

Synoptic circulation and its influence on spring and summer surface ozone concentrations in southern Sweden

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Received 18 Mar. 2008, accepted 30 Nov. 2008 (Editor in charge of this article: Veli-Matti Kerminen)

Tang, L., Chen, D., Karlsson, P.-E., Gu, Y. & Ou, T. 2009: Synoptic circulation and its influence on spring and summer surface ozone concentrations in southern Sweden. *Boreal Env. Res.* 14: 889–902.

The influence of synoptic circulation patterns on surface ozone concentrations at three monitoring sites in southern Sweden was investigated for the spring (April–May) and summer (June–August) periods 1990–2005. Synoptic circulation patterns were classified into six groups based on the Lamb Weather Types (LWTs). The analyses show that the anticyclonic weather pattern (A) and the directional flows from southeast/east (SEE) and southwest/south (SWS) were most frequently associated with high ozone levels. It was estimated that 85.5%, 73.3% and 83.5% of the ozone episode days at Rörvik/Råö, Norra Kvill and Vavihill, respectively, were observed under these three circulation patterns. There were apparent spatial differences in the ozone concentrations during nighttime under condition A that were related to the high altitude position of Norra Kvill and Vavihill. The wind component indices u and v , and the total vorticity index ζ for each circulation pattern reflect the intensity of synoptic circulation and they all have an impact on the variation of surface ozone concentrations. The total vorticity index seems to be the key variable in terms of synoptic influence on surface ozone. A statistic model for the relations between synoptic circulation and ozone concentrations was established based on the frequencies and intensities of the six LWTs. It is able to explain 85% and 71% of the total variances in the observed mean ozone concentrations in spring and summer, respectively, over the period 1998–2005. The results demonstrated the strong impacts of synoptic circulations on surface ozone concentrations in southern Sweden.

Introduction

Tropospheric ozone is an important secondary air pollutant as well as a well-known greenhouse gas contributing to climate change (Levy 1971, Le Treut *et al.* 2007). Surface ozone is primarily produced by photochemical reactions from precursor compounds including methane

(CH₄), volatile organic compounds (VOCs) and carbon monoxide (CO) in the presence of nitrogen oxides (NO_x). Variation in ozone concentration depends not only on emissions but also on meteorological conditions. Meteorological variables such as solar radiation, near surface wind, temperature and precipitation influence ozone formation, deposition and transport processes by

affecting photochemical reactions, stomatal and non-stomatal uptake, and atmospheric dynamic conditions (Lennartson and Schwartz 1999, Solberg *et al.* 2005b, Andersson *et al.* 2007). Furthermore, increasing air temperatures as well as reduced cloudiness and precipitation due to climate change may also promote high ozone concentrations (Meleux *et al.* 2007).

It has long been established that the total ozone column is strongly influenced by synoptic weather variations (Chen and Nunez 1998). Previous studies have also shown that prevailing synoptic-scale circulation govern variation in local climate (Achberger *et al.* 2006, Chen *et al.* 2006), thus having a strong impact on surface ozone concentrations (Kallos *et al.* 1993, Zhang *et al.* 1998, Lennartson and Schwartz 1999, Cheng 2001, Helmis *et al.* 2003, Makra *et al.* 2006). For example, blocking anticyclone was identified as an important cause for ozone episodes over west-central Europe (Gangoiti *et al.* 2002). In the Nordic countries, meteorological variability is even more influential for elevated ozone since they are on the outskirts of the main European ozone precursor emission area (Solberg *et al.* 2005a) and the meteorological conditions vary greatly and quickly. The typical situation of high ozone events in this area is often associated with the breaking up of an extensive high-pressure system due to the approach of a marked cold front system (Solberg *et al.* 2005a). Ozone episode induced by long-range transport in northern Fennoscandia is also related to the activity of high pressure system and frontal systems (Lindskog *et al.* 2007).

Synoptic-scale circulations are usually classified by certain schemes. Distinctive circulation patterns have the advantage of introducing a dynamic element aiding synoptic interpretations (Beaumont and Hawksworth 1997). Lamb weather types (LWTs) are one of the circulation classification being widely used in describing synoptic weather conditions based on a manual scheme developed by Lamb (1950), and automatic scheme developed by Jenkinson and Collinson (1977). O'Hare and Wilby (1995) examined the relation between LWTs and surface ozone concentrations in the UK (United Kingdom) and Ireland. They pointed out the importance of anticyclone, cyclone and westerly types in spa-

tial distributions of surface ozone. LWTs over southern Scandinavia have been calculated by using gridded monthly and daily mean sea level pressure (MSLP) data and applied to determine local variability of meteorological variables such as temperature and extreme precipitation (Chen 2000, Linderson 2001, Hellström 2005).

The overall aim of this study was to investigate the relation between synoptic circulation patterns and surface ozone concentrations in spring (April–May) and summer (June–August) in southern Sweden. Spring and summer were chosen because their relatively high ozone concentrations which have the potential to affect vegetation and human health. The specific aims were to (1) identify the predominant circulation patterns associated with low and high surface ozone concentrations, (2) reveal the spatial difference in ozone distributions, and (3) quantify the impact of circulation patterns on inter-annual variability of ozone concentrations.

