Atmospheric Pollution Research 5 (2014) 741-752

Atmospheric Pollution Research

www.atmospolres.com

Downscaling of monthly PM_{10} indices at different sites in Bavaria (Germany) based on circulation type classifications

Christoph Beck, Claudia Weitnauer, Jucundus Jacobeit

University of Augsburg, Institute of Geography, Alter Postweg 118, D–86135 Augsburg, Germany

ABSTRACT

Atmospheric circulation affects local concentrations of particulate matter with an aerodynamic diameter of 10 µm or less (PM_{10}) in different ways: Via the determination of local meteorological conditions favoring or suppressing the formation and the accumulation of PM_{10}, and through its control on short– and long–range transport of particles and precursors. The quantitative assessment of the connections between the large–scale atmospheric circulation and local PM_{10} is relevant not only for the understanding of observed variations in PM_{10} concentrations. It is even more important for estimating the potential effects of projected future changes in large–scale atmospheric circulation on PM_{10}. In this contribution, daily atmospheric circulation types (CTs), resulting from variants of three different classification methods, and their monthly occurrence frequencies have been utilized in three different downscaling approaches for estimating monthly indices of PM_{10} for the period 1980–2010 at 16 locations in Bavaria (Germany). All variants of approaches have been evaluated via a leave–one–out cross validation procedure in order to attain reliable performance ratings to detect the most suitable downscaling approaches. Results indicate that the highest performance of downscaling approaches is achieved in winter when the best performing models explain on average roughly 50% of the observed PM_{10} variance. From this it can be concluded that classification–based approaches are generally suitable for the downscaling of PM_{10}, particularly during winter when PM_{10} concentrations in Bavaria reach maximum values. As preferable settings of the downscaling approaches, the usage of rather small spatial domains and a relatively high number of classes for circulation type classification and furthermore the utilization of multiple linear regression analyses or random forest analyses for relating CTs to PM_{10} have been ascertained. These findings provide the basis for further enhancements of the classification–based downscaling of monthly PM_{10} that will be realized in successive investigations.

Keywords: *Particulate matter, Bavaria (Germany), atmospheric circulation, circulation types*

Corresponding Author:
Christoph Beck
☎ : *+49-821-598-2129*
🖷 : *+49-821-598-2264*
✉ : *christoph.beck@geo. uni-augsburg.de*

Article History:
Received: 14 February 2014
Revised: 22 April 2014
Accepted: 27 May 2014

doi: 10.5094/APR.2014.083

1. Introduction

Besides various gaseous atmospheric pollutants like ozone (O_3), sulfur dioxide (SO_2) or different nitrogen oxides (NO_X), particulate matter (PM) that includes solid particles and liquid droplets have distinct adverse effects on human health. This is especially true for particles with an aerodynamic diameter of 10 µm or less (PM_{10}) which may penetrate into the respiratory system, the farther the smaller they are (Harrison and Yin, 2000). Numerous epidemiological studies have shown the association between particulate air pollution and varying serious health effects like the aggravation of existing respiratory and cardiovascular diseases or even lung cancer incidence (e.g. Brunekreef and Holgate, 2002; Medina et al., 2004; Dominici et al., 2005). PM_{10} can be directly emitted from varying natural and anthropogenic sources (primary PM_{10}) or can be formed as secondary PM_{10} via the oxidation of gaseous precursor substances like sulfur dioxide (SO_2), nitrogen oxides (NO_X), ammonia (NH_3) or various organic compounds (e.g. Querol et al., 2004; Seinfeld and Pandis, 2006; Weijers et al., 2011). Natural PM_{10} may comprise geogenic or marine aerosols (e.g. Saharan dust, volcanic ash, sea salt) but as well biogenic particulates like pollens, fungal spores, bacteria or viruses (e.g. Querol et al., 2004; Seinfeld and Pandis, 2006; Weijers et al., 2011). Anthropogenic PM_{10} mainly stems from industrial and domestic fossil fuel burning, road traffic, bulk handling of cargoes and other production processes (e.g. Lenschow et al., 2001).

Spatial and temporal variations in local PM_{10} concentrations are determined by changes in local emission rates of natural and anthropogenic PM_{10}. In addition, they are strongly influenced by varying meteorological and climatological conditions on the local and synoptic scales. Numerous studies have identified varying local meteorological variables that affect local PM_{10} concentrations. These include as most important ones: boundary layer height (Hooyberghs et al., 2005; Holst et al., 2008; Rost et al., 2009) and atmospheric stability (Smith et al., 2001; Triantafyllou, 2001; Holst et al., 2008; Stadlober et al., 2008), air pressure (Cheng et al., 2007; Demuzere et al., 2009; Gietl and Klemm, 2009), wind speed (Smith et al., 2001; Triantafyllou, 2001; Cheng et al., 2007; Stadlober et al., 2008; Demuzere et al., 2009; Gietl and Klemm, 2009) and wind direction (Smith et al., 2001; Hooyberghs et al., 2005; Sanchez–Reyna et al., 2006), global radiation (Pitz et al., 2008; Demuzere et al., 2009) and cloud cover (Hooyberghs et al., 2005; Demuzere et al., 2009), air temperature (Cheng et al., 2007; Stadlober et al., 2008; Demuzere et al., 2009; Gietl and Klemm, 2009), humidity (Wise and Comrie, 2005; Cheng et al., 2007; Demuzere et al., 2009; Gietl and Klemm, 2009) and precipitation (Holst et al., 2008; Stadlober et al., 2008; Demuzere et al., 2009; Rost et al., 2009).

