from 14 to 29 $\mu\text{g m}^{-3}$ for the south-westerly and cyclonic easterly types, respectively. Since summary data for the two pollution metrics were computed for different time periods, they are not directly comparable, but the observation that highest concentrations were associated with different weather types supports the assertion that BS and PM10 may be affected by synoptic meteorology in different ways.

The particle concentrations observed in Edinburgh are considerably lower than in some of the larger conurbations in England. For example, mean PM10 concentrations in Edinburgh between 1993 and 1996 were 84% and 72% of mean concentrations observed in equivalent urban centre locations in Birmingham Centre and London Bloomsbury, respectively (APEG, 1999; QUARG, 1996). Hence the contribution of long-range transport of particles from European sources is likely to represent a higher proportion of the overall PM10 amount in Edinburgh.

3.2. Influence of Jenkinson weather type on particle concentration

Descriptive statistics for BS and PM₁₀ divided by JWT(v) and JWT(d) categories are shown in Table 4 and Table 5 for the three time periods considered. Note that the highest concentrations of BS were associated with unclassified days, for both the JWT(v) and JWT(d) categories, for all periods tested (Table 4 and Table 5). However, this is a consequence of the excessive influence of a small number of high pollution days, which may be sampling or contamination artifacts. These unclassified days were excluded from further analyses. The 5 unclassified back trajectories out of the 366 within the TS (95–96) period were also excluded from the analyses presented here.

	BS				PM ₁₀			
	<i>n</i>	Mean	Median	10–90 percentile	<i>n</i>	Mean	Median	10–90 percentile
TS (81–96)								
Anticyclonic(v)	1794	13.6	10	3.0–28.0				
Directional(v)	2259	10.1	7	2.0–20.0				
Cyclonic(v)	1270	12.5	9	3.0–25.0				
Unclassified days	61	16.0	12	4.0–34.0				
All	5384	11.9	8	2.0–35.0				
TS (92–96)								
Anticyclonic(v)	448	9.0	7	2.0–17.0	449	22.8	21	13.0–35.0
Directional(v)	599	7.3	5	2.0–14.0	562	19.3	17	11.0–30.9
Cyclonic(v)	331	8.5	7	3.0–16.0	326	19.7	18	12.0–29.0
Unclassified days	14	10.4	9.5	3.0–15.0	12	20.7	20	14.0–24.9
All	1392	8.2	7	2.0–15.0	1349	20.6	18	12.0–32.0
TS (95–96)								
Anticyclonic(v))	98	8.0	7	2.0–14.0	137	23.2	20	13.0–37.4
Directional(v)	125	6.7	5	2.0–12.0	146	20.0	17	11.0–33.0
Cyclonic(v)	51	6.9	5	2.0–12.0	75	20.8	17	12.4–30.0
Unclassified days	2	13.5	13.5	12.3–14.7	2	20.0	20	20.0–20.0
All	276	7.2	6	2.0–12.0	360	21.4	19	12.0–35.1

Table 4. Descriptive statistics for BS and PM₁₀ concentrations ($\mu\text{g m}^{-3}$) by JWT(v) categories

In all categorisations (by vorticity, direction, or back-trajectory), median PM₁₀ concentrations were much higher than those of BS (typically by a factor 2–3), confirming that BS is a sub-set of PM₁₀ as expected from the estimated size-fractionating characteristic of the BS sampler ($\approx\text{PM}_{4.4}$ (COMEAP, 1995)).

3.2.1. Influences on BS concentrations

Kruskal–Wallis, K–W tests showed that there were highly significant differences ($p < 0.001$ in all cases) in median BS concentration across both JWT(v) and JWT(d) categories, for both TS (81–96) and TS (92–96) periods. Highest median BS concentrations were consistently associated with anticyclonic(v) JWT(v) and southerly JWT(d) categories (Table 4 and Table 5, respectively).