Materials and methods

Ozone data

Three monitoring sites with long records of hourly ozone concentrations were used in this study: Rörvik/Råö, Norra Kivill and Vavihill. They are situated in rural areas of southern Sweden and represent regional background levels (Fig. 1 and Table 1). The site Rörvik/Råö was first positioned at Rörvik (57.40°N, 11.92°E) and was then moved 2 km south to Råö (57.25°N, 11.56°E) on 1 January 2002. Both sites are located at the coast but Rörvik is positioned approximately 500 m from the seashore while Råö is located right at the seashore. Norra Kivill (57.49°N, 15.34°E) is an inland site in central southern Sweden, positioned high in the local landscape (261 m a.s.l.). It is less prone to nighttime air temperature inversions and associated stable air layers as compared to many sites in southern Sweden that are positioned low in the local landscape (Sundberg *et al.* 2006). Vavihill (56.01°N, 13.09°E) is situated in the most southern part of Sweden and is the nearest to the mainland Europe. Also this site is positioned relatively high in the landscape (175 m a.s.l.). As monitoring sites in the EMEP (Euro-

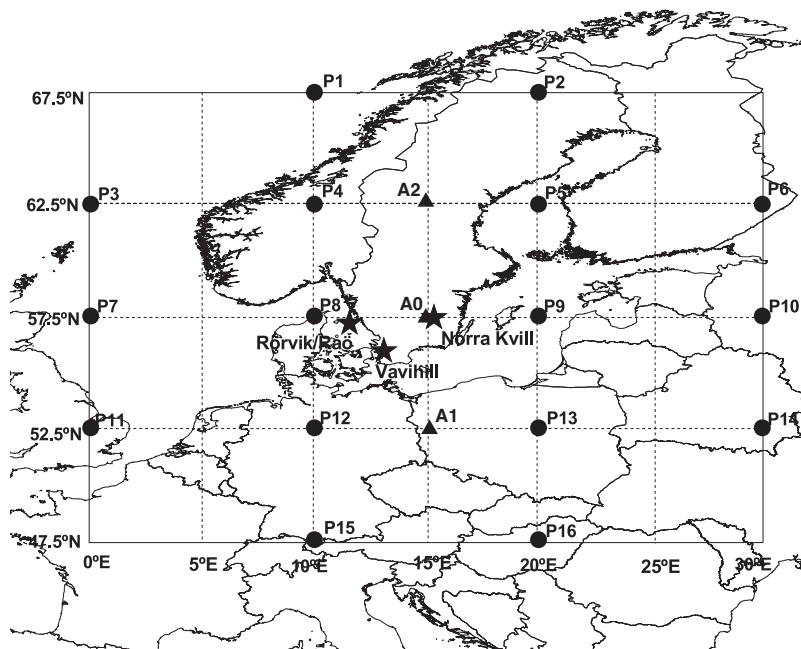


Fig. 1. Locations of 16 grid points (circle) (P1–P16) and three points (triangles) (A0, A1, A2) used to calculate the six Lamb indices. The stars represent the positions of ozone monitoring sites Rörvik/Råö, Norra Kvill and Vavilhill.

pean Monitoring and Evaluation Programme) network, these three sites have been operated since the 1980s with high data quality and small amounts of missing values. Hourly ozone concentrations at these sites for 1990–2005 are available from the official Swedish database hosted by the Swedish Environmental Institute (<http://www.ivl.se/>) and are used for this study.

Lamb weather types (LWTs)

The classification of the synoptic circulation was based the manual scheme developed by Lamb (1950) for the British Isles. This scheme was automated by Jenkinson and Collison (1977) by

defining a number of indices and classification rules. The method is based on a set of indices describing geostrophic wind and vorticity conditions. Zhu *et al.* (2007) described the classification scheme in detail. According to this description, the circulation types over southern Sweden during 1990–2005 were calculated from MSLP data on a 5° latitude by 10° longitude grid-point in southern Sweden (Fig. 1). In this study, the pressure data from 1990 to 2005 were extracted for an area bounded by 47.5°–67.5°N, and 0°–30°E, essentially centred in southern Sweden. The daily MSLP at 16 points were obtained from NCEP Reanalysis data I with 2.5° latitude by 2.5° longitude grid (Kalnay *et al.* 1996) and were used to calculate the following six circulation indices:

Table 1. Positions and the site description of the three ozone monitoring sites used in this study. The period with ozone data is 1 January 1990–31 December 2005 for all the sites. The inlet sampling point is 5 m above ground. The altitude refers to the height above the sea level.

Site	Lat. °N	Long. °E	Altitude (m)	Comments
Rörvik/Råö	57.40/57.25	11.92/11.56	10	Situated on the seashore, surrounded by an open Scots pine forest
Norra Kvill	57.49	15.34	261	Situated at a hilltop, very high in the local topography, surrounded by grassland and an open, mixed forest.
Vavilhill	56.01	13.09	175	Situated on the southern slope of a ridge. On open land, surrounded by a beech forest.

$$u = 0.5[P(12) + P(13) + P(4) + P(5)] \quad (1)$$

$$v = \frac{1}{\cos \alpha} \times \frac{1}{4} \begin{bmatrix} P(5) + 2P(9) + P(13) \\ -P(4) + 2P(8) - P(12) \end{bmatrix} \quad (2)$$

$$V = \sqrt{u^2 + v^2} \quad (3)$$

$$\zeta_u = \frac{\sin \alpha}{\sin \alpha_1} \times \frac{1}{2} [P(15) + P(16) - P(8) - P(9)] \quad (4)$$

$$- \frac{\sin \alpha}{\sin \alpha_2} \times \frac{1}{2} [P(8) + P(9) - P(1) - P(2)]$$

$$\zeta_v = \frac{1}{2 \cos^2 \alpha} \times \frac{1}{4} \begin{bmatrix} P(6) + 2P(10) + P(14) \\ -P(5) - 2P(9) - P(13) \\ +P(3) + 2P(7) + P(11) \\ -P(4) - 2P(8) - P(12) \end{bmatrix} \quad (5)$$