Besides the local meteorological conditions, the synoptic scale circulation is a highly relevant controlling factor of local aerosol concentrations. Synoptic circulation patterns largely determine the local meteorological conditions that are relevant for PM_{10} concentrations. Furthermore, the synoptic circulation controls short– and long–range transport of primary particulate matter and secondary particulate matter precursors.

In a number of studies, the connection between synoptic scale atmospheric circulation and local PM_{10} concentrations has been analyzed, mainly on the basis of weather and circulation type classifications (CTCs). Flocas et al. (2009) detected specific large–scale circulation patterns related to exceedances of several air pollutants–including PM_{10}–in Thessaloniki (Greece) via manual classification of gridded sea level pressure data. Also on the basis of manual CTCs, Dayan and Levy (2005) and Makra et al. (2007) investigated the relationship between large–scale atmospheric circulation and PM_{10} concentrations in Tel Aviv (Israel) and Szeged (Hungary), respectively. For manually derived circulation types (CTs) over the New England region (USA), Keim et al. (2005) detected distinctly different levels of particulate matter concentrations in Durham, New Hampshire (USA). A statistically significant influence of automatically derived large–scale CTs on local PM_{10}–concentrations in Edinburgh (UK) has been found by Buchanan et al. (2002) applying non–parametric analysis of variance.

From these findings a significant influence of large–scale synoptic conditions (reflected by CTs and weather types) on local air pollution levels including PM_{10} concentrations can be deduced. Taking into consideration recent and potential future climate change, it can furthermore be argued that associated variations in large–scale synoptic circulation will probably provoke corresponding changes in PM_{10} concentration levels (Bernard et al., 2001; Ebi and McGregor, 2008).

To the author's best knowledge, there has been no study so far that applies and quantitatively evaluates such a comprehensive set of classification–based statistical downscaling models for monthly PM_{10}, thus identifying the most suitable approach for varying locations and seasons. Against this background, the main objective of this contribution is to establish quantitative relationships between atmospheric CTs and local PM_{10} concentrations at different sites in Bavaria (Germany).

To this end varying CTCs were applied to daily gridded sea level pressure (SLP) data for the period 1980–2010. Monthly occurrence frequencies of the resulting CTs were then utilized as predictors in different statistical models to estimate monthly PM_{10} values at several locations. The skill of each model was evaluated via a cross validation procedure. Finally, the most suitable approaches for estimating local PM_{10} from the large–scale atmospheric circulation were identified. In future research these approaches can then be applied to CTs that are determined from data of climate model scenario runs, to estimate possible future PM_{10} concentration levels under climate change conditions.

The paper is structured as follows: Section 2 introduces the data sets used in our analyses and explicates the varying approaches for circulation type classification and for relating CTs to local PM_{10}. Main results are presented and briefly discussed in Section 3. Section 4 provides a short summary and the main conclusions.

2. Data and Methods

2.1. Daily PM_{10} concentration data

Measurements of PM_{10} concentrations (µg/m^3) at several sites in Bavaria are available from the official air quality monitoring network in Bavaria (LfU, 2014). Prior to 1987, daily mean values were recorded while 3–hourly and 0.5–hourly data were determined since 1987 and 2000, respectively. In order to provide a consistent data base, all sub–daily data have been aggregated to daily mean values. The change from the recording of total suspended particles (TSP) to direct PM_{10} measurements that took place in the year 2000, was accounted for by applying an empirical correction factor of 0.83 – as recommended by the 1999/30/EC directive (EU, 1999) – to all daily mean data until December 31, 1999.

From the 46 stations from the Bavarian air quality monitoring network for which PM_{10} concentrations are available, 16 stations for which complete data are available for at least 90% of all months in the period 1980 to 2010 have been selected for further analyses. The locations of the 16 stations are displayed in Figure 1, a listing of the stations together with some additional specifications is given in Table 1.

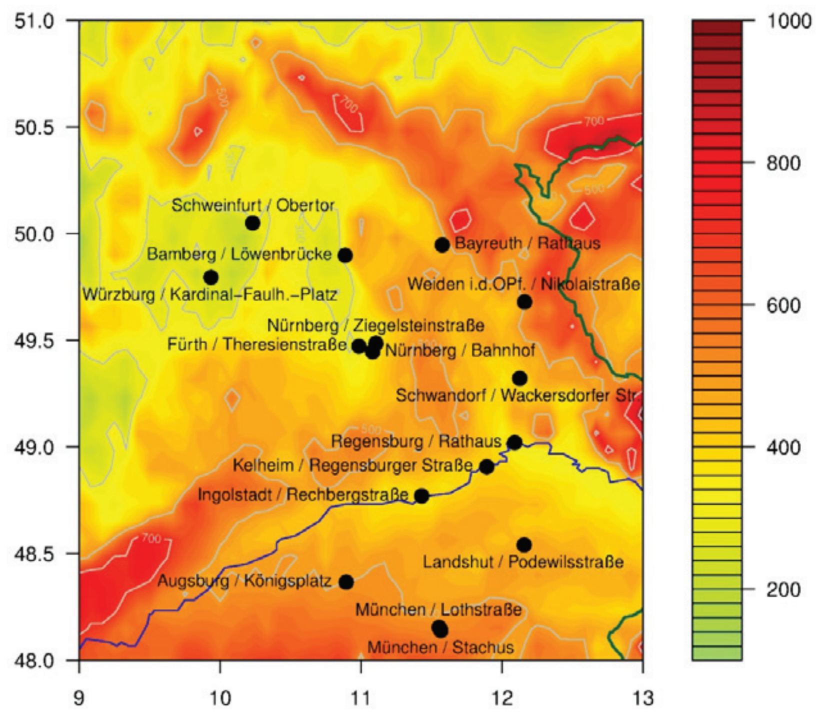

Figure 1. *Map of the locations of 16 selected stations from the Bavarian air quality monitoring network for which PM_{10} concentrations are available for at least 90% of all months in the period 1980 to 2010. Color shading indicates heights in m a.s.l.*

