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A method to assess the inter-annual weather-dependent variability in air pollution concentration and deposition based on weather typing

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HIGHLIGHTS

- Yearly anomalies in air pollution were explained by variation in Lamb Weather Types.
- A novel method to assess annual anomalies in air pollution is suggested.
- Adjusting for anomalies improved significance of temporal trends in air pollution.
- Most pollutants showed no trend or a negative trend but urban ozone had a positive trend.

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ABSTRACT

Annual anomalies in air pollutant concentrations, and deposition (bulk and throughfall) of sulphate, nitrate and ammonium, in the Gothenburg region, south-west Sweden, were correlated with optimized linear combinations of the yearly frequency of Lamb Weather Types (LWTs) to determine the extent to which the year-to-year variation in pollution exposure can be partly explained by weather related variability. Air concentrations of urban NO_2, CO, PM_{10}, as well as O_3 at both an urban and a rural monitoring site, and the deposition of sulphate, nitrate and ammonium for the period 1997–2010 were included in the analysis. Linear detrending of the time series was performed to estimate trend-independent anomalies. These estimated anomalies were subtracted from observed annual values. Then the statistical significance of temporal trends with and without LWT adjustment was tested. For the pollutants studied, the annual anomaly was well correlated with the annual LWT combination (R^2 in the range 0.52–0.90). Some negative (annual average [NO_2], ammonia bulk deposition) or positive (average urban [O_3]) temporal trends became statistically significant ($p < 0.05$) when the LWT adjustment was applied. In all the cases but one (NH_4 throughfall, for which no temporal trend existed) the significance of temporal trends became stronger with LWT adjustment. For nitrate and ammonium, the LWT based adjustment explained a larger fraction of the inter-annual variation for bulk deposition than for throughfall. This is probably linked to the longer time scale of canopy related dry deposition processes influencing throughfall being explained to a lesser extent by LWTs than the meteorological factors controlling bulk deposition. The proposed novel methodology can be used by authorities responsible for air pollution management, and by researchers studying temporal trends in pollution, to evaluate e.g. the relative importance of changes in emissions and weather variability in annual air pollution exposure.

1. Introduction

Air pollution exposure is in many cases highly temporally variable. Emission rates, (photo)chemical activity and dispersion depend on meteorological conditions (Arain et al., 2007; Jones et al., 2010; Pleijel et al., 2004). Hence, the meteorological conditions strongly affect the short-term variability in the concentration of key air pollutants such as nitrogen oxides ($NO_x = NO_2 + NO$), particulate matter (PM), carbon monoxide (CO) and ozone (O_3) as

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well as the deposition of sulphate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+) and base cations.

Even on an annual time scale, there is a considerable variation in air pollution levels. This may pose a problem for managers and policy makers responsible for air pollution monitoring and control regarding e.g. assessments of the results of emission reductions. When the air quality standards (AQS) are met in one year, it may be concluded that the situation is fine and further action to abate emissions are not required. The following year, there is once again exceedance of AQS, which may lead to the conclusion that this is caused by increasing emissions. However, in addition to possibly existing upward or downward time series trends related to emissions, a large part of the inter-annual variability in air pollution concentrations may be the result of the variation in the pattern of weather conditions characterizing a particular year. Thus the yearly anomaly in air pollution concentration would to a certain extent be a function of a considerable year-to-year variation in those conditions (e.g. dispersion, photochemistry and precipitation) promoting high or low air pollution concentrations or deposition. The annual time unit, which is most commonly used when evaluating AQSs, is therefore too short to evaluate effects of abatement measures and trends in pollutants, since large year-to-year variation occur depending on the weather profiles. However, adjustments for the influence of inter-annual variation in weather, if this influence can be quantified, may enhance the detection of temporal trends in air pollution concentration and deposition caused by long term changes in emissions.

Similar to air pollution concentrations, deposition of compounds such as SO_4^{2-}, NO_3^- and NH_4^+, presents substantial inter-annual variation, possibly superimposed on temporal trends caused by emission changes. The deposition of these compounds is mostly monitored on a monthly basis. Acidification of forest soils and surface waters remain a serious problem in south-west Sweden (Akselsson et al., 2013; Sverdrup et al., 2005). Furthermore, there is a substantial risk of N leaching from forest soils (Akselsson et al., 2010). Hence, it is important to assess the changes of both sulphur and nitrogen deposition over time. The deposition of sulphur and nitrogen to Norway spruce forests at northern latitudes is dominated by wet deposition. In southwest Sweden, the share of dry deposition constitute approximately 30% of the total deposition of inorganic nitrogen (Karlsson et al., 2011), while in northern Sweden this share is close to zero. Approximately the same applies to sulphur deposition.

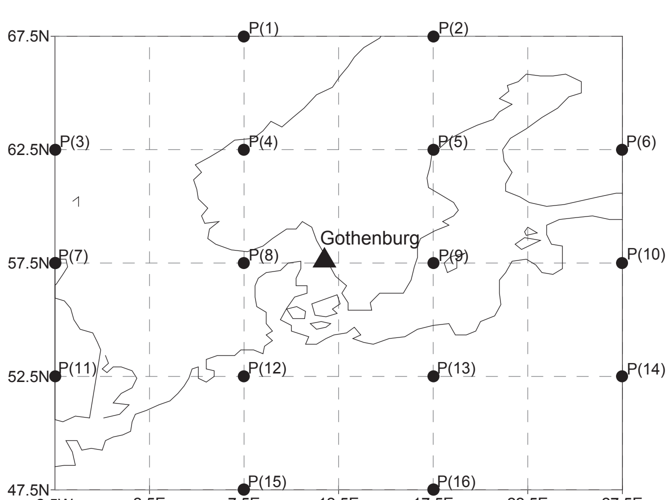

Fig. 1. Map showing the atmospheric pressure grid points used to calculate the Lamb Weather Types.

Grundström et al. (2015) showed that the urban concentrations of NO_2 are strongly influenced by the prevailing weather types for Gothenburg, south-west Sweden, during the winter. Similarly, Tang et al. (2009) showed that ozone concentrations in south Sweden are linked to weather types. Both these studies used Lamb Weather Types (LWTs), which represent an efficient tool for characterising local meteorological conditions based on synoptic scale sea level pressure (Chen, 2000). The distribution of the sea level pressure provides information about the synoptic air mass movement and vorticity, which has been proven to be a useful summary of the local meteorological conditions (Chen, 2000; Demuzere and van Lipzig, 2010; Grundström et al., 2015), based on a set of objective rules (Jenkinson and Collison, 1977). LWTs provide an effective way to classify the prevailing local weather from the regional directional flow of air masses. Similarly, Buchholz et al. (2010) used an air pollution index together with so called Grosswetterlagen (GWL), weather types for Central Europe, to identify situations with high air pollution levels. LWT and GWL schemes both started as subjective schemes for weather typing (Lamb, 1950), but have been developed into objective schemes that can be automated (James, 2007). The GWL scheme represents the weather pattern on a continental scale prevailing for several consecutive days i.e. differing from the LWTs which are normally defined with a higher resolution (regional and daily to sub-daily). More recently, Pope et al. (2014) and Russo et al. (2014) used similar weather classification schemes to successfully identify situations with large/small potential for high air pollution concentrations. Finally, Zhang et al. (2016) successfully used synoptic weather patterns to assess the effect of the East Asian Monsoon on the air quality over the North China Plains.