$$\zeta = \zeta_u + \zeta_v \quad (6)$$

Here $P(N)$ is the MSLP at the grid-point N ($N = 1, 2, \dots, 16$) and α , α_1 and α_2 are the latitude at points A0, A1 and A2, respectively (Fig. 1). Indices u and v represent westerly (zonal) and southerly (meridional) components of the geostrophic wind, V is the combined wind speed, ζ_u (meridional gradient of u) and ζ_v (zonal gradient of v) are westerly and southerly shear vorticity, respectively, and ζ is the total shear vorticity index. Since all pressures have the units of hecto Pascals (hPa), the indices have units of hPa per 10° longitude (hPa/ 10° long.) at 57.5° N (see Zhu *et al.* (2007) for the detailed derivation process). Once u and v are known, the wind direction can be determined. As a result, two main categories of circulation type are classified. The so-called directional flow types (north, N; northeast, NE; east, E; southeast, SE; south, S; southwest, SW; west, W; northwest, NW) are characterized by coherent wind direction ($|\zeta| \leq V$). The other category emphasizes rotation of the atmosphere ($|\zeta| \geq 2V$), which can either be cyclonic (C) or anticyclonic (A). There is also a possibility of hybrid categories ($V < |\zeta| < 2V$), which can be any combination of the two main categories. In the case where $V < 6$ hPa/ 10° long. and $|\zeta| < 6$ hPa/ 10° long., the circulation is unclassified (UD). In this study, in the totally twenty-six daily LWTs, not including UD, were classified to identify their influences on surface ozone.

Definition of ozone episodes

Ozone episodes are characterized as short periods with higher than normal ozone concentrations, which can do harm to human health and vegetation. The Swedish ordinance (2000/2001:130) on environmental quality standards for ambient air use the target value 60 ppb as the maximum 8-h mean ozone concentration in order to protect human health by the year 2010. This is identical to environmental quality standards of the EU directive (2002/3/EG) for the year 2020. Therefore, the exceedance of the value 60 ppb for an 8-h moving average was used to define an ozone episode in this study. The 8-h moving average was calculated as a running average for every hour throughout the day, producing 24 values for each day. According to the EU directive, setting the time for each period was based on the last hour. Hence, the first 8-h average in a day (01:00) is calculated as the average of the eight 1-h values from 17:00 to 01:00 of the previous day. If any of the moving 8-h averages during the day was greater than or equal to 60 ppb, this day is defined as an episode day.

Statistic analysis

Trends in mean ozone concentrations were calculated for the different periods. In order to establish if the trends were statistically significant, a Mann-Kendall test was applied. This method has been used to detect air pollutant trends (Salmi *et al.* 2002) as well as long-term changes in Swedish precipitation (Busuioac *et al.* 2001). The Mann-Kendall test is a non-parametric test that has the advantage of robustness against outliers and can be applied to non-normally distributed data with missing values. To estimate the slope of an existing trend Sen's non-parametric method was used (Salmi *et al.* 2002).

The stepwise regression method has been widely used in synoptic climatological and air pollution studies due to its ability to identify sequentially the optimum subset of independent variables. (Wolff *et al.* 1986, Eder *et al.* 1994, Lam and Cheng 1998, Kim Oanh *et al.* 2005). A stepwise regression can start with no model term (forward), all the terms (backward) or a subset

of all the terms. In this study, the backward stepwise method was used to establish linear regression models between the dependent variable ozone concentration and the three independent variables (Lamb indices u , v and ζ). If the index was significant at $p < 0.05$, it was included as the variables in the model. If there was no index that was statistically significant, then all the three indices were used as model variables. The stepwise analyses were carried out using functions packaged in MATLAB 7.2.

Results and discussion

LWTs and surface ozone

Six merged LWTs

Including infrequent weather types in the statistical analysis makes it difficult to establish reliable statistical relations. Previous studies have stressed the need of reduced number of LWTs for a specific application and region (Goodess and Palutikof 1998, Linderson 2001). In this study, the 26 original LWTs were merged into fewer groups according to their influences on surface ozone concentrations in this region. Firstly, the anticyclonic (A) and cyclonic (C) types were kept because of their distinct features and high frequencies. Secondly, the anomalies of averaged ozone concentrations at the three monitoring sites under the directional types, A-hybrid and C-hybrid types were compared (Fig. 2). The A-hybrid and C-hybrid types did not show big deviation from directional types so they were merged into directional types. Thirdly, eight directional types were combined into four types based on the possible source regions from these directions: west (W), southeast and east (SEE), southwest and south (SWS), north, northeast and northwest (N+). W is the most common directional type in southern Sweden and represents one possible emission source: the Great Britain. SEE and SWS are the directions of eastern and central Europe which are two identified emission sources for ozone formation in the Nordic countries (Solberg *et al.* 1997). N+ represents relative clean air direction from North Atlantic Ocean and Arctic region. Finally, the 26 LWTs

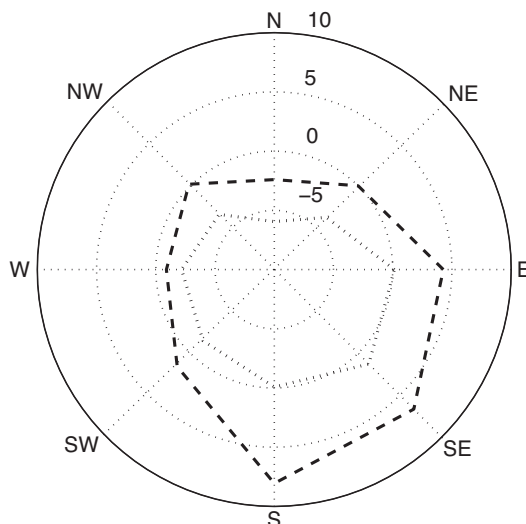


Fig. 2. Mean ozone deviations from the daily-averaged ozone concentrations at the three sites from April to August over the period of 1990–2005 for eight directional types (solid line), A-hybrid types (dashed line) and C-hybrid types (dotted line).

were combined into the six LWT groups: A, C, W, SEE, SWS and N+.