Table 1. *Stations from the Bavarian air quality monitoring network for which PM_{10} concentrations are available for at least 90% of all months in the period 1980 to 2010*

Station Number	Station Name	Longitude	Latitude	Height (m, a.s.l.)	Site Environment Type
L1.1	Ingolstadt/Rechbergstrasse	11.43	48.77	377	Urban traffic
L2.1	Kelheim/Regensburger Strasse	11.89	48.91	351	Urban traffic
L2.3	Landshut/Podewilsstrasse	12.16	48.54	393	Urban traffic
L3.1	Regensburg/Horatius	12.09	49.02	345	Urban traffic
L3.3	Weiden i. d. OPf./Nikolaistrasse	12.16	49.68	402	Urban background
L3.4	Schwandorf/Wackersdorfer Str.	12.13	49.32	383	Suburban background
L4.2	Bayreuth/Rathaus	11.58	49.95	338	Urban traffic
L4.3	Bamberg/Lowenbrucke	10.89	49.90	236	Urban background
L5.1	Nürnberg/Bahnhof	11.08	49.45	312	Urban traffic
L5.2	Nurnberg/Ziegelsteinstrasse	11.11	49.49	321	Urban traffic
L5.5	Furth/Theresienstrasse	10.98	49.47	300	Urban traffic
L6.3	Schweinfurt/Obertor	10.23	50.05	232	Urban background
L6.4	Wurzburg/Kardinal–Faulh.–Platz	9.94	49.79	184	Urban traffic
L7.1	Augsburg/Konigsplatz	10.89	48.37	497	Urban traffic
L8.1	Munchen/Stachus	11.57	48.14	527	Urban traffic
L8.3	Munchen/Lothstrasse	11.55	48.15	519	Urban background

Two data sets of monthly PM_{10} data have been compiled from the daily mean PM_{10} concentration data: (1) monthly mean PM_{10} concentrations (μg/m^3) (PM_{mean} hereinafter), (2) monthly exceedances of a daily mean value of 50 μg/m^3 (days/month) (PM_{e50} hereinafter). The exceedance of 50 μg/m^3 is allowed for not more than 35 days/year according to the 2008/50/EC directive (EU, 2008).

2.2. Daily gridded SLP data

From the NCEP/NCAR reanalysis 1 data archive (Kalnay et al., 1996), 2.5° by 2.5° gridded daily SLP data are globally available for the period since 1948 to present. For the determination of CTs in our study, we used SLP data for 12 UTC for varying sub–regions within the superordinate North Atlantic European region (62.5°W to 82.5°E; 12.5°S to 87.5°N). The dimensions of the sub–regions are given in Table S1 (see the Supporting Material, SM).

2.3. Circulation type classifications

CTCs are often applied for categorizing the continuum of atmospheric circulation into a reasonable number of discrete CTs. The resulting CTs describe main characteristics of the atmospheric circulation dynamics. A large variety of classification approaches is utilized in synoptic climatological studies to investigate the relationship between the atmospheric circulation and varying environmental target variables (see for example Huth et al., 2008; Huth et al., 2010). Recent studies have shown that the applicability of CTCs varies distinctly with respect to season, location and target variable and is also dependent on specific method configurations (e.g. Beck and Philipp, 2010; Beck et al., 2013). In this study we therefore employ not only one CTC, but multiple variants of three different classification approaches to daily gridded SLP for the period 1980 to 2011 in order to figure out those CTCs that are best suited for PM_{10} related analyses.

The three classification approaches can briefly be described as follows: The Grosswettertypes approach or prototype classification (GWT) assigns cases (daily SLP fields) to classes (CTs) depending on their zonality, meridionality and vorticity characteristics that are determined as spatial correlation coefficients between daily SLP fields and prototypical SLP patterns (Beck et al., 2007). The Lund classification (LND) estimates the spatial Pearson correlation coefficients between daily SLP fields as similarity measure and based thereupon merges most similar cases to CTs (Lund, 1963). The third classification (DKM) uses non–hierarchical k–means cluster analysis (Hartigan, 1975) for deriving CTs thereby utilizing most dissimilar cases (SLP fields) included in the data set to determine the initial starting partition (Enke and Spekat, 1997; Philipp et al., 2010).

These three classifications have been selected from the multitude of available methods because of two reasons: First, they represent three basic approaches (threshold based, correlation based, optimization algorithms) commonly used for the classification of circulation types (see Philipp et al. (2010) for details). And secondly, their above–average suitability for PM_{10} related synoptic analyses turned out from preliminary studies.