For the full period TS (81–96), categorised by vorticity, the Mann–Whitney pairwise comparison with Bonferroni correction confirms that the anticyclonic(v) (and cyclonic(v)) JWT(v) categories had significantly higher median BS concentration than the directional(v) category (Table 6a). Median BS concentration in the anticyclonic(v) category ($10 \mu\text{g m}^{-3}$) (Table 4) was $3 \mu\text{g m}^{-3}$ greater than in the directional(v) category ($7 \mu\text{g m}^{-3}$), and $2 \mu\text{g m}^{-3}$ greater than the median of all BS data ($8 \mu\text{g m}^{-3}$). The association of the highest concentrations of BS with anticyclonic conditions is consistent with the hypothesis that BS is a marker of emissions from local sources that

become trapped in the non-dispersive stable conditions that typically occur in anticyclonic weather types (King and Dorling, 1997).

	BS				PM ₁₀			
	<i>n</i>	Mean	Median	10–90 percentile	<i>n</i>	Mean	Median	10–90 percentile
TS(81–96)								
Anticyc/cyc(d)	1798	14.2	10	3.0–29.0				
NE	163	12.1	10	3.0–21.8				
E	158	12.4	9	2.0–25.6				
SE	236	15.6	12	4.0–32.5				
S	452	17.5	13	4.0–37.0				
SW	822	9.4	7	2.0–17.0				
W	862	7.2	5	2.0–13.0				
NW	498	8.8	7	2.0–17.0				
N	334	10.9	9	2.3–21.7				
Unclassified days	61	16.0	12	4.0–34.0				
TS(92–96)								
Anticyc/cyc(d)	448	9.3	8	3.0–17.0	452	22.2	20	13.0–34.0
NE	50	9.8	8	4.0–17.0	48	18.7	17	11.7–27.6
E	54	7.9	8	3.0–12.0	52	23.7	21	15.0–34.8
SE	72	9.5	8	3.0–16.0	75	29.3	26	16.2–46.6
S	138	11.0	9	3.0–22.3	134	25.2	23.5	14.0–38.0
SW	200	7.4	5	2.0–14.0	184	16.7	15	10.0–26.0
W	228	5.0	4	2.0–9.0	214	16.6	16	11.0–22.0
NW	110	6.8	5	2.0–14.0	105	17.7	17	12.0–24.0
N	78	7.1	7	2.0–11.3	73	18.1	18	11.0–24.0
Unclassified days	14	10.4	9.5	3.0–15.0	12	20.7	20	14.0–24.9
TS(95–96)								
Anticyc/cyc(d)	97	8.1	7	2.0–13.4	131	22.7	20	13.0–37.0
NE	15	7.0	5	3.4–9.0	18	16.3	15	10.7–24.1
E	17	7.2	8	3.0–13.4	18	22.9	18.5	14.4–31.8
SE	32	7.8	6.5	2.0–12.0	38	32.1	26.5	18.7–55.1
S	35	8.0	8	2.0–15.6	41	22.3	21	13.0–34.0
SW	38	6.3	5	2.0–11.3	47	16.7	15	11.0–26.0
W	19	5.3	5	1.8–8.4	32	16.3	16.5	13.0–19.9
NW	9	3.9	4	2.0–6.0	15	16.5	15	13.4–20.6
N	12	4.5	4.5	2.1–6.0	18	16.8	16.5	11.0–23.3
Unclassified days	2	13.5	13.5	12.3–14.7	2	20.0	20	20.0–20.0

Table 5. Descriptive statistics for BS and PM10 concentrations ($\mu\text{g m}^{-3}$) by JWT(d) categories

(a)								
Cyclonic (9)	ND							
Directional (7)	***		***					
		Anticyclonic (10)		Cyclonic (9)				
(b)								
SE (12)	ND							
AC/C (10)	***		*					
NE (10)	***		*	ND				
E (9)	***		*	ND	ND			
N (9)	***	***	***	***	ND	ND		
SW (7)	***	***	***	***	***	***	***	
NW (7)	***	***	***	***	***	***	***	ND
W (5)	***	***	***	***	***	***	***	***
	S (13)		SE (12)		AC/C (10)		NE (10)	
							E (9)	
							N (9)	
							SW (7)	
								NW (7)