LWT groups and mean ozone concentrations

The influence of the six LWT groups on the surface ozone concentrations were investigated by calculating the deviations from the averaged ozone concentrations at the three sites during April–August for the entire period 1990–2005 (Fig. 3). The comparisons were carried out for the three different time periods: daily (24-h), daytime (08:00–20:00 UTC) and nighttime (20:00–08:00 UTC). In general, positive deviations were detected under A, SEE and SWS, whilst negative deviations were found under C, W and N+. Spatial differences between the different monitoring sites were most evident during nighttime. Sites that are positioned high in the local topography experience less nighttime ozone depletion than low-level sites under stable boundary layer when relatively ozone rich air is transported from aloft with down-sloped cold air (Coyle *et al.* 2002, Sundberg *et al.* 2006, Karlsson *et al.* 2007). Therefore, the hilltop sites

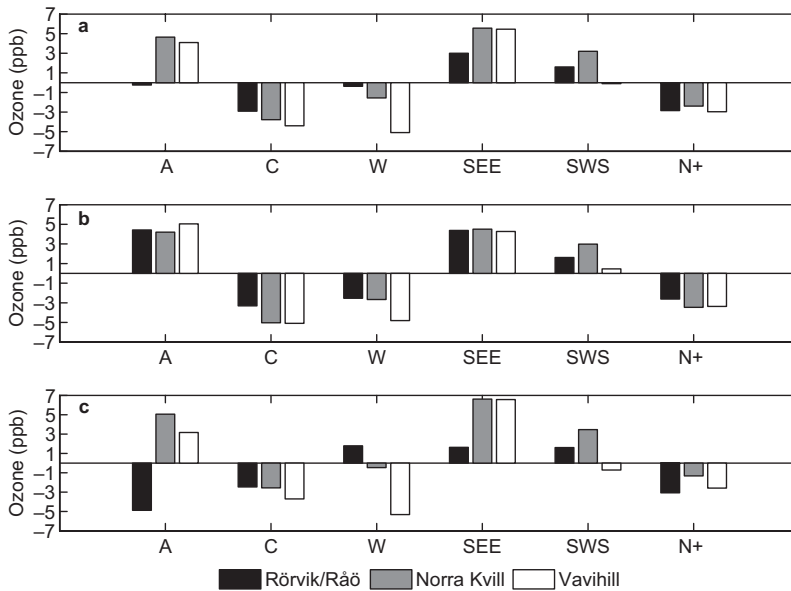


Fig. 3. Mean ozone deviations from the averaged ozone concentrations at the three sites under the six LWTs from April to August over the period of 1990–2005 at Rörvik/Råö, Norra Kville and Vavihill. (a) daily mean (24-h), (b) daytime (08:00–20:00 UTC) mean and (c) nighttime (20:00–08:00 UTC) mean.

Norra Kville (261 m a.s.l.) and Vavihill (174 m a.s.l.) showed higher positive deviations in nighttime ozone concentrations under type A. In addition, Norra Kville showed higher positive deviations than Vavihill during nighttime since the degree of nighttime ozone depletion decreases with altitude (Entwistle *et al.* 1997, Coyle *et al.* 2002). Rörvik/Råö, on the other hand, showed frequent nighttime air temperature inversions which resulted in low nighttime ozone concentrations under type A. The positive deviations of daytime ozone concentrations under anticyclonic weather pattern were similar at both coastal and elevated sites, due to relatively deep and efficient mixing. Under the unstable cyclonic weather pattern, ozone and its precursors seldom accumulate to high levels and are more easily dispersed under high mixing levels and excellent ventilation (O'Hare and Wilby 1995). Dominant flows from south, southwest, east and southeast tend to transport ozone and its precursors from the identified source regions in central and eastern Europe resulting in elevated ozone concentrations.

LWT groups and ozone episodes

The influence of LWTs on ozone episodes can be

demonstrated by their frequencies under episode days (Fig. 4). Higher values for the frequencies of LWT groups during episode days (grey bars), as compared with the total frequencies of LWT groups (black bars) under A, SEE and SWS indicate their higher correlation with ozone episodes. Indeed, 85.5%, 73.3% and 83.5% of the episode days occurred under A, SEE and SWS at Rörvik/Råö, Norra Kville and Vavihill, respectively. Relative frequency clarifies the percent of ozone episode for each LWT, giving the relative importance of the circulation pattern to ozone episode. The higher white bars (relative frequency) under SEE demonstrate the closer relationship between SEE and high ozone levels despite its fewer occurrences. The probability that there will be an ozone episode when there was a SE/E directional flow was close to 20% at Rörvik/Råö (17.0%) and Vavihill (17.9%), while the corresponding value for the anticyclonic weather type was somewhat lower, slightly above 10% (12.5% at Rörvik/Råö, 13.3% at Vavihill, 10.1% at Norra Kville). The empirical cumulative distribution for the mean daily maximum of 8-h moving average at the three sites were calculated under the three high ozone level related patterns A, SEE and SWS (Fig. 5). The major differences among the three groups occurred between 40–60 ppb and nearly 50% of data were larger than 50 ppb under SEE.

Fig. 4. Comparison of the frequencies for daily LWTs at (a) Rörvik/Råö, (b) Norra Kivill and (c) Vavihill from April to August over the period of 1990–2005. The black bars represent the frequencies of LWTs based on all days, the grey bars represent the frequencies of LWTs based on the episode days and the white bars represent the relative frequencies of episode days associated with a certain LWT.