Previous studies have shown that varying configurations of CTCs have distinct effects on their applicability for relating atmospheric circulation to environmental variables (e.g. Beck and Philipp, 2010; Beck et al., 2013). Therefore the three classification approaches have been run in several variants concerning the following features:

The size of the spatial domain to which the CTCs are applied varies among 8 sizes ranging from 17.5° to 87.5° longitudinal and from 12.5° to 47.5° latitudinal extension (all domains are centered around 11.25°E/48.75°N). All classifications have been run for three different numbers of CTs (10, 18, and 27). All CTCs based on LND and DKM, respectively, have furthermore been applied not only to SLP fields of single days but as well to three–day sequences of daily SLP fields. Table S1 in the SM gives an overview of the CTC variants conducted in this study.

All CTC variants have been run using the software package cost733class (Philipp et al., 2014) that has been developed in the COST733 action "Harmonisation and Applications of Weather Type Classifications for European Regions" (Huth et al., 2010).

As main results of each CTC we get (1) the CT catalog that comprises for each day the information which CT occurred on that specific day and (2)–as spatial representation of each CT–the CT composites (or centroids) which are calculated as the arithmetic mean of all cases (daily SLP fields) assigned to the respective CT (see Figure 2 and Figures S1 to S2 in the SM for exemplary illustrations of CT composites resulting from three selected CTCs).

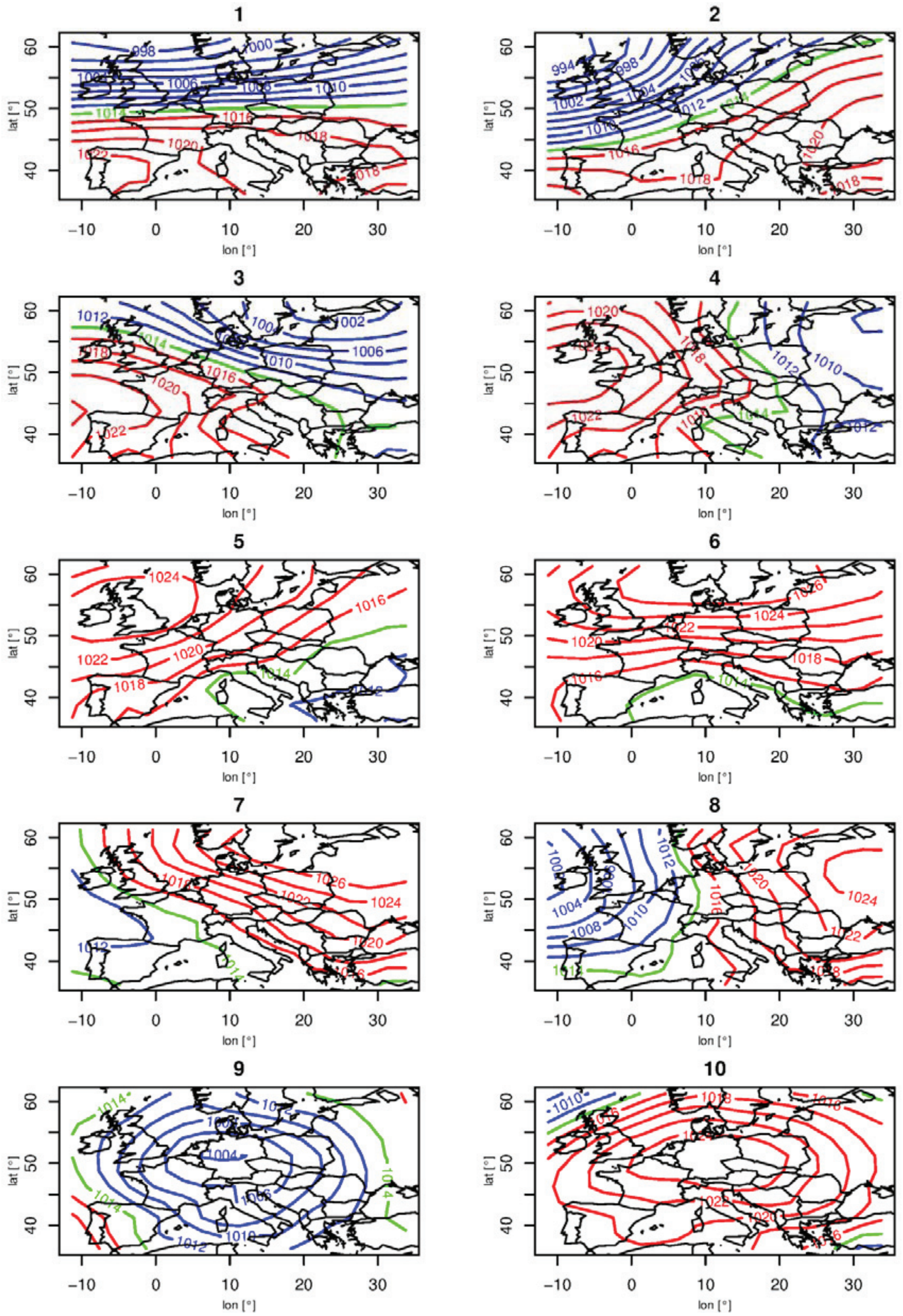

Figure 2. *SLP (hPa) composites for 10 circulation types resulting from the GWT classification applied to daily SLP (1980–2011) for the spatial domain 12.5°W–35°E/35°N–62.5°N.*

2.4. Relating circulation types to monthly PM_{10}

The estimation of monthly PM_{10} indices from CTs assumes that different CTs feature distinctly diverse PM_{10} concentration levels. It is not possible to depict for each CTC used in this study in how far this assumption is fulfilled. Thus one example may serve as proof for the general validity of this assumption.