In this study, we suggest a novel method to quantitatively estimate the influence of the weather conditions represented, by Lamb Weather Types, on the annual anomalies of urban air concentrations of NO_2, PM_{10}, CO, urban and rural air concentrations of O_3, as well as the deposition of SO_4^{2-}, NO_3^- and NH_4^+ as throughfall (precipitation that passed through the forest canopy, TF) and bulk (deposition with the precipitation over open field, BD) deposition at a rural site in the Gothenburg region. The aim of our study was to provide an objective tool to assess the weather influence on annual anomalies in air pollution levels and to improve the possibility to detect temporal trends due to emission changes. Our hypotheses were that: 1. A large fraction of the inter-annual variability in air pollution concentrations can be explained by the frequency distribution of LWTs, 2. The trends for the air pollutants caused by emission changes is easier to be detected, when the LWT related annual anomalies are removed.

2. Methods

2.1. Data

Data were obtained from three monitoring sites. From the rooftop monitoring station Femman in central Gothenburg (30 m above street level, 57°42′N, 11°58′E) data regarding air concentrations of NO_2 (Tecan CLD 700 AL chemiluminescence), CO (Unor 610), O_3 (Monitor Labs 9810) and PM_{10} (Tapered Element Oscillating Microbalance, Series 1400b) were obtained. The Femman house is one of the tallest buildings in the City of Gothenburg and surrounding buildings are not likely to influence the flow of air pollutants. To compare the urban O_3 data with a nearby rural site, data from the Råö monitoring station (Thermo Environmental Model 49 UV monitor) of the EMEP (European Monitoring and Evaluation Programme) network (www.emep.int) 40 km S of Gothenburg (57°23′N, 11°54′E) was used. From the Hensbacka rural site 90 km N of Gothenburg (58°26′N, 11°44′E) data on BD and TF

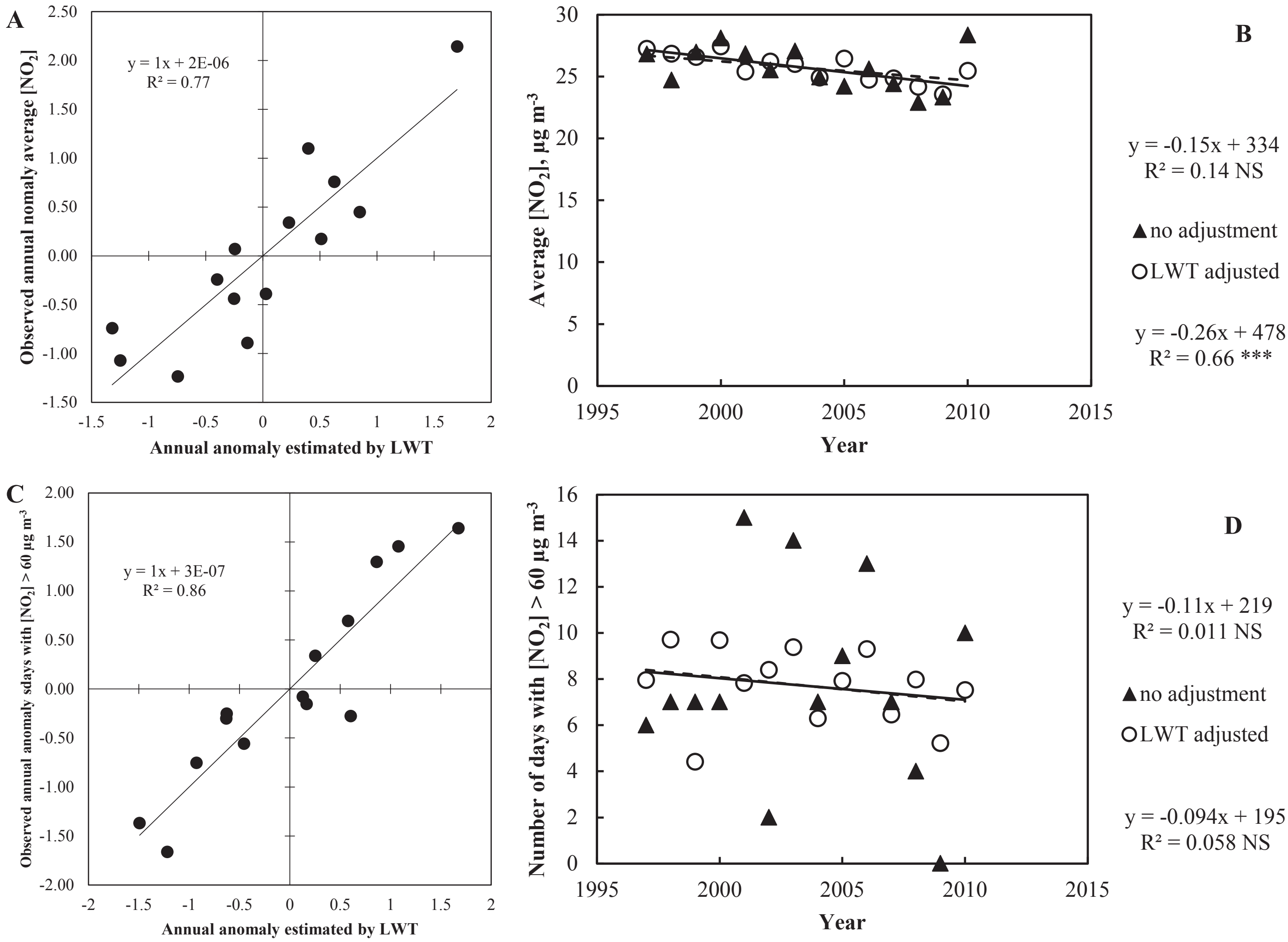

Fig. 2. A: Observed annual anomaly vs. the corresponding annual anomaly resulting from the optimized LWT combination for average [NO_2] measured at the monitoring station Femman, B: Temporal trend for observed and LWT adjusted average [NO_2], C: Observed annual anomaly vs. the anomaly resulting from the optimized LWT combination for days with average [NO_2] > 60 µg m^{-3}, D: Temporal trend for observed and LWT adjusted days with average [NO_2] > 60 µg m^{-3}. Broken line, no adjustment; solid line, LWT adjusted.

deposition of sulphate, nitrate and ammonium were retrieved. The Hensbacka site forms part of the Swedish Throughfall Monitoring (SWETHRO) network (Pihl Karlsson et al., 2011). This is an environmental monitoring network that measures air concentrations of pollutants, deposition and soil water chemistry at forest sites in Sweden. Monitoring sites are positioned in closed, mature, managed forests with no major roads or other pollution sources in the vicinity. The methods for the TF and BD deposition are described in detail in Pihl Karlsson et al. (2011).

For NO_2, the annual average (µg m^{-3}) and the number of hours >200 µg m^{-3} (both connected to EU legislation target values, http://ec.europa.eu/environment/air/quality/standards.htm), as well as the annual number of days > 60 µg m^{-3} and the number of hours >90 µg m^{-3} (domestic AQS target values in Sweden, http://www.naturvardsverket.se/upload/in-english/legislation/the-swedish-environmental-code/environmental-quality-standards/air-quality-ordinance-sfs-2010-477.pdf), were evaluated. In the case of PM_{10} and CO the annual average was evaluated. For ozone the annual growing season (April–September) average concentrations (ppbv) were used for the urban and the rural site.