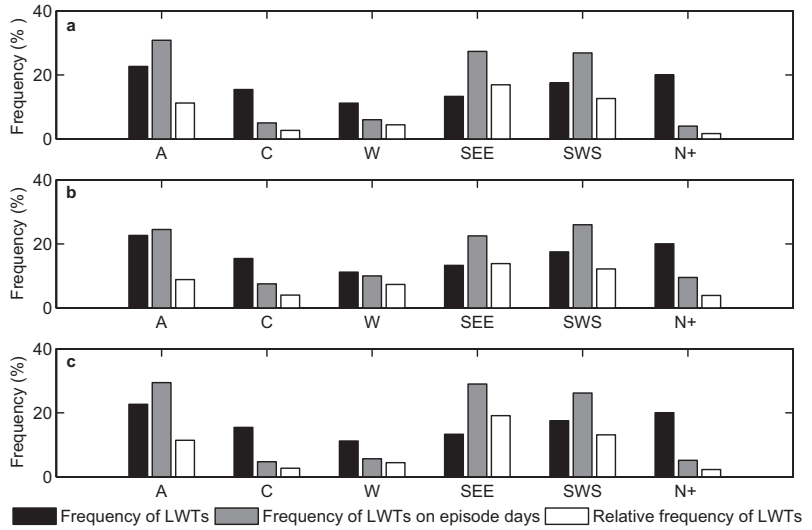
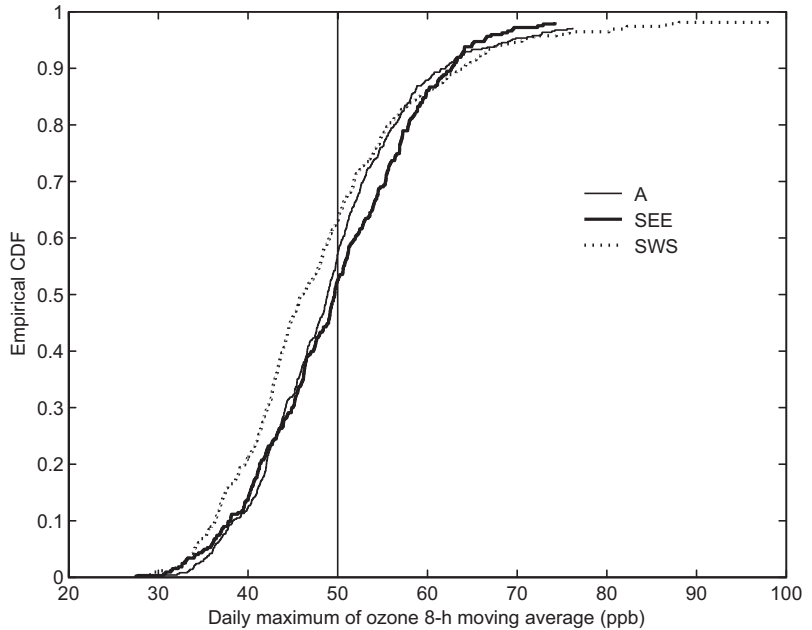


Fig. 5. The empirical cumulative distribution function (CDF) plot for the mean daily maximum of 8-h moving average (ppb) at the three sites under high ozone level related circulation patterns A, SEE and SWS.



The extreme high ozone level (8-h moving average > 80 ppb) occurred only under pattern SWS.

Evaluation of the LWTs-ozone relationship

A stagnant or slow-moving high-pressure system is frequently associated with high ozone levels on the regional scale (Vukovich 1994). High-pressure systems during summer is known to give

rise to sunny, dry and calm weather conditions during daytime, which promotes photochemical reactions and ozone production (Kallos *et al.* 1993, Zhang *et al.* 1998). At nighttime, ozone and its precursors are trapped aloft the nocturnal residual layer. On the following day they can again be entrained downward into the mixing layer, as the surface-based inversion starts to break up (Neu *et al.* 1994, Zhang and Rao 1999, Athanassiadis *et al.* 2002). Furthermore, strong subsidence enhances vertical transport from the

lowermost stratosphere. The enhanced lowermost stratospheric ozone levels have been found to influence surface ozone variability and contribute to the positive trend of background ozone concentrations over Europe during the 1990s (Ordóñez *et al.* 2007). Therefore, anticyclonic weather pattern (A), in particular the duration of high pressure systems (Zhang and Rao 1999), is often associated with high ozone.

Cyclonic weather pattern (C) represents an unstable weather condition and is often associated with clouds and rainy weather. In Sweden, cyclonic pattern together with its hybrid types are associated with 70% extreme precipitation events due to high mean vertical velocities and high mean specific humidity (Hellström 2005). Low air temperatures and weak solar radiation restrict the photochemical reaction and ozone production. Effective convective mixing in the main ascending branch of an extratropical cyclone is likely to ventilate the boundary layer air into the upper troposphere (Stohl 2001, Esler *et al.* 2003). Therefore cyclonic weather is usually associated with low ozone levels. However, frontal systems with strong convection, naturally included in the cyclonic weather types, can cause elevated ozone in Nordic countries by long-range transport from the European continent (Solberg *et al.* 2005a). This is indicated as the small but significant proportion of ozone episodes observed under C (Fig. 4).

Directional flows are associated with long-range transport from source regions by horizontal advection. According to EMEP emission inventory in 2000 (<http://www.emep.int>), the potential source regions of high NO_x and VOCs were the Great Britain, Netherlands, Belgium, western and eastern Germany, northwest Czech Republic, southern Poland, central Belarus, western Russia and eastern Ukraine. The higher ozone levels under SEE and SWS were associated with the air masses transported from these emission source regions. Compared with those under SEE and SWS, ozone levels under W were relatively low even though Great Britain is also a well-known pollution source. A possible explanation is that the directional flow from westerly flow contained a large proportion of air masses from the Atlantic, which has not passed over the polluted areas of the Great Britain.

Trends of ozone concentrations and the link with LWTs

The relationship between LWTs and surface ozone concentrations at the three sites shows a strong influence from synoptic patterns on the ozone formation and transport processes. This indicates a link between the inter-annual variability of synoptic patterns and surface ozone concentrations. However, this link can be modulated by other factors such as photochemical reactions that are controlled not only by meteorological variables, but more importantly also by emissions of the precursors. Indeed, previous studies have showed that the high ozone levels in western Europe during the periods 1989–1997 have been substantially reduced from 1998 onwards, which is mainly attributed to the substantive control measures to reduce anthropogenic NO_x and VOCs emissions (Vestreng 2001, Derwent *et al.* 2003, Simmonds *et al.* 2004). Since the photochemical reactions are active in the study seasons (April–August), the contribution of the changes in emissions to the ozone concentration variation can be substantial. To minimise this effect, we focused on the period from 1998 to 2005 during which there were small changes in the anthropogenic emissions. This way the impact from the atmospheric circulation can be readily identified.