Figure 3 shows seasonal box–plots of daily PM_{10} concentrations at the air quality monitoring station Nurnberg/ Ziegelsteinstrasse grouped according to the ten CTs shown in Figure 2. Distinctly varying PM_{10} concentration levels among CTs can be identified. Disregarding seasonal variations, highest concentrations are related to CTs featuring easterly, south–easterly and southerly air mass advection (CTs 6 to 8) or a central high pressure pattern (CT 10). Accordingly high PM_{10} levels at Nurnberg/Ziegelsteinstrasse may be related either to short and long–range transport of PM_{10} from regions with high emission levels (CTs 6 to 8) or to the accumulation of in situ emissions due to decelerated horizontal and vertical air mass mixing (CT 10). Lower concentrations of PM_{10}, on the other hand, appear to be related to the occurrence of CTs implying the advection of relatively unpolluted air masses (CTs 1 to 4) or central low pressure patterns (CT 9) associated with above normal precipitation. Very similar relationships are apparent for the other stations as well.

Therefore it is justified to utilize CTs or their occurrence frequencies for relating atmospheric circulation dynamics to PM_{10}. In this study, three different approaches for relating CTs to monthly PM_{10} indices (PM_{mean}, PM_{e50}) have been applied and compared. Taking into account intra–annual variations in relationships (see Figure 3), all downscaling approaches have been applied separately to the four 3–month seasons winter (DJF), spring (MAM), summer (JJA) and autumn (SON) and as well separately to the individual months January to December.

Synoptic downscaling. Within the synoptic downscaling approach (SD), daily PM_{10} concentrations were estimated from daily CTs by (1) calculating long–term conditional mean PM_{10} concentrations for each circulation type and (2) using these conditional mean values to estimate daily PM_{10} concentrations for all days with the occurrence of the corresponding circulation type. Monthly PM_{10} indices were subsequently determined by averaging the daily estimates (PM_{mean}) or by counting the estimated daily exceedances of 50 µg/m^3 (PM_{e50}).

Multiple linear regression. Monthly occurrence frequencies of CTs were utilized as predictors in multiple linear regression analyses (MRA) to estimate the predictand variables PM_{mean} and PM_{e50}, respectively. In order to determine the adequate predictor variables, stepwise MRA was performed for 100 random samples, each comprising 20 years of data. The Akaike information criterion (Akaike, 1974) was used for model selection. From these explorative MRAs the most frequent number of predictors and the most frequently chosen predictors were determined. All subsequent applications of the MRAs were then run using these specific combinations of predictor variables.

Random forests. Random forests (RF) were introduced by Breiman (2001) and are based on classification and regression tree (CART) analysis (Breiman et al., 1984). CART models are represented as binary decision trees, they have the advantage that the input data do not need to be normally distributed and that linear as well as nonlinear relationships between predictors and predictand are captured. However, one weakness of conventional CART analysis is its instability concerning changes in the training data. RF uses two strategies to overcome this restriction. Firstly, many binary decision trees determined for random subsamples of the observation data are combined. Secondly, at each node the best split is determined on the basis of a random subsample of explanatory variables. In the present study, RF has been applied using monthly occurrence frequencies of CTs as predictors and monthly PM_{mean} and PM_{e50} as predictands (response variables). For each run 500 trees have been used and each split has been determined on the basis of $\sqrt{p}$ randomly chosen predictors (where p is the number of explanatory variables).

2.5. Model validation

To each of the 16 Bavarian stations and for 4 seasons and 12 months, respectively, 360 downscaling models (120 CTCs utilized in 3 different downscaling approaches) have been applied. Each approach has finally been subjected to a leave–one–out cross validation procedure in order to substantiate assertions concerning the robustness of the model.

Leave–one–out cross validation uses a single observation (month or season) from the whole sampling (period 1980–2011) for validation and the remaining observations for calibration of the model. This procedure was repeated until each observation in the sample has been used once for validation (see Arlot and Celisse, 2010 for an extended discussion on cross validation techniques). Finally the skill of the validated models has been quantified by calculating the squared Pearson correlation coefficient (r^2) and the mean error (ME) or bias between observed and modeled series.

3. Results and Discussion

The application of the varying downscaling approaches yields numerous results of which only a deliberate selection can be presented here. We thereby focus on those approaches that – in terms of r^2 – turned out to be most suitable for estimating monthly PM_{10} indices at a specific location.

Main characteristics of these best performing approaches for downscaling of monthly PM_{10} indices from CTs are summarized for PM_{mean} in Table 2 (for the seasonal models) and in Tables S2 to S4 in the SM (for PM_{e50} for the seasonal models and for PM_{mean} and PM_{e50} for the monthly models, respectively).