Meteorological data obtained from the rooftop monitoring station Femman were atmospheric pressure (Vaisala PA11A), air temperature and relative humidity (Campbell Rotronic MP101 thermometer/hygrometer), wind speed and wind direction (Gill ultrasonic anemometer), solar radiation (Skye SKL2650) and precipitation (Tipping Bucket – Casella cell).

2.2. Lamb Weather Types

Daily mean sea level pressure (MSLP) for a 16 point-grid (Fig. 1) centred over the Gothenburg city centre (57°7′N, 11°97′E), were obtained from the NCEP/NCAR Reanalysis database 2.5 × 2.5° pressure fields (Kalnay et al., 1996). Circulation indices, u (westerly or zonal wind), v (southerly or meridional wind), V (combined wind strength), ξ_u (meridional gradient of u), ξ_v (zonal gradient of v) and ξ (total shear vorticity) describing the geostrophic winds and Lamb Weather Types (Jenkinson and Collison, 1977) were calculated following Chen (2000). This classification scheme has 26 weather types: anticyclone (A), cyclone (C), eight directional types (NE, E, SE, ...), 16 hybrid types (ANE, AE, ASE, CNE, CE, CSE, ...). In this study, the 26 weather types were consolidated into 10 LWTs according to the directions of the geostrophic wind, eight directional: NE, E, SE, S, SW, W, NW, N, and two rotational: A and C.

2.3. Data analysis

To account for the obviously large year-to-year variation in air pollution concentration/deposition a detailed analysis of anomalies

Table 1
Statistics related to the analysis of yearly anomalies of different air pollution variables. LWT, Lamb Weather Type; SSR, sum of squares of residuals; NS, non-significant; *, p < 0.05; **, p < 0.01; ***, p < 0.001.

Air pollution variable	R^2 anomaly vs. LWT combination	Unadjusted SSR	LWT adjusted SSR	Ratio SSR adj/SSR LWT adj	Unadjusted p slope	LWT adjusted p slope	Unadjusted sign level	LWT adjusted sign level	Direction of trend
NO_2 average	0.773	32.3	6.1	0.19	0.183	0.000	NS	***	negative
NO_2 days>60	0.862	236.3	32.6	0.14	0.726	0.408	NS	NS	
NO_2 hours>90	0.753	12022.7	2968.7	0.25	0.570	0.245	NS	NS	
NO_2 hours>200	0.613	80.0	28.0	0.35	0.940	0.337	NS	NS	
PM_{10} average	0.74	96.6	24.4	0.25	0.569	0.107	NS	NS	
O_3 average Urban	0.843	463.2	71.9	0.16	0.158	0.001	NS	**	positive
O_3 average Rural	0.756	62.1	15.1	0.24	0.783	0.506	NS	NS	
CO average	0.904	40407.8	3453.0	0.09	<0.001	<0.001	***	***	negative
SO_4 bulk deposition	0.776	3.8	0.8	0.22	<0.001	<0.001	***	***	negative
NO_3 bulk deposition	0.84	6.7	1.1	0.16	0.010	<0.001	**	***	negative
NH_4 bulk deposition	0.736	10.9	2.5	0.23	0.462	0.010	NS	*	negative
N_{tot} bulk deposition	0.708	26.7	8.0	0.30	0.067	<0.001	NS	***	negative
SO_4 throughfall	0.56	13.7	6.0	0.44	<0.001	<0.001	***	***	negative
NO_3 throughfall	0.683	13.1	4.0	0.31	0.037	<0.001	*	***	negative
NH_4 throughfall	0.523	12.7	6.0	0.48	0.842	0.958	NS	NS	
N_{tot} throughfall	0.583	45.3	18.7	0.41	0.199	0.103	NS	NS	

Table 2
Coefficients (k) of the optimized LWT combinations for different air pollution variables. A, anticyclonic; C, cyclonic; NE, north-east; E, east … .

Air pollution variable	A	NE	E	SE	S	SW	W	NW	N	C
	k_A	k_N	k_{NE}	k_E	k_{SE}	k_S	k_{SW}	k_W	k_{NW}	k_C
NO_2 average	11.2	19.3	−16.0	−14.7	37.4	−24.3	−10.5	16.3	−55.6	9.0
NO_2 days>60	9.1	−23.4	−7.5	−25.0	12.2	−9.6	−18.2	10.7	−11.6	20.0
NO_2 hours>90	3.7	−35.7	1.5	−17.6	3.5	3.0	−23.2	25.6	−17.6	13.8
NO_2 hours>200	5.7	−49.4	−1.3	−11.4	6.5	−0.5	−21.3	10.8	5.8	15.0
PM_{10} average	1.9	−74.0	30.2	9.8	−24.6	26.8	−25.3	19.0	14.3	−2.6
O_3 average Urban	14.5	−33.3	−8.8	−6.3	−2.8	0.5	−16.5	19.1	−17.2	0.2
O_3 average Rural	12.8	−38.2	9.2	9.3	−16.9	13.8	−26.3	11.4	2.7	−4.2
CO average	−6.5	17.0	8.6	3.1	4.1	−1.9	4.7	−15.3	14.7	2.2
SO_4 bulk deposition	−4.4	−16.8	8.7	−1.9	−3.7	7.5	1.2	5.2	−0.8	−1.8
NO_3 bulk deposition	−3.9	−6.1	−2.4	−8.2	4.4	9.6	10.2	−10.2	−6.4	−0.9
NH_4 bulk deposition	−7.7	−15.0	16.8	−7.6	−32.3	37.4	−8.1	17.4	13.3	−12.9
N_{tot} bulk deposition	2.7	−7.6	3.6	−16.5	−5.4	19.1	1.1	−0.7	−18.0	−0.9
SO_4 throughfall	1.2	−11.5	3.8	−6.5	−12.5	13.8	−2.7	−5.9	9.4	1.4
NO_3 throughfall	3.1	−12.7	7.9	−5.3	−25.3	29.8	−6.2	−7.4	23.3	−9.2
NH_4 throughfall	6.3	7.4	−25.0	−2.4	−14.4	19.7	−5.8	−3.4	3.4	−4.1
N_{tot} throughfall	4.0	−21.1	6.4	−2.6	−20.9	31.7	−10.8	−1.7	12.6	−5.0

was made. The annual average anomaly z_i and air pollutant concentration/deposition level (C_i) for year i was defined as (Grumm and Hart, 2001):

$$z_i = \frac{(C_i - x_C)}{\sigma_C} \tag{1}$$

where x_c and σ_c is the full period (1997–2010) air pollutant concentration/deposition level mean and standard deviation, respectively. Linear detrending of the annual data was made to estimate trend-independent anomalies; overarching trends over the period were thus assumed to be the result of changes in emissions, possibly with some influence from long-term climate change and altered atmospheric chemistry. The time fraction (f) of each of the ten LWTs was calculated for each year. The estimated annual anomaly caused by variation in the frequency of different LWTs, z_{i_LWT}, was represented by a linear combination of the time fractions of the different LWTs of year i:

$$z_{i\_LWT} = k_A f_A + k_N f_N + k_{NE} f_{NE} + \ldots + k_{NW} f_{NW} + k_C f_C \tag{2}$$

Numerical optimization of the coefficients k_A, k_N, k_{NE} … was made separately for each pollutant index to minimize the deviation between observed z_i, and z_{i_LWT}, using the least square approach, over the study period. Then the observed anomaly z_i was regressed vs. z_{i_LWT} representing the estimated LWT contribution to the anomaly, to evaluate how much of the variation in z_i that could be explained by the linear combination of annual LWT time fractions. The relationship was evaluated using linear regression with respect to the coefficient of determination (R^2).