There are accumulating evidences that the background tropospheric ozone concentrations are increasing on a large geographical scale (Prather *et al.* 2003, Simmonds *et al.* 2004, Laurila *et al.* 2004a, Carslaw 2005, Derwent *et al.* 2007), as well as in northern Sweden (Lindskog 2003, Karlsson *et al.* 2007), Norway (Solberg 2003) and Finland (Laurila *et al.* 2004b). The increasing trends of 0.5 ± 0.3 ppb year⁻¹ in spring (March–May) and 0.4 ± 0.3 ppb year⁻¹ in summer (June–August) were observed at background station at Mace Head, Ireland, from 1987 to 2003 (Simmonds *et al.* 2004). However, the slightly but not statistically significant increasing trends were found in the mean summertime ozone concentration during May–July, 1989–2001 in Finland (Laurila *et al.* 2004b) and April–September, 1990–2002 in Norway (Solberg 2003). At Vindeln in northern Sweden, annual mean ozone concentration (April–September) showed

an increasing trend of 0.2 ppb year⁻¹ over the period 1990–2006 (Karlsson *et al.* 2007). A statistical analysis of the period from 1998 to 2005 showed that the annual mean ozone concentrations in spring (April–May) at Rörvik/Råö and Norra Kvill had a significant upwards trend of 0.7 ± 0.3 ppb year⁻¹ ($p < 0.05$) and 0.5 ± 0.4 ppb year⁻¹ ($p < 0.1$), respectively (Table 2). Interestingly, the significant increasing trends occurred under A, SEE or SWS circulation patterns. The summer (June–August) ozone concentrations for 1998–2005 showed a slightly increasing but less significant trend at Rörvik/Råö and Vavihill (Table 2).

The cause for the increase in the background level remains unclear since many processes are interacting, such as photochemistry on a continental or hemispheric scale, long-range transport, stratosphere-troposphere exchange, biomass burning at a large scale, climate change and policy-mandated emission control. In this study, the influence from synoptic weather conditions was reflected in correlation between inter-annual mean ozone concentration and some of the frequencies of the six LWT groups (Table 3). A significant correlation coefficients (r) were found under C ($r = -0.85, p < 0.01$) and SEE ($r = 0.75, p < 0.05$) in summer. Together with the positive impact of SEE on the ozone levels, the significantly increased SEE frequency may be linked to the weakly increased summer ozone concentrations over the period 1998–2005 (Table 2). In spring, the role played by the six LWT groups is less clear, which is demonstrated by poor correlation between the LWT frequencies and the ozone variation.

It has been proposed that there exist a potential link between observed inter-annual variability of ozone and the intensity of weather circulation patterns (Hegarty *et al.* 2007). In this study, Lamb indices based on MSLP reflect the variation of mean position, size and central pressure of dominant circulation, thus reflecting the variation of circulation intensity. Among the six Lamb indices, combined wind speed index V is dictated by u ; ζ_u, ζ_v are highly correlated with ζ which has a clear synoptic and physical interpretation. Therefore, we used wind component index u and v to reflect the feature of synoptic flow, and the total vorticity index

Table 2. Results of the Mann-Kendall test for trends ('+' upwards and '-' downwards) of annual mean ozone concentration in six LWTs in spring (April–May) from 1998 to 2005 at Rörvik/Råö, Norra Kvill and Vavihill. The slopes of significant trend for all types are estimated by Sen's method. Significance: * $p < 0.05$, # $p < 0.1$.

	Daily mean (24-h)			Daytime mean (08:00–20:00 UTC)			Nighttime mean (20:00–08:00 UTC)		
	Rörvik/Råö	Norra Kvill	Vavihill	Rörvik/Råö	Norra Kvill	Vavihill	Rörvik/Råö	Norra Kvill	Vavihill
Spring									
A	#	+	+	+	+	+	*	+	+
C	+	+	+	+	+	+	+	+	+
W	+	+	+	+	+	+	+	+	+
SEE	+	+	#	+	+	*	+	#	+
SWS	+	+	#	#	+	+	#	*	+
N+	+	+	+	-	-	+	+	-	+
All types	+	#	+	+	+	+	+	*	+
	(0.7 ± 0.3 ppb y ⁻¹)(0.5 ± 0.4 ppb y ⁻¹)						(1.1 ± 0.4 ppb y ⁻¹)(0.6 ± 0.4 ppb y ⁻¹)		
Summer									
A	+	+	+	+	+	+	+	+	+
C	+	+	-	-	+	-	+	+	-
W	+	+	+	+	+	+	+	+	+
SEE	+	+	-	-	-	-	+	-	-
SWS	+	+	+	+	-	+	+	-	+
N+	+	+	+	+	+	+	+	+	+
All types	+	-	+	+	+	+	+	+	+

Table 3. Correlation coefficient between annual mean ozone concentrations averaged over the three sites and frequencies of LWTs in spring (April–May) and summer (June–August) over the period 1998–2005.

	A	C	W	SEE	SWS	N+
Spring	0.34	−0.58	0.22	−0.35	0.49	−0.07
Summer	0.48	−0.85**	−0.39	0.75*	−0.03	−0.22

Significance: ** $p < 0.01$, * $p < 0.05$.

ζ as an indicator of rotation of the atmosphere and strength of a high/low pressure system. The positive and negative values of u and v and their correlation coefficients with ΔC (the difference between yearly averaged ozone concentrations at three sites and corresponding 16-year average, 1990–2005) under the six types were examined separately (Table 4). The result showed that ozone concentrations decreased with increased westerly wind index ($+u$) in summer. This strong negative correlation ($r = -0.84$) provided an evidence of lower ozone concentrations under W. Positive vorticity index (ζ) indicates low pressure system whilst negative vorticity indicates high pressure system. The significant correlation between ΔC and ζ reflects ozone build up under anticyclonic weather and ozone vertical transport by strong ventilation and mixture under cyclonic weather (Fig. 6). The stronger the high pressure systems were, the higher the ozone concentrations presented; whilst the stronger the low pressure systems were, the lower the ozone concentrations observed. Thus, the total vorticity index ζ seem to be able to serve as a key vari-

able in terms of its ability in describing synoptic impact on surface ozone.