Table 2. *Features of the best performing (according to r^2 for cross validation) seasonal approaches for estimating PM_{mean} at Bavarian air quality measurement stations. Beside the two skill metrics r^2 and ME, the method (CM; Prototype classification–GWT, Lund classification–LND, k–means classification–DKM), the number of circulation types (NT; 10, 18, 27), the domain size (DS; increasing from 1 to 8), the sequence–length (SL; 1 or 3 days) of the underlying circulation type classification and the used downscaling approach (DSA; synoptic downscaling–SD, multiple linear regression–MRA, random forests–RF) are given*

Station	Season	r^2	ME	CM	NT	DS	SL	DSA
Ingolstadt/Rechbergstrasse	DJF	0.61	–0.18	LND	27	7	3	MRA
	MAM	0.52	0.04	DKM	10	2	3	SD
	JJA	0.36	0.05	LND	10	6	1	MRA
	SON	0.31	0.11	LND	18	7	1	MRA
Kelheim/Regensburger Strasse	DJF	0.42	–0.22	DKM	18	1	3	MRA
	MAM	0.34	0.05	DKM	27	1	3	MRA
	JJA	0.24	–0.14	GWT	27	2	1	MRA
	SON	0.50	0.39	LND	27	8	1	RF
Landshut/Podewilsstrasse	DJF	0.44	–0.02	LND	18	5	3	MRA
	MAM	0.40	0.60	LND	27	8	3	RF
	JJA	0.50	0.15	GWT	27	5	1	RF
	SON	0.38	0.04	DKM	27	2	1	RF

Table 2. *Continued*

Station	Season	r^2	ME	CM	NT	DS	SL	DSA
Regensburg/Rathaus	DJF	0.42	0.01	GWT	18	2	1	MRA
	MAM	0.42	0.31	DKM	27	8	3	RF
	JJA	0.76	0.09	GWT	27	5	1	RF
	SON	0.45	0.01	LND	27	6	1	RF
Weiden i.d.OPf./Nikolaistrasse	DJF	0.46	–0.25	LND	18	2	3	RF
	MAM	0.46	0.06	GWT	18	1	1	MRA
	JJA	0.32	0.08	LND	10	8	1	RF
	SON	0.28	–0.05	DKM	27	1	1	RF
Schwandorf/Wackersdorfer Str.	DJF	0.48	0.03	LND	18	2	3	MRA
	MAM	0.42	0.06	DKM	18	6	3	MRA
	JJA	0.41	0.16	LND	27	5	3	RF
	SON	0.37	–0.08	LND	27	4	1	MRA
Bayreuth/Rathaus	DJF	0.48	0.15	LND	27	8	3	MRA
	MAM	0.30	0.48	DKM	10	4	3	RF
	JJA	0.58	0.23	DKM	27	1	3	RF
	SON	0.58	0.29	DKM	18	1	3	RF
Bamberg/Lowenbrucke	DJF	0.53	–0.04	LND	27	3	1	MRA
	MAM	0.53	0.05	DKM	27	4	1	MRA
	JJA	0.41	0.01	LND	18	5	1	MRA
	SON	0.36	0.13	LND	27	6	1	MRA
Nurnberg/Bahnhof	DJF	0.46	0.18	LND	18	3	1	RF
	MAM	0.40	0.02	DKM	27	8	3	MRA
	JJA	0.31	0.10	DKM	10	3	3	RF
	SON	0.27	0.10	DKM	27	1	1	RF
Nurnberg/Ziegelsteinstrasse	DJF	0.59	–0.15	LND	18	2	1	MRA
	MAM	0.45	0.13	LND	27	8	1	RF
	JJA	0.59	0.26	DKM	27	2	1	RF
	SON	0.42	0.24	DKM	27	1	3	RF
Furth/Theresienstrasse	DJF	0.52	0.43	LND	27	2	3	MRA
	MAM	0.40	0.24	GWT	18	6	1	SD
	JJA	0.31	0.19	LND	10	1	1	RF
	SON	0.23	–0.10	LND	18	1	1	MRA
Schweinfurt/Obertor	DJF	0.55	–0.15	LND	27	6	1	MRA
	MAM	0.41	0.45	LND	27	8	1	RF
	JJA	0.42	0.17	LND	27	3	3	RF
	SON	0.23	–0.05	LND	27	6	3	MRA
Wurzburg/Kardinal–Faulh.–Platz	DJF	0.45	–0.18	GWT	18	4	1	MRA
	MAM	0.41	0.40	LND	27	8	1	RF
	JJA	0.34	0.20	DKM	27	8	1	RF
	SON	0.46	0.30	LND	27	2	1	RF
Augsburg/Konigsplatz	DJF	0.44	0.66	DKM	27	2	3	RF
	MAM	0.50	0.74	LND	27	4	3	RF
	JJA	0.55	0.60	GWT	18	3	1	RF
	SON	0.52	0.75	LND	18	6	3	RF
Munchen/Stachus	DJF	0.40	0.13	LND	27	1	1	MRA
	MAM	0.30	–0.19	DKM	27	8	3	MRA
	JJA	0.31	–0.08	DKM	27	2	3	MRA
	SON	0.41	0.02	LND	18	5	1	MRA
Munchen/Lothstrasse	DJF	0.48	0.00	LND	27	6	1	SD
	MAM	0.41	–0.05	DKM	18	6	3	MRA
	JJA	0.58	0.14	GWT	18	6	1	RF
	SON	0.44	0.14	GWT	10	6	1	RF