Table 6. Mann–Whitney pairwise comparisons of BS TS (81–96) classified by (a) JWT(v) category, (b) JWT(d) category

Categories are tabulated in order of median concentration ($\mu\text{g m}^{-3}$), given in parentheses. *** Significant difference at the Bonferroni-corrected critical value of $p=0.05/(0.5k(k^{-1}))=0.05/3=0.017$ or $p=0.05/(0.5k(k^{-1}))=0.05/36=0.001$, where k is the no. of categories compared, for data in (a) and (b), respectively. *Not significant at Bonferroni but significant at standard pairwise critical value of $p=0.05$. ND=no significant difference ($p>0.05$).

For the full period TS (81–96) categorised by directional weather-type (Table 5), the median BS concentration in the southerly JWT(d) category ($13 \mu\text{g m}^{-3}$) was $8 \mu\text{g m}^{-3}$ greater than the JWT(d) category associated with the lowest median BS concentration (westerly, $5 \mu\text{g m}^{-3}$) and $5 \mu\text{g m}^{-3}$ greater than the median ($8 \mu\text{g m}^{-3}$) of all BS concentrations. The results of M–W pairwise comparisons of BS with JWT(d) category, are given in Table 6b. For example, BS concentrations in the southerly category were significantly higher than BS concentrations in all other directional categories apart from south-easterly. BS concentrations in the westerly category were significantly lower than BS concentrations in all other directional categories. Overall, the JWT(d) categories essentially form four groups (S and SE; AC/C, N, NE and N; SW and NW; W) with BS concentration that differ significantly from the others.

The magnitude of the ranges of BS concentration across weather type categories were very significant proportions of the median absolute BS concentrations experienced. The median BS concentration for the whole TS (81–96) dataset was $8 \mu\text{g m}^{-3}$ (Table 4), whilst the range of median BS concentration across JWT(v) and JWT(d) categories were 3 and $8 \mu\text{g m}^{-3}$, respectively. It was therefore clear that, weather type is an important determinant of the magnitude, on average, of the concentration of BS in Edinburgh.

3.2.2. Influences on pm10 concentrations

Highest median PM10 concentrations for the TS (92–96) period were also consistently associated with anticyclonic(v) days for JWT(v) categories (Table 4), whilst for JWT(d) categories, highest median PM10 concentrations were associated with the south-easterly category (Table 5). K–W tests showed that, as for BS, median PM10 concentrations were highly significant different ($p<0.001$) across both JWT(v) and JWT(d) categories. For the period TS (92–96), the highest median PM10 concentrations for JWT(v) and JWT(d) categories (21 and $26 \mu\text{g m}^{-3}$ for anticyclonic(v), Table 4, and south-easterly, Table 5, respectively) were 4 and $11 \mu\text{g m}^{-3}$ greater than the JWT(v) and JWT(d) categories with lowest median PM10 concentrations (directional(v) ($17 \mu\text{g m}^{-3}$) and south-westerly ($15 \mu\text{g m}^{-3}$)), respectively. The median PM10 concentration in Edinburgh over the TS (92–96) time period was $18 \mu\text{g m}^{-3}$ (Table 4). Therefore, it was again clear that weather type influenced a significant proportion, on average, of the PM10 concentration experienced in Edinburgh.

3.3. Influence of air mass back trajectory on particle concentration

Table 7 contains mean and median PM10 and BS concentrations for all trajectory categories in dataset TS (95–96). Median BS concentrations ranged from 4 to $10 \mu\text{g m}^{-3}$, and median PM10 concentrations ranged from 17 to $30 \mu\text{g m}^{-3}$. Highest and lowest median concentrations of BS were associated with Western European and

Eastern European and trajectory categories respectively. Highest and lowest median PM10 concentrations were observed for the Eastern European and Atlantic/Arctic/UK trajectory categories respectively.