Reconstruction of the inter-annual variability of ozone concentrations

To elucidate the influence of circulation patterns on the surface ozone concentrations, statistical models were established to estimate annual averaged ozone concentrations at the three sites. Following Hegarty *et al.* (2007), inter-annual variability of ozone concentrations was reconstructed by considering the frequency and intensity of the six weather patterns as follows:

$$\bar{C}_k = \sum_{i=1}^6 (\bar{C}^i + \Delta C_k^i) f_k^i \quad (7)$$

where \bar{C}_k is the annual mean ozone concentration averaged over the three sites in year k ; \bar{C}^i is the mean ozone concentration for LWT group i

Table 5. Selected LWTs indices incorporated into stepwise regression equations to predict the ΔC_k^i in Eq. 7 for each LWT in spring (April–May) and summer (June–August) in year k ; R^2 values for each equation are listed.**Table 4.** Correlation coefficient between ΔC (the difference between annual mean ozone concentration averaged over the three sites and the corresponding 16-year average, 1990–2005) and circulation indices in spring (April–May) and summer (June–August) over the period 1998–2005. $+u$ is the index of wind component from west, $-u$ is the index of wind component from east, $+v$ is the index of wind component from south, $-v$ is wind component from north and ζ is the index of the total vorticity.

	$+u$	$-u$	$+v$	$-v$	ζ
Spring	−0.16	−0.53	−0.06	/	−0.78*
Summer	−0.84**	/	0.63	0.77	−0.82*

Significance: ** $p < 0.01$, * $p < 0.05$. ‘/’ means no value or only one value.

	Regression model	R^2
Spring		
A	$\Delta C_k^1 = -18.91 - 0.89\zeta$	0.46
C	$\Delta C_k^2 = -0.18 + 0.24u - 0.10v + 0.02\zeta$	0.08
W	$\Delta C_k^3 = 8.04 - 0.61u$	0.47
SEE	$\Delta C_k^4 = -7.83 - 0.48u + 0.48v - 0.54\zeta$	0.44
SWS	$\Delta C_k^5 = -14.38 + 0.80u + 1.13v - 0.12\zeta$	0.60
N+	$\Delta C_k^6 = 5.24 - 0.47u + 0.30v - 0.44$	0.95
Summer		
A	$\Delta C_k^1 = -1.25 - 1.85u$	0.71
C	$\Delta C_k^2 = -10.03 - 2.86u + 0.13v + 0.69\zeta$	0.34
W	$\Delta C_k^3 = 0.88 - 0.07u - 1.83v - 0.06\zeta$	0.23
SEE	$\Delta C_k^4 = -9.34 - 0.72u + 0.77v + 0.29\zeta$	0.23
SWS	$\Delta C_k^5 = -17.19 + 1.25u + 1.48v - 1.58\zeta$	0.62
N+	$\Delta C_k^6 = 5.60 + 0.55u + 0.97v - 0.09\zeta$	0.08

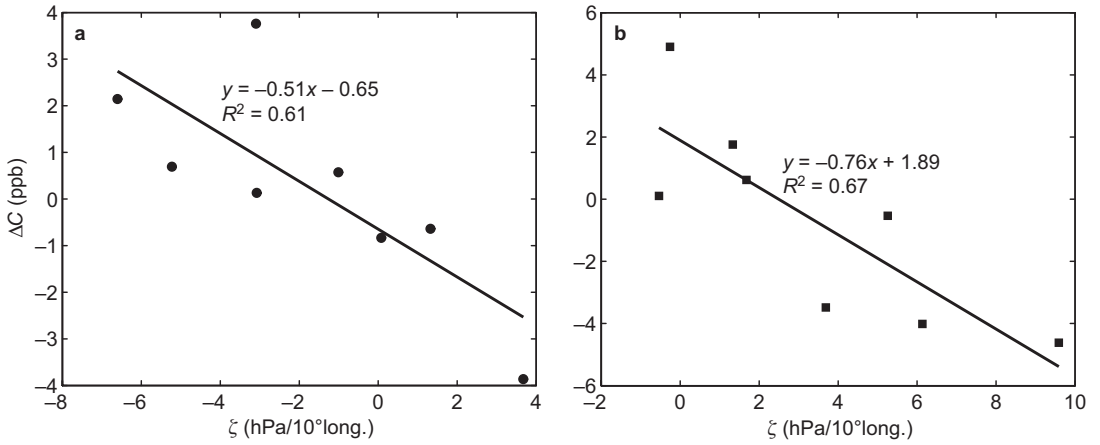


Fig. 6. Scatter plot of ΔC (ppb) (the difference between annual mean ozone concentration averaged over the three sites and the corresponding 16-year average, 1990–2005) vs. the circulation indices total vorticity (ζ) for all types in (a) spring and (b) summer. The unit of circulation index is hPa/10° longitude (hPa/10°long.). The best fit line and its expression are also shown in the figures.