Further, time series of observed annual concentrations/depositions were compared to time series of LWT adjusted annual values. This was made by multiplying the z_{i_LWT} with σ_C and subtracting the resulting value from C_i. The observed and LWT adjusted

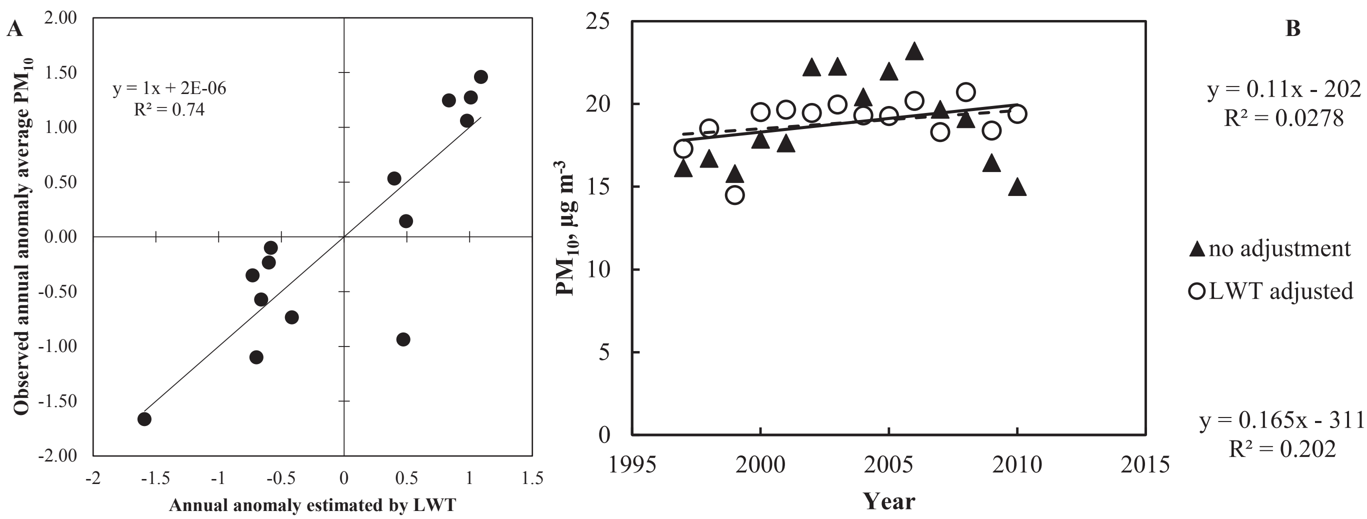

Fig. 3. A: Observed annual anomaly vs. the corresponding annual anomaly resulting from the optimized LWT combination for average [PM_{10}] measured at the monitoring station Femman, B: Temporal trend for observed and LWT adjusted average [PM_{10}]. Broken line, no adjustment; solid line, LWT adjusted.

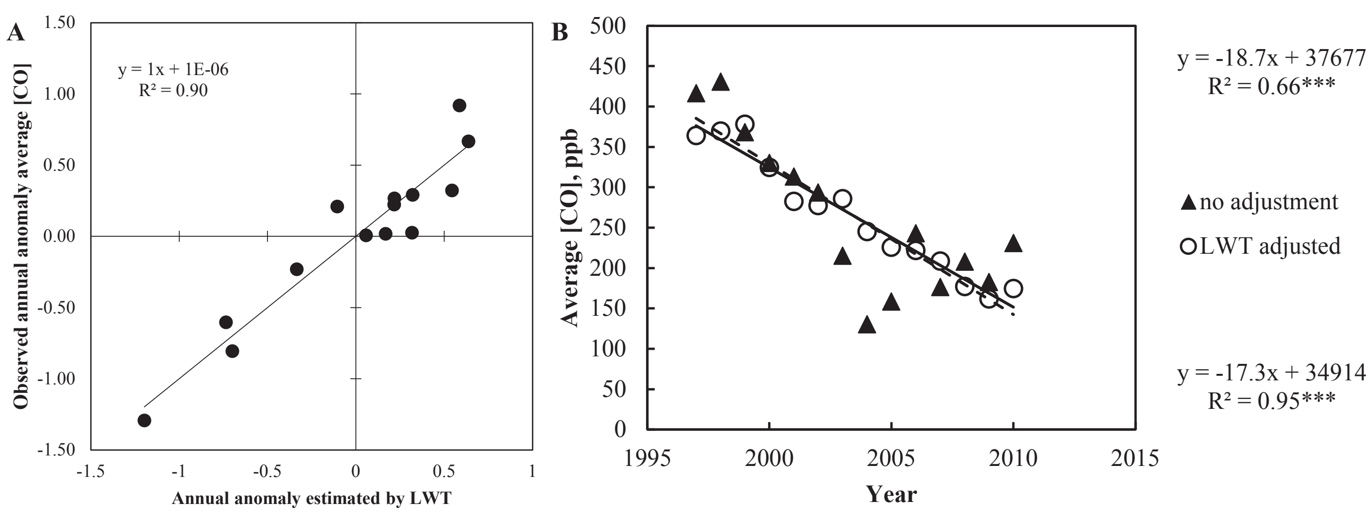

Fig. 4. A: Observed annual anomaly vs. the corresponding annual anomaly resulting from the optimized LWT combination for average [CO] measured at the monitoring station Femman, B: Temporal trend for observed and LWT adjusted average [CO]. Broken line, no adjustment; solid line, LWT adjusted.

time series were evaluated by R^2, the sum of squares of residuals (SSR, quantifying to what extent the data deviate from the regression line) and the statistical significance of the regression slope.

3. Results

3.1. NO_2

The optimized LWT combination explained 77% of the variation in the yearly average [NO_2] anomaly at the monitoring station Femman (Fig. 2A, Table 1). There was a weak declining temporal trend in the annual average [NO_2], which was not statistically significant for the unadjusted data, but highly significant ($p < 0.001$) for LWT adjusted data (Fig. 2B, Table 1). An even stronger relationship between observed and the optimized LWT combination ($R^2 = 0.86$) was obtained for the number of days with daily [NO_2] > 60 µg m^{-3} (Fig. 2C, Table 1), but for this variable there was no significant temporal trend (Fig. 2D, Table 1). However, the interannual variability as expressed by the SSR was strongly reduced when comparing unajdusted with LWT adjusted data also for this variable (Table 1). Similar results as for [NO_2] > 60 µg m^{-3} were found for the exceedance of hourly values of 90 and 200 µg m^{-3} (presented in Tables 1 and 2).

We compared the coefficients presented in Table 2 for the four different indices used for NO_2. The correlation was not very strong between LWT coefficients (values of k) for average [NO_2] and daily [NO_2] > 60 µg m^{-3} ($R^2 = 0.24$). It was much stronger when correlating LWT coefficients for the NO_2 exposure indices including a concentration threshold. The LWT coefficients for daily [NO_2] > 60 µg m^{-3} correlated strongly with those for hourly [NO_2] > 90 µg m^{-3} ($R^2 = 0.72$) and fairly strongly also with those for hourly [NO_2] > 200 µg m^{-3} ($R^2 = 0.59$).

3.2. PM_{10}

As shown in Fig. 3A and Table 1, the optimized linear combination of LWTs explained 74% of the variation of the annual anomalies in PM_{10}. For this pollutant a weak positive temporal trend was observed (Fig. 3B), which was not statistically significant (Table 1), neither for unadjusted data, nor for LWT adjusted data.