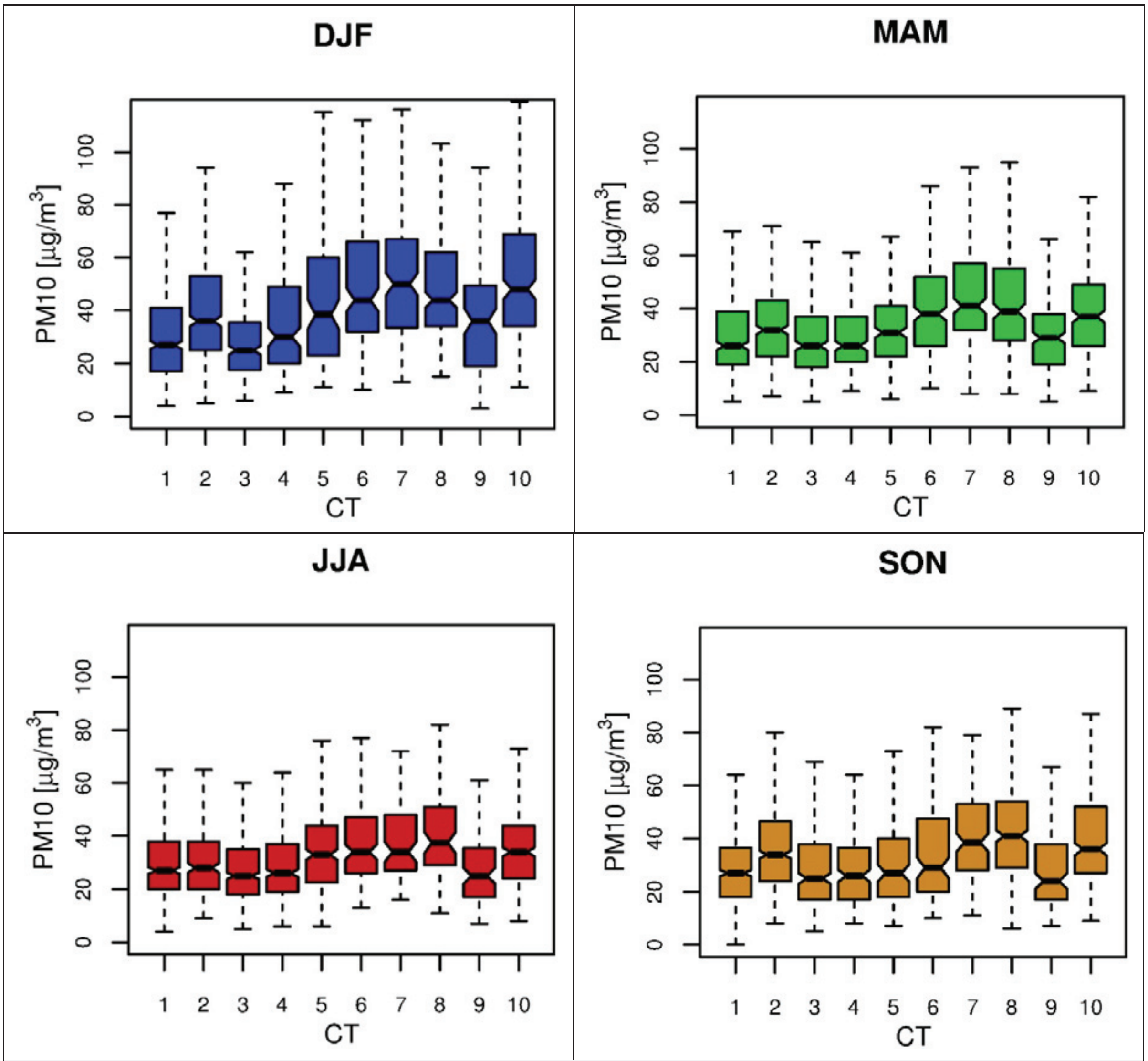

Figure 3. *Box–plots of daily PM_{10} concentrations (µg/m^3) at the air quality monitoring station Nürnberg/Ziegelsteinstrasse. Separate plots are given for the four 3–month seasons (DJF, MAM, JJA, SON). Box–plots are grouped according to 10 circulation types (CT) resulting from the GWT classification applied to daily SLP (1980–2011) for the spatial domain 12.5°W–35°E/35°N–62.5°N (see Figure 2 for the corresponding SLP (hPa) composites). Upper/lower whiskers indicate the 1.5 interquartile range from the upper/lower quartile. Notches represent estimates of the 95% confidence intervals of the respective medians. Thus non–overlapping notches point to significant differences of the respective medians.*

3.1. Skill of the best performing downscaling approaches

The skill of the seasonal downscaling models for PM_{mean} as measured by r^2 ranges between 0.23 and 0.76. Distinct variations in r^2 appear between seasons (with highest values of r^2 – averaged over all stations – appearing in DJF) and as well between stations. However, no clear–cut connection between skill and station location or type of environment becomes evident. Differences in skill between suburban background, urban background and urban traffic stations (Figure S3 in the SM) were not statistically significant.

The superior mean skill in DJF may be at least partly attributed to the fact that in mid–latitude winter the connection between synoptic circulation and local meteorological conditions (which in turn are influencing PM_{10} concentration levels) is much stronger than during the other seasons, especially JJA when small scale dynamic processes (e.g. local convection) are more dominant (e.g. Schiemann and Frei, 2010).

Marked variations in skill and as well in specific features of the best performing models between – often nearby – stations may appear suspicious at first view. However, comparable variations between locations appear as well when relating CTs to surface climate variables (e.g. Beck et al., 2013). The marked spatial variations in the skill of the downscaling models can be explained by variations of the manifold factors that affect local PM_{10} on different spatial and temporal scales. Concerning ME, Table 2 indicates that the bias of the selected best models is positive in most cases – indicating an overestimation of PM_{mean} – however, generally of rather minor magnitude.