TS (95–96)	BS				PM ₁₀			
	<i>n</i>	Mean	Median	10–90 percentile	<i>n</i>	Mean	Median	10–90 percentile
Trajectory category								
Vicinity of the UK	33	8.5	6	2.2–18.4	41	20.7	17	11.0–39.0
Atlantic	108	6.7	5.5	2.0–12.0	152	18.2	17	12.0–24.0
Arctic	45	7.1	7	3.0–12.0	54	19.0	17	11.3–31.1
Western Europe	21	10.2	10	2.0–18.0	24	25.4	22.5	15.3–35.8
Scandinavia	15	6.2	6	2.0–10.8	20	18.6	19	11.0–27.3
Maritime	12	7.2	7	3.1–12.0	17	23.9	22	15.2–30.4
Eastern Europe	41	7.0	4	2.0–12.0	47	32.8	30	17.6–55.8
Unclassified	1	4.0	4	4.0–4.0	5	24.0	22	17.2–32.4
All data	276	7.2	6	2.0–12.0	360	21.4	19	12.0–35.1

Table 7. Descriptive statistics for BS and PM10 concentrations by trajectory category. All concentrations are expressed in $\mu\text{g m}^{-3}$

K–W tests applied to the TS (95–96) data showed that there was no significant difference in median BS concentrations, whether categorised by JWT(v), JWT(d) or trajectory. However, median PM10 concentrations did differ significantly across all three categorisations ($p < 0.001$). BS is a good indicator of local combustion-related primary particulate matter. Correspondingly this pollution metric may only be influenced to a relatively small extent by long-range transport. In contrast, the differences in PM10 concentration may have been the consequence of trans-boundary transport of secondary (non-BS) particles (APEG, 1999; Stedman, 1998; King and Dorling, 1997). Although BS concentrations did not differ significantly with weather type or trajectory for TS (95–96) (Table 7), this metric did differ with weather type for the longer periods (Table 4, Table 5 and Table 6). This may be a consequence of the smaller dataset, but is more likely due to the reductions in transport of primary (dark) particles from heavily populated areas in continental Europe and England over this time period (APEG, 1999), arising from large declines in domestic and industrial coal combustion. Although PM10 concentrations for TS (92–96) and TS (95–96) have remained relatively similar, there have been notable reductions in BS concentrations between TS (81–96), TS (92–96) and TS (95–96), especially for the weather type categories associated with higher BS levels. This has led to a reduction in the range in median BS concentration across categories (Table 4 and Table 5).

The results of the pairwise M–W comparisons of PM10 concentrations categorised by back-trajectory are shown in Table 8. PM10 concentrations associated with Eastern European back-trajectories are significantly higher than PM10 associated with the other trajectory categories. Overall, there are three broad groups of trajectories (although not all are significantly different at the Bonferroni-corrected critical value): EE; WE & MT; and SC, UK, AR & AT. These findings are consistent with earlier work. Malcolm et al. (2000) have shown that concentrations of particulate pollution are highest when air parcels pass over European source regions en route to the UK. Similarly, Katsoulis (1999) used a flow climatology for Aliartos, Greece, to determine the potential for long-range transport of air pollutants, finding low pollutant concentrations when air masses originated in the Atlantic Ocean, and higher concentrations when air masses originated in continental Europe.

WE (22.5)	*					
MT (22)	*	ND				
SC (19)	***	*	*			
UK (17)	***	***	*	ND		
AR (17)	***	***	*	ND	ND	
AT (17)	***	***	***	ND	ND	ND
	EE (30)	WE (2.5)	MT (22)	SC (19)	UK (17)	AR (17)

Table 8. Mann–Whitney pairwise comparisons of PM10 TS (95–96) classified by back trajectory category

Categories are tabulated in order of median concentration ($\mu\text{g m}^{-3}$), given in parentheses. *** Significant difference at the Bonferroni-corrected critical value of $p=0.05/(0.5k(k-1))=0.05/21=0.002$, where k is the no. of categories compared. *Not significant at Bonferroni but significant at standard pairwise critical value of $p=0.05$. ND=no significant difference ($p>0.05$)

The range in median PM10 concentration across trajectory categories was $13 \mu\text{g m}^{-3}$, and the median PM10 concentration of the back trajectory category with highest concentration was $11 \mu\text{g m}^{-3}$ higher than the median PM10 concentration of $19 \mu\text{g m}^{-3}$ for the whole TS (95–96) dataset. These values are important since epidemiological evidence has consistently demonstrated that increases of the magnitude shown here for PM10 from different back-trajectory source regions are significantly linked to a range of related health outcomes (Pope and Dockery, 1999), and clearly point to the inherent difficulty of controlling PM10 concentrations within local AQMA by local measures alone.