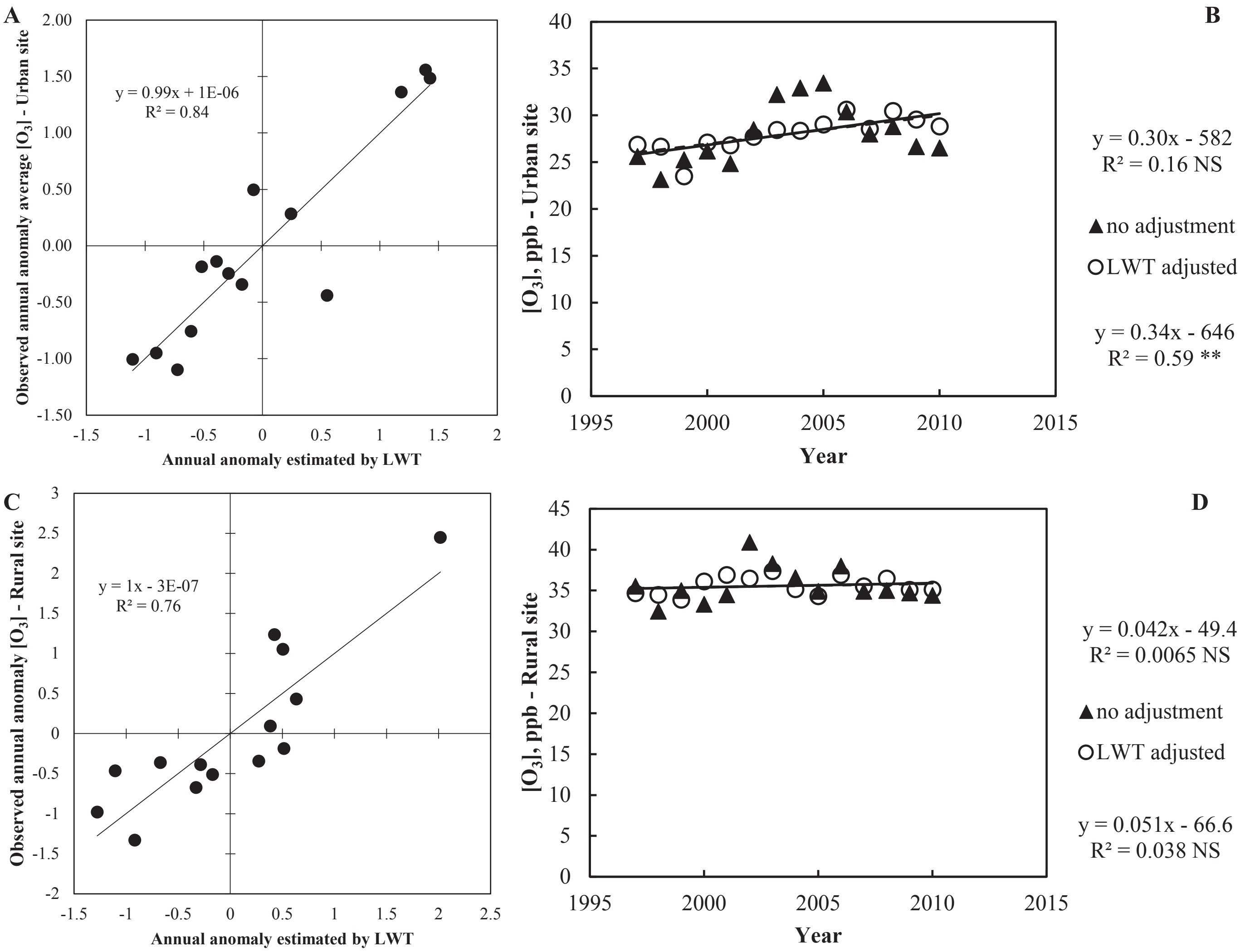

Fig. 5. A: Observed annual anomaly vs. the corresponding annual anomaly resulting from the optimized LWT combination for average urban $[O_3]$ measured at the monitoring station Femman, B: Temporal trend for observed and LWT adjusted average urban $[O_3]$, C: Observed annual anomaly vs. the anomaly resulting from the optimized LWT combination for average rural $[O_3]$ measured at the rural monitoring station Råö, D: Temporal trend for observed and LWT adjusted days with average rural $[O_3]$. Broken line, no adjustment; solid line, LWT adjusted.

However, the SSR was strongly reduced, from 96.6 to 24.4 (Table 1), by the LWT adjustment.

3.3. CO

For CO, the optimised linear combination of LWTs was highly efficient in explaining the annual anomalies in average concentration ($R^2 = 0.90$, Fig. 4A, Table 1). The temporal trend for CO was steeply declining (Fig. 4B) and strongly statistically significant for unadjusted data as well as for LWT adjusted data. The SSR was, however, reduced by more than a factor of ten, when comparing unadjusted and LWT adjusted values (Table 1).

3.4. O_3

At the urban site, annual $[O_3]$ anomalies were explained by the optimized LWT linear combination by 84% (Fig. 5A, Table 1). Here, there was an increasing temporal trend for $[O_3]$ which was statistically significant for LWT adjusted data but not for non-adjusted (Fig. 5B, Table 1). The SSR was reduced by more than a factor of five for LWT adjusted data compared with non-adjusted (Table 1).

At the rural site, the optimized LWT linear combination explained 76% of the variation in annual anomaly of $[O_3]$ (Fig. 5C, Table 1). Unlike the urban site there was no indication of any temporal trend at this site (Fig. 5D, Table 1). LWTs A and NW promoted both urban and rural $[O_3]$ while LWTs W and NE were associated with low $[O_3]$ (Table 2).

3.5. Deposition of sulphate, nitrate and ammonium

Regarding SO_4^{2-}, the annual anomaly was explained by the LWT model by 78% and 56%, respectively, for BD and TF deposition (Fig. 6A and C, Table 1). The declining temporal trend was very strong for SO_4^{2-}, and strongly statistically significant both for BD and TF deposition (Figs. 6B and D, Table 1).

For NO_3^-, the annual anomaly was explained by the LWT model by 84% and 68%, respectively, for open-field and TF deposition (Fig. 7A and C, Table 1). The declining trend was weaker than for SO_4^{2-}, but still statistically significant both for BD and TF deposition (Fig. 7B and D, Table 1). It should be noted that in general the deposition values for NO_3^- are lower for TF as compared to BD, which is explained by the fact that the tree canopies directly take