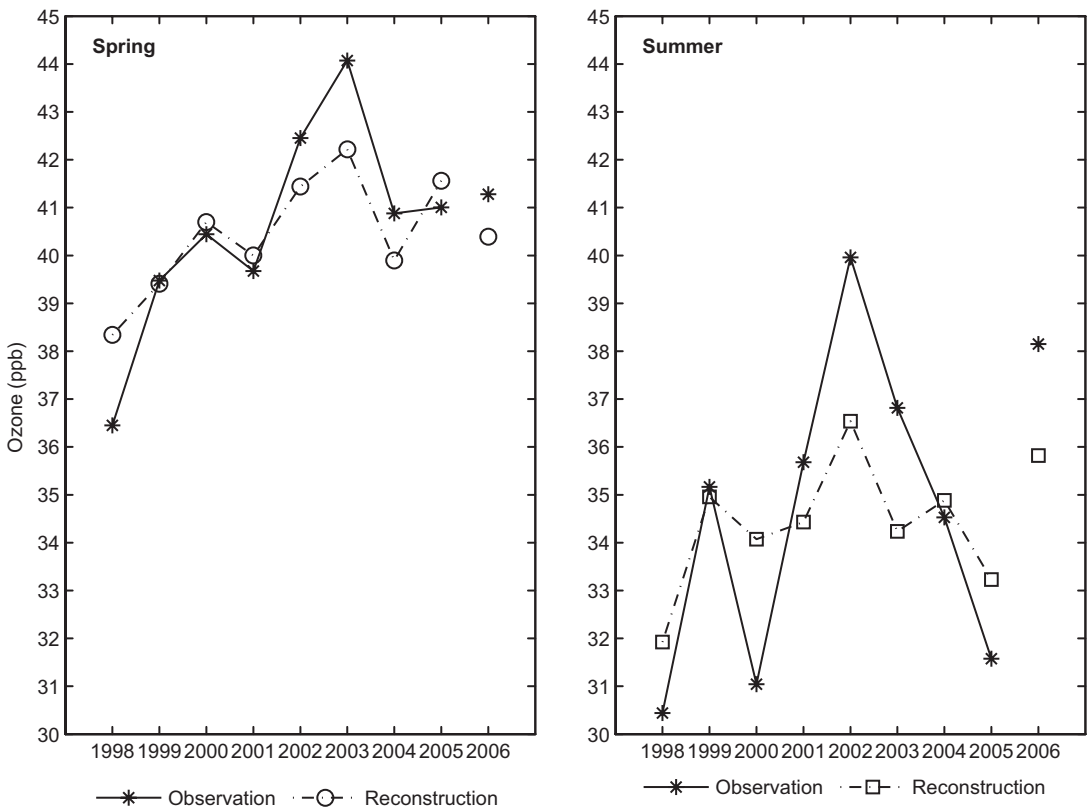


Fig. 7. Reconstruction of inter-annual averaged ozone concentrations at the three sites from 1998 to 2005 with considering both frequencies and intensity of six circulation patterns (Eq. 7). The ozone values in 2006 are calculated independently by using the indices of circulation patterns in 2006.

from 1990 to 2005; f_k^i is the frequency of LWT group i ($i = 1, 2, 3, \dots, 6$) in year k , and ΔC_k^i is the differences between the annual mean ozone concentration averaged over the three sites and the corresponding 16-year average for each pattern i and year k . To determine the ΔC_k^i , the three Lamb indices (u, v, ζ) were used to establish linear regression models for each type in this study. By implementing the backward stepwise regression method, ΔC_k^i was calculated from the regression model for each type (Table 5). Then, the predicted ΔC_k^i were used in Eq. 7 and the annual mean ozone concentrations in spring and summer obtained. As a simple independent test of the models, ozone concentrations in 2006 were predicted independently by using the indices of LWTs in 2006 (Fig. 7).

The reconstructed inter-annual averaged ozone concentrations by the statistic models are in reasonable agreement with the observations. The curves show that 85% and 71% of the observed variances are reconstructed in spring and summer respectively by Eq. 7. Interestingly the modeled maxima (e.g. the spring maximum in 2002 and the summer maximum in 2003) appear in the correct years, although the values are underestimated due to the nature of the statistical model. These underestimations may be attributed to factors that are not included in the model, such as large scale boreal biomass burning events, which have been estimated as the contribution to the larger than average annual rates of ozone concentrations (Simmonds *et al.* 2005). Other possible influencing factors include nonlinear relations with climate change, and policy-mandated reductions of anthropogenic emissions (Hegarty *et al.* 2007). The predicted ozone concentrations in 2006 are underestimated but in an acceptable range.

Conclusions

This study used ozone observation at the three sites in southern Sweden and the six grouped LWTs (A, C, W, SEE, SWS, N+) centered in the southern Sweden to examine the influence of synoptic circulation patterns on surface ozone concentration. The synoptic circulation is expressed by the intensity and frequencies of LWTs. The main results are summarized as follows:

1. The Lamb circulation patterns A, SEE and SWS were closely associated with regionally elevated ozone concentrations, while the Lamb circulation pattern W was often associated with low ozone levels.
2. Local climate and topography most likely resulted in spatial differences in ozone concentrations between the monitoring sites under same circulation pattern. Under the condition of anticyclonic weather, the altitude effect at Norra Kvill and Vavihill could in part explain the spatial ozone difference.
3. Among the six Lamb indices, the total vorticity index ζ was to a large extent able to explain the influence of high/low pressure system on surface ozone and seems to constitute a key index for the description of synoptic impact on surface ozone.
4. The annual mean ozone concentrations over the three sites were reconstructed by the use of the linear regression models with the frequencies and indices of LWTs as predictors. The models, explaining 85% and 71% of the observed ozone concentrations in spring and summer, respectively, are able to reasonably well estimate the inter-annual variability of the regional ozone concentrations.

Acknowledgements: The authors thank Professors Øystein Hov, Peringe Grennfelt and Håkan Pleijel for constructive comments on the primary research. Discussions with Hans Linderholm and Christine Achberger on ozone topics are also acknowledged. The Adlerbertska Research Fund, Wilhelm and Martina Lundgrens Scientific Fund are thanked for their support to Lin Tang. The Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning (FORMAS), the Swedish Research Council (VR), and Tellus are thanked for their support to Deliang Chen. The contribution by Per Erik Karlsson was supported by the programmer Abatement Strategies for Transboundary Air pollution (ASTA) funded by the Foundation for Strategic Environmental Research (MISTRA). The ozone monitoring at Rörvik/Råö, Norra Kvill and Vavihill was funded by the Swedish Environmental Protection Agency. Also, we would like very much to express our great thanks to the editor and referees for valuable comments and constructive suggestions to improve the manuscript.

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