The 1995–1996 period was deliberately chosen to study long-range transport effects since it was known to contain a relatively large number of days dominated by advection from Eastern European sources (Stedman, 1998; King and Dorling, 1997). Correspondingly any extrapolation of health burden impacts from our data should account for different frequencies of such long-range transport episodes during other time periods.

3.4. Influence of climate change on weather type frequency

The trend between 1881 and 2000 of the annual % frequencies of all of the JWT(v) and JWT(d) weather type categories was examined using the Mann–Kendall trend test (Manly, 2001). In most cases the trends observed were non-significant. However, there has been a very significant ($P=0.006$) long-term decreasing trend in annual frequency of the anticyclonic(v) weather type category (Fig 2a) and a significant ($P=0.05$) increasing trend in the frequency of the directional(v) type (Fig 2b). Both of these trends are in a direction to support the suggestion that changing synoptic weather patterns have the potential to reduce median particle concentrations in Edinburgh. Overall therefore, there is no evidence to suggest that the effects of long-range transport of particulate material to Edinburgh from industrialised areas of Europe, may be becoming more pronounced. However, there is some evidence that suggests particle concentrations may decrease slightly because of a small decrease in the frequency of anticyclonic(v) conditions that may tend to inhibit the dispersion of locally emitted pollutants. These findings may also apply to other areas of the UK and suggest that efforts to ameliorate local air quality may be influenced to small extent

by regional scale climate change. It would be useful to confirm these effects by studying climatologies of air mass back trajectories and long-term measurements of meteorological metrics of local dispersion conditions.