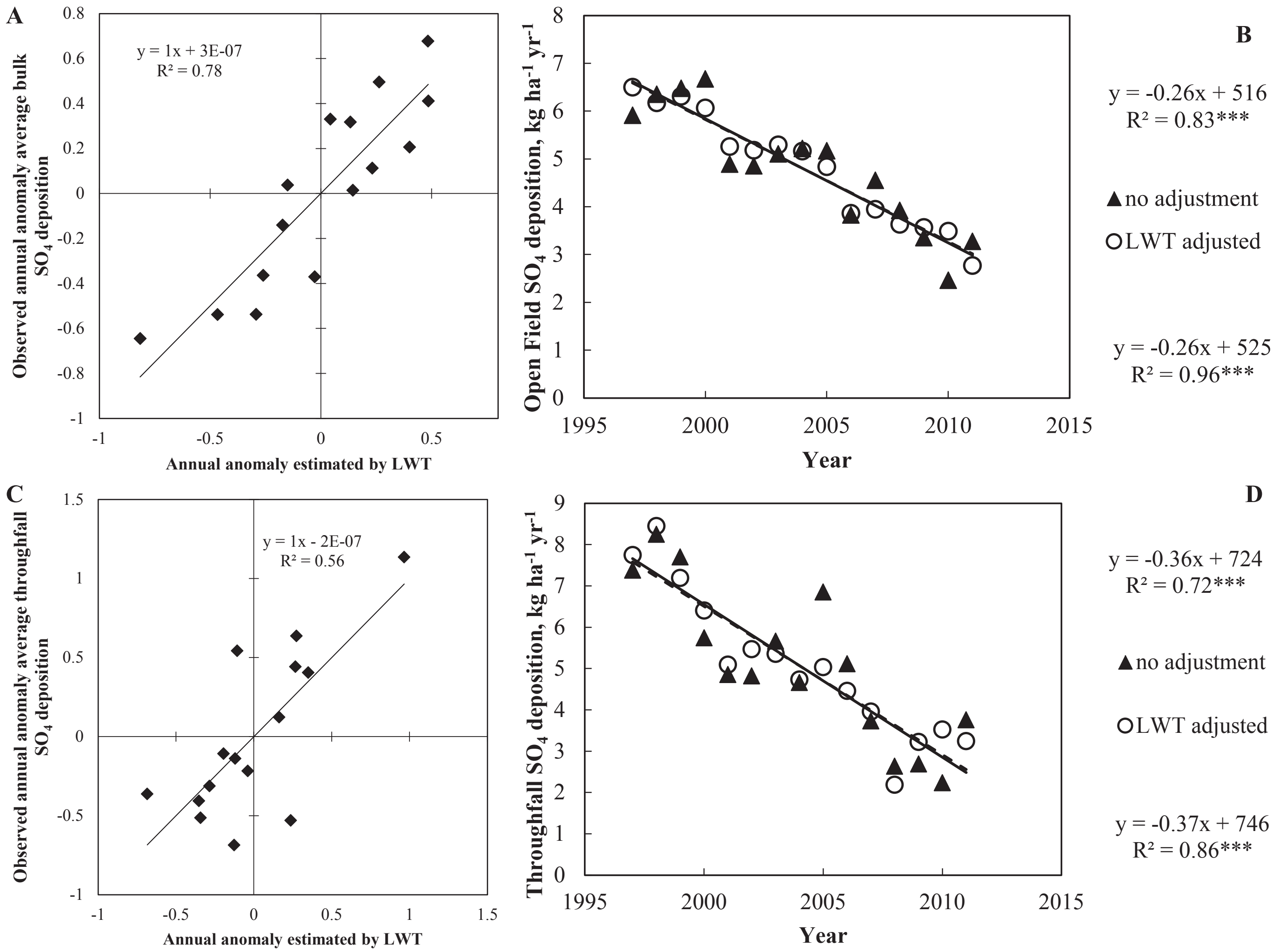

Fig. 6. A: Observed annual anomaly vs. the corresponding annual anomaly resulting from the optimized LWT combination for sulphate open-field deposition measured at the rural monitoring station Hensbacka, B: Temporal trend for observed and LWT adjusted open-field sulphate deposition, C: Observed annual anomaly vs. the anomaly resulting from the optimized LWT combination for sulphate throughfall deposition, D: Temporal trend for observed and LWT adjusted days with sulphate throughfall deposition. Broken line, no adjustment; solid line, LWT adjusted.

up a fraction of the deposited NO_3^- and NH_4^+ (e.g. Ferm, 1993).

In the case of NH_4^+, the annual anomaly was explained by the LWT model by 74% and 52%, respectively, for BD and TF deposition (Fig. 8A and C, Table 1). The bulk NH_4^+ deposition was weakly declining. The temporal trend was statistically significant when using LWT adjusted data, but non-significant for data not LWT adjusted (Fig. 8B, Table 1). For TF NH_4^+ data, there was no indication for a temporal trend, neither for non-adjusted nor for LWT adjusted data (Fig. 8D, Table 1).

It can be noted that for all three ions the LWT linear combination model provided stronger results (R^2 for the anomaly vs. LWT linear combination higher) for BD data as compared with TF (Table 1). This is also highlighted by the smaller reduction in SSR by applying the LWT adjustment for TF as compared to BD (Table 1). From Table 2 it can be inferred that the LWT SW promoted large deposition of sulphate, nitrate and ammonium, both for BD and TF deposition. As evident from Table 3, the LWT SW had the clearly highest precipitation. For the remaining LWTs the result was more complex.

It can be observed for TF of sulphate, nitrate as well as ammonium that there was a large positive anomaly for the year 2005 (Figs. 6D, 7D and 8D). In January 2005 there was a major storm affecting south Sweden with very high deposition values observed. Due to the storm some sampling equipment was damaged and the total sampling volume had to be estimated. Undoubtedly, the deposition during this month with the storm event was very high, but there is some uncertainty around the exact values of the depositions during this month. However, it seems that the LWT adjustment worked well also for 2005 (Figs. 6D, 7D and 8D) and the uncertainty of estimations of BD and TF for January 2005 is not likely to have affected the assessment for 13-year period studied.

3.6. Weather patterns in different LWTs

In Table 3 the weather patterns of the different LWTs during the study period is summarised. The table shows that there was a considerable variation in different meteorological variables between LWTs. For example, C and southern/western LWTs showed low average atmospheric pressure while A and eastern LWTs exhibited high atmospheric pressure. LWT A had the lowest average wind speed while rainfall was by far highest in LWT SW. N was the coldest LWT, while SW, W, S and C had the highest relative humidity. Solar radiation also varied considerably between LWTs. This variable is of importance for promoting ozone formation and

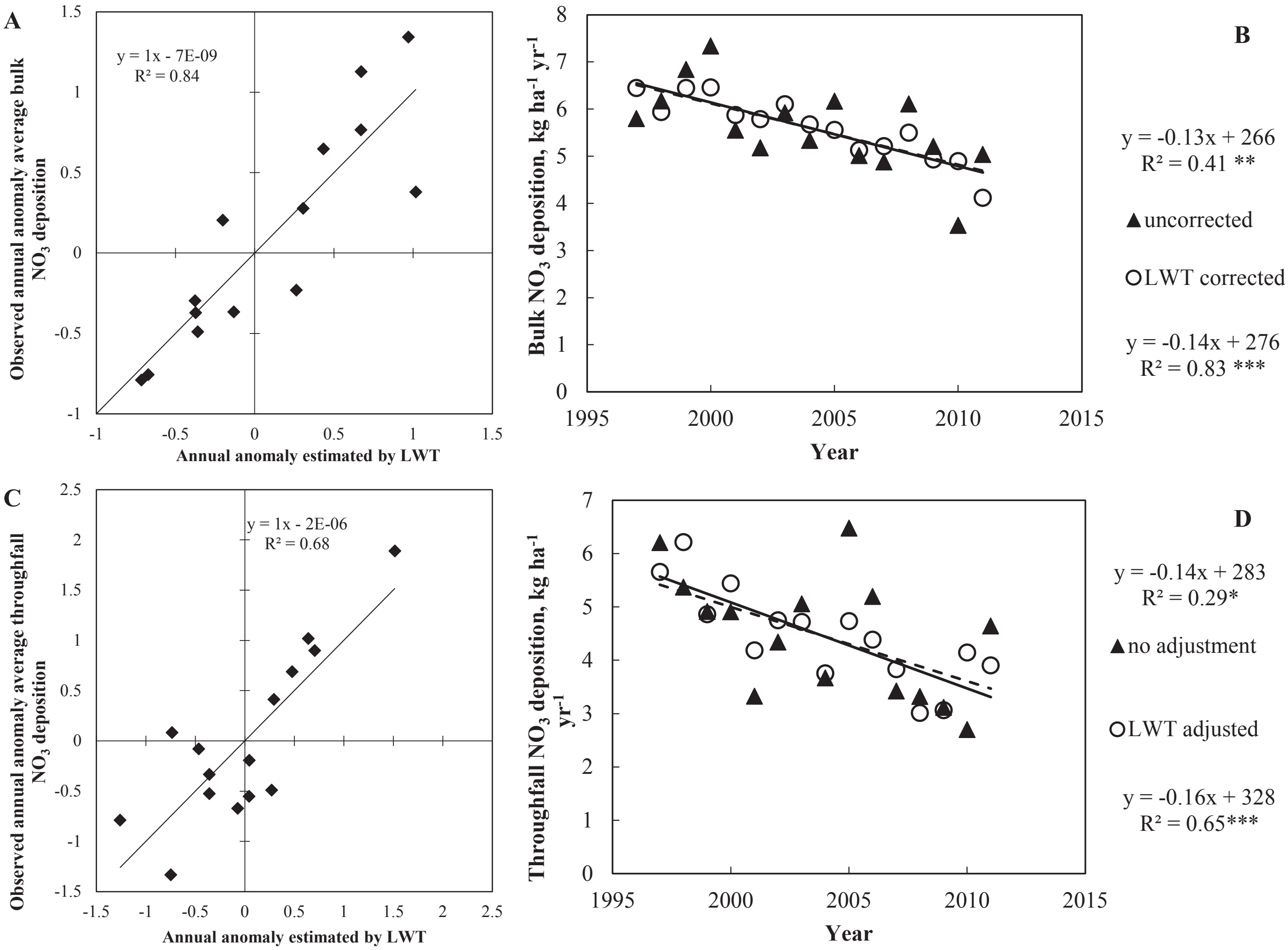

Fig. 7. A: Observed annual anomaly vs. the corresponding annual anomaly resulting from the optimized LWT combination for nitrate open-field deposition measured at the rural monitoring station Hensbacka, B: Temporal trend for observed and LWT adjusted open-field nitrate deposition, C: Observed annual anomaly vs. the anomaly resulting from the optimized LWT combination for nitrate throughfall deposition, D: Temporal trend for observed and LWT adjusted days with nitrate throughfall deposition. Broken line, no adjustment; solid line, LWT adjusted.

was highest in LWTs A, E, N and NW.

4. Discussion

This study showed that a large fraction of the year-to-year variation in air pollution concentrations/deposition can be explained by the year-to-year variation in weather conditions as expressed by the ten LWTs. When studying temporal trends for the pollution/deposition the SSR was strongly reduced in all cases and in three cases (average [NO_2], urban [O_3] and BD of ammonium) trends which were non-significant for unadjusted data became statistically significant for LWT adjusted data. For all but one (TF NH_4^+ deposition) pollution variable, the temporal trends became more strongly significant for LWT adjusted data. Thus, the approach presented in this paper is useful to study the strength and significance of temporal trends in pollution concentrations and deposition that are related to emission changes.

In the case of O_3 the rural concentrations were higher than the urban, which is expected from the titration of O_3 with NO occurring in the urban environment (Clapp and Jenkin, 2001). Interestingly, the urban [O_3], unlike rural, showed a significant increase, which was paralleled by a significant decline in [NO_2]. A similar pattern has been observed in several European and North American cities (Paoletti et al., 2014) and is most likely the result of reduced local NO_x emissions, which leads to less O_3 titration by NO and a converging trend for rural and urban [O_3].

The strong decline in [CO] and sulphate deposition, both BD and TF, is easily explained by reduced emissions. The three-way catalyst system of vehicles is very efficient in removing CO and this has led to a strong CO decline in many cities, for example in Helsinki (Anttila and Tuovinen, 2010). The Gothenburg Protocol has resulted in very large emission reductions for sulphur in Europe (Gauss et al., 2014).

The LWT adjustment was less efficient in explaining the inter-annual variation in TF deposition of SO_4^{2-}, NO_3^- and NH_4^+ as compared to BD. There are some basic differences in how BD and TF depend on meteorological conditions. BD consists mainly of wet deposition and it is strongly dependent on precipitation rates. Furthermore, the raindrops accumulate pollutants during the air mass movements over relatively long time periods and over long geographical distances. There is no influence of the forest canopies on the BD. It appears that the variation in the factors determining the wet deposition is relatively well described by LWTs. TF on the other hand includes also the contribution from dry deposition,