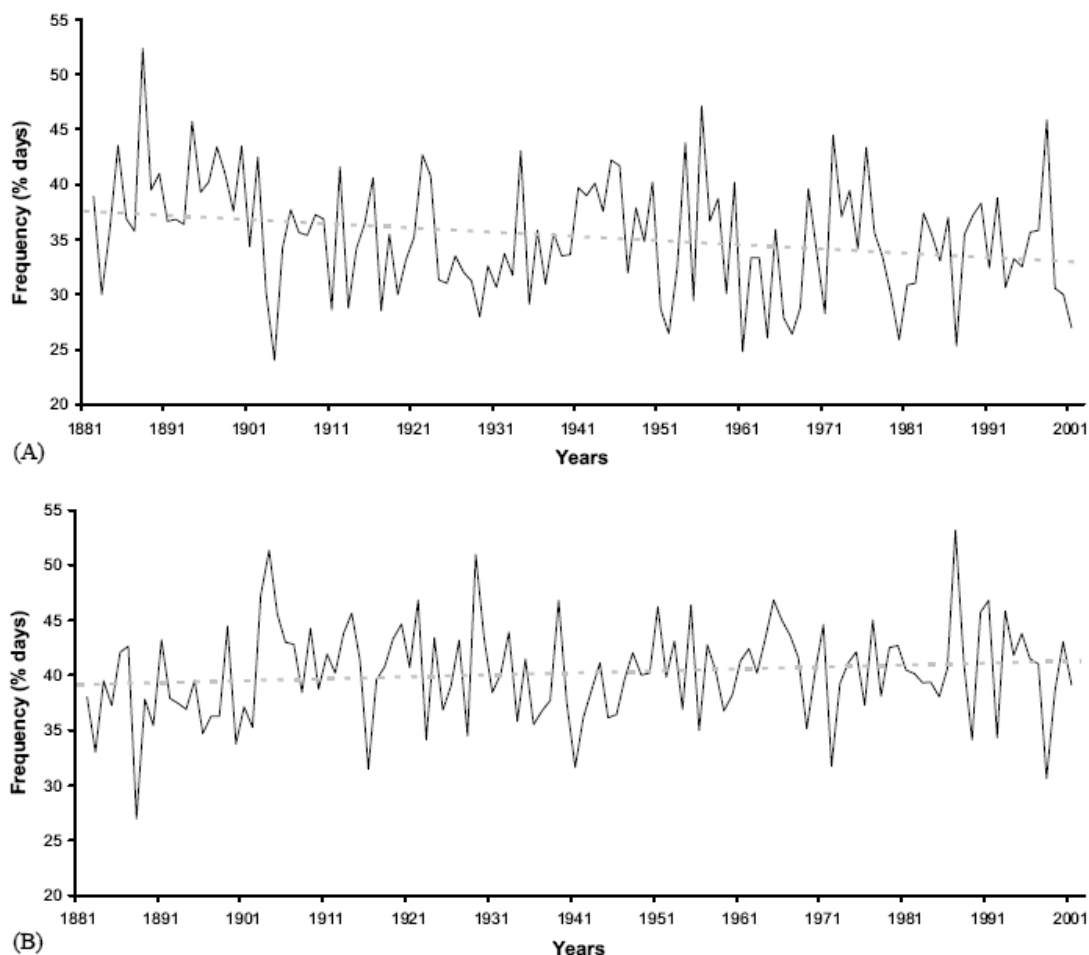


Fig. 2. Annual frequencies of (a) anticyclonic(v) and (b) directional(v) weather types (1881–2000 inclusive). Dotted lines are least-squares linear trends. Time series trends were significant using the Mann–Kendall trend test.

4. Conclusions

This study has shown that synoptic meteorology and long-range transport significantly influenced BS and PM₁₀ pollution in Edinburgh between 1981–1996 and 1992–1996, respectively.

PM₁₀ concentrations remained relatively constant over the 1992–1996 period, although significant reductions were observed in BS concentrations over the same period (and the longer period 1981–1996), particularly for weather type categories associated with higher BS levels.

Classification of weather type by vorticity (anticyclonic(v), cyclonic(v) or directional(v)) showed that median concentrations of BS and PM₁₀ were 3 and 4 μg

m^{-3} higher for the anticyclonic(v) weather type category than for the directional(v) weather type category for the periods 1981–1996 and 1992–1996, respectively. The observation that anticyclonic conditions over the UK leads to elevated particle concentrations, via import from Europe, concurs with the study of King and Dorling (1997).

Median BS concentrations for the period 1981–1996 were 1–8 $\mu\text{g m}^{-3}$ higher for the southerly directional weather type category than for all other directional weather type categories. Similarly, median PM10 concentrations for the period 1992–1996 were approximately 2.5–11 $\mu\text{g m}^{-3}$ higher for the south-easterly directional category than for all other directional weather type categories. During 1995–1996 PM10 concentrations in Edinburgh increased by 11 $\mu\text{g m}^{-3}$ (compared with annual mean) when air mass back-trajectories were from Eastern Europe. However, in 1995–1996, no significant differences were observed for median BS concentration categorised by weather type or trajectory. This suggests that contemporary BS is influenced mostly by local emissions and meteorological conditions, while PM10 continues to be influenced by long-range transport processes.

The differences in median particle concentration between weather type and back-trajectory categories cited above were significant proportions of the overall median BS and PM10 concentrations, clearly demonstrating that PM10 in local areas is influenced by sources out-with local authority control. Since epidemiological research shows that increases in PM10 of 10 $\mu\text{g m}^{-3}$ are associated with increased mortality and morbidity, this study suggests that long-range pollution transport mechanisms may have significant health effects on the population of Edinburgh. Furthermore, it is evident that changes in future climate that impacts upon the relative distribution of synoptic weather type has the potential to change the magnitude (on average) of both urban PM10, and the PM10:BS ratio.

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