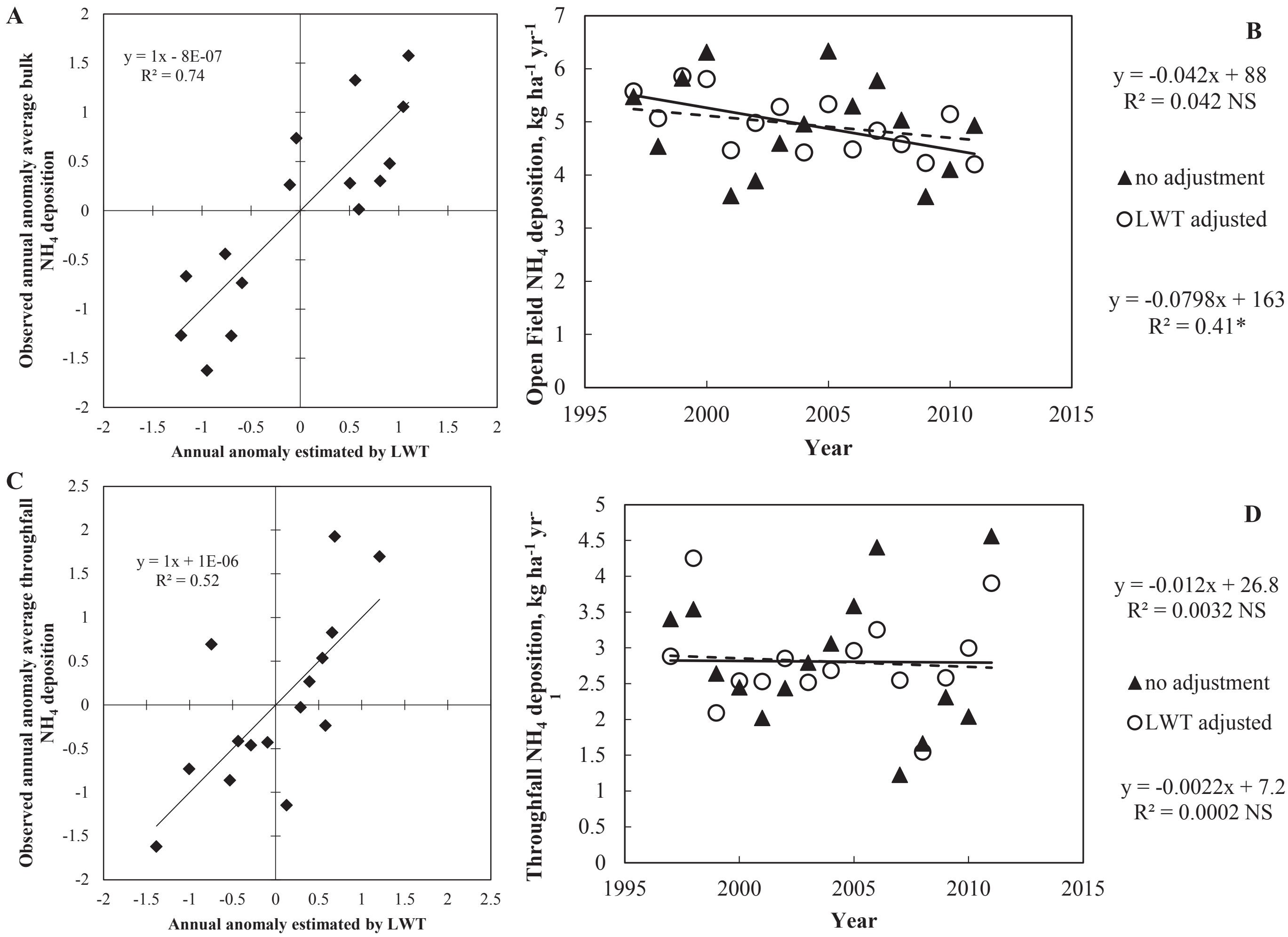

Fig. 8. A: Observed annual anomaly vs. the corresponding annual anomaly resulting from the optimized LWT combination for ammonium open-field deposition measured at the rural monitoring station Hensbacka, B: Temporal trend for observed and LWT adjusted open-field ammonium deposition, C: Observed annual anomaly vs. the anomaly resulting from the optimized LWT combination for ammonium throughfall deposition, D: Temporal trend for observed and LWT adjusted days with ammonium throughfall deposition. Broken line, no adjustment; solid line, LWT adjusted.

which depends on the local concentrations of gases and particles at the forest stand in combination with local meteorological factors such as wind speed and air turbulence. Furthermore, there is a strong influence of the forest canopy on the TF. Some of the deposition can occur as snow. Since data were used on a calendar year basis, snow can remain on the trees from one year to the next. Part of the wet deposition is during warmer weather conditions intercepted and water is evaporated back to the atmosphere. Hence, deposited compounds can remain in the canopy for relatively long time and thus interact with the canopies before they are washed off. A substantial fraction of the nitrogen deposition to forests can be taken up directly by the canopies (Adriaenssens et al., 2012) and thus not reaching the collectors for TF at the ground. It is not known to which extent this nitrogen is taken up by the tree shoots or by the epiphytes living on them, but in both cases the uptake rates may depend on the local conditions, which are not well reflected by the LWTs. There is however no evidence for direct canopy uptake of sulphur, so this aspect does not explain why LWTs were less efficient in explaining the inter-annual variation in TF deposition, compared to BD, of SO_4^{2-}.

It should be noted that LWTs do not only represent the direction of transport of pollutants, but to a large extent weather patterns associated with synoptic scale atmospheric pressure dynamics, which in turn affect precipitation, wind speed and direction, atmospheric stability and temperature (Table 3; Chen, 2000; Grundström et al., 2015). This is both an advantage and a

Table 3
Meteorological variables (averages) for the different Lamb Weather Types during the study period: P, atmospheric pressure; RH, relative humidity; T, atmospheric temperature; u, wind speed; R, global solar radiation; prec, precipitation; A, anticyclonic; C, cyclonic; NE, north-east; E, east… . The percentage of data occurrence, for each meteorological variable, recorded in the time period (1997–2010), has been presented in the bottom row.

LWT	P, hPa	RH, %	T, °C	u, m s^{-1}	R, W m^{-2}	prec, mm day^{-1}
A	1022	73.6	8.3	2.5	144.1	0.34
NE	1015	73.3	5.0	3.8	119.6	0.61
E	1015	71.7	7.6	4.1	131.3	0.94
SE	1013	73.7	8.7	3.3	111.6	1.93
S	1008	80.6	8.8	3.6	79.8	3.74
SW	1005	85.7	9.0	4.3	65.5	6.48
W	1007	83.3	9.4	4.8	87.1	3.23
NW	1009	74.8	8.7	3.8	131.8	0.76
N	1013	71.8	6.7	2.9	136.5	0.37
C	997	84.5	8.4	3.5	84.8	3.94
% Data	100%	67%	100%	91%	60%	74%

limitation. The composition of weather patterns represented by LWTs efficiently encapsulates the prevailing mix of meteorological conditions, which is an advantage. On the other hand, it is a complex task to analyse the impacts of the LWTs in the optimised linear combinations shown in Table 2, since the positive and negative effects of different LWTs on different pollutants is the net effect of many meteorological influences associated with each LWT, and these influences change with season and depend on a whole set of interactions among various factors.

For some air pollutants, such as NO_2 and to a large extent PM_{10} and CO, local emissions more or less dominate the pollution levels, especially in urban environments and situations with high concentrations. Thus, the ventilation of the urban air is critical for the air pollution levels. Grundström et al. (2015) showed that the strong variation of NO_2 in different LWTs was associated with the ventilation in turn associated with wind speed of the different LWTs rather than associated transport direction. Similarly, Zhang et al. (2016) found a strong relationship between stagnant weather and stable conditions with high values of a ground-based air pollution index over a 34 year period. In the case of tropospheric ozone, the anticyclonic LWT A promoting ozone formation by modest ventilation and high photochemical activity (Tang et al., 2009), but transport of air masses containing abundant O_3 and its precursors from source areas may also be of large importance. Regarding sulphate, nitrate and ammonium deposition, transport direction may be of even larger importance, major sources being located mainly in the in directions from west over south to east (Gauss et al., 2014), but also other conditions promoting/reducing deposition, especially precipitation, may also be very important.

This study shows that the inter-annual variation in air pollution levels can be accounted for by applying adjustments based on an objective weather typing, in this case LWTs. This approach has, to our best knowledge, not been tested before. Thus, the methodology presented in this paper forms a novel methodology to assess annual anomalies in air pollution concentration/deposition. The proposed methodology can be used e.g. by authorities responsible for air pollution management, and by researchers studying temporal trends in pollution, to evaluate e.g. the relative importance of changes in emissions and weather variability in annual air pollution exposure.

Possibly the results can be improved by analysing the data by season, since LWTs may represent somewhat different meteorological conditions e.g. in winter compared to summer. Also, it would be possible to include the hybrid type LWTs, increasing the number of weather types from 10 to 28, and at the same time possibly resolving important variation within the ten consolidated LWTs. An approach including seasonality and hybrid type LWTs will make up a more complex model and the simpler model used in our study was largely very efficient in explaining observed anomalies in air pollution concentration and deposition.

5. Conclusions

The most important conclusions of this study were:

- The LWT system is able to account for a substantial fraction of the inter-annual variability of air pollutant concentrations/deposition, making it a useful tool for policy and scientific accounting for weather dependent year-to-year variation in air pollution. This supports hypothesis 1 and forms the basis of a novel and efficient method to assess the weather influence on annual anomalies in air pollution concentration/deposition.
- The significance of temporal trends in the pollution variables studied is increased after the weather dependent inter-annual variability has been removed, which provides an effective means to focus on the roles played by other processes such as long term emission changes, in line with hypothesis 2.
- Throughfall deposition was accounted for by LWT to a lesser extent than bulk deposition, which most likely points to an effect of dry deposition and some long-term processes influencing throughfall but not bulk deposition.

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