Turning to the seasonal models applied to PM_{e50} (Table S2 in the SM), quite similar variations in r^2 among seasons and stations become obvious. However, on the whole (mean over all stations and seasons) r^2 for PM_{e50} (0.38) is lower than for PM_{mean} (0.43). Furthermore ME – although again generally small – reaches distinctly higher negative values for some stations/seasons. This lower performance of the PM_{e50} models can be attributed to the general incapability of CTCs to clearly resolve values from the tails of the frequency distribution of the target variable, as each CT represents the mean of all cases assigned to this CT.

Values of PM_{e50} determined on a monthly basis are often 0 indicating that no daily mean value exceeding 50 µg/m^3 appeared in the specific month. As this may influence model performance, all PM_{e50} models have been additionally run using CT frequencies and PM_{e50} summed up for each of the 3–month seasons (DJF, MAM, JJA, SON), thus reducing the number of zero values of PM_{e50}. However, results of these analyses (not shown) generally confirm the above stated findings.

As an example time series of observed and modeled PM_{mean} and PM_{e50} are displayed in Figures 4 and 5 for the station Nurnberg/Ziegelsteinstrasse featuring highest mean skill (in terms of r^2) averaged over the four 3–month seasons and over PM_{mean} and PM_{e50} (see Tables 2 and S2).

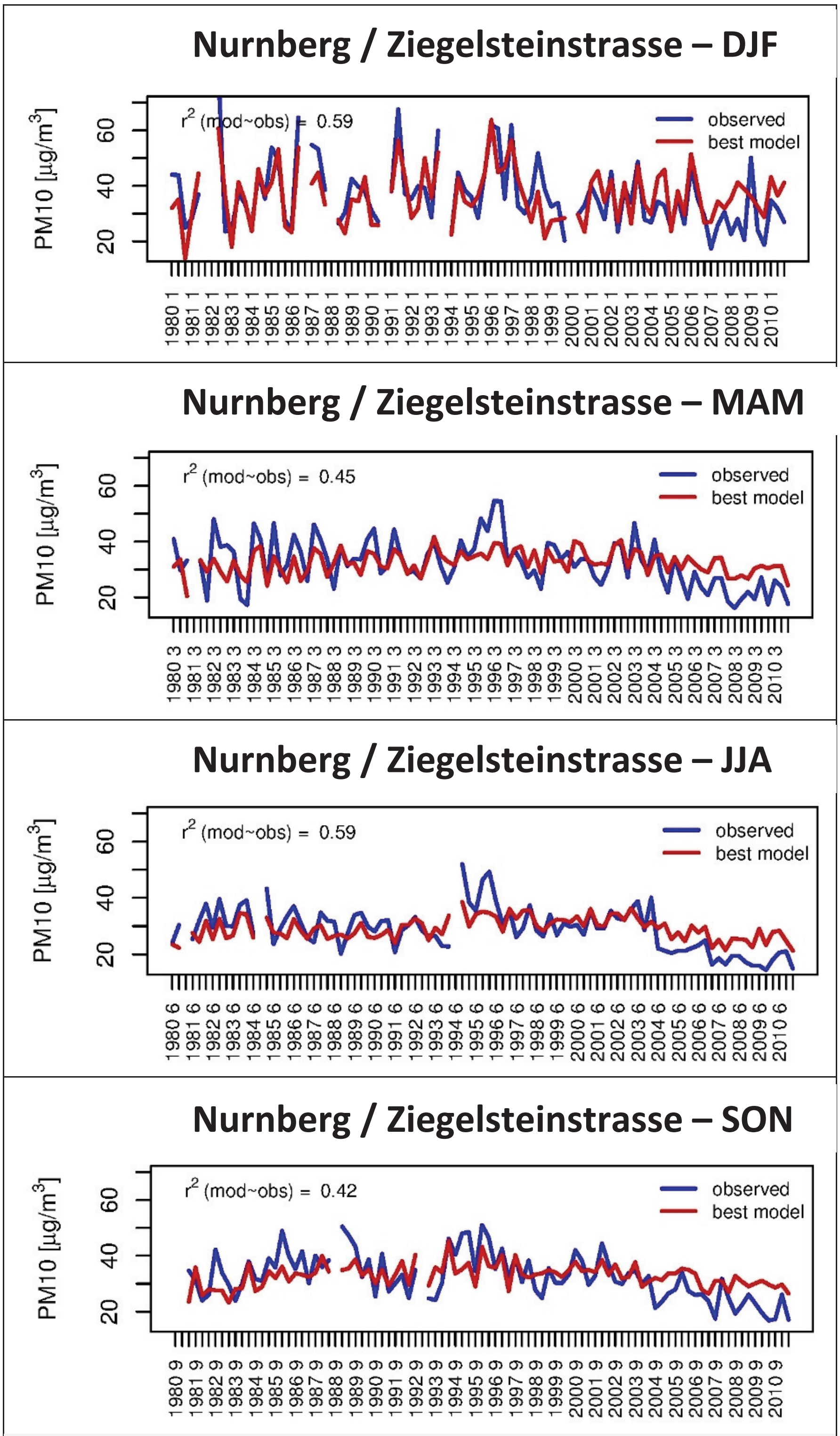

Figure 4. *Time series of observed and modeled (best seasonal models according to Table 2) PM_{mean} at the air quality monitoring station Nurnberg/Ziegelsteinstrasse for the four 3–month seasons (DJF, MAM, JJA and SON). The r^2 between observed and modeled series is given in the top left part of each plot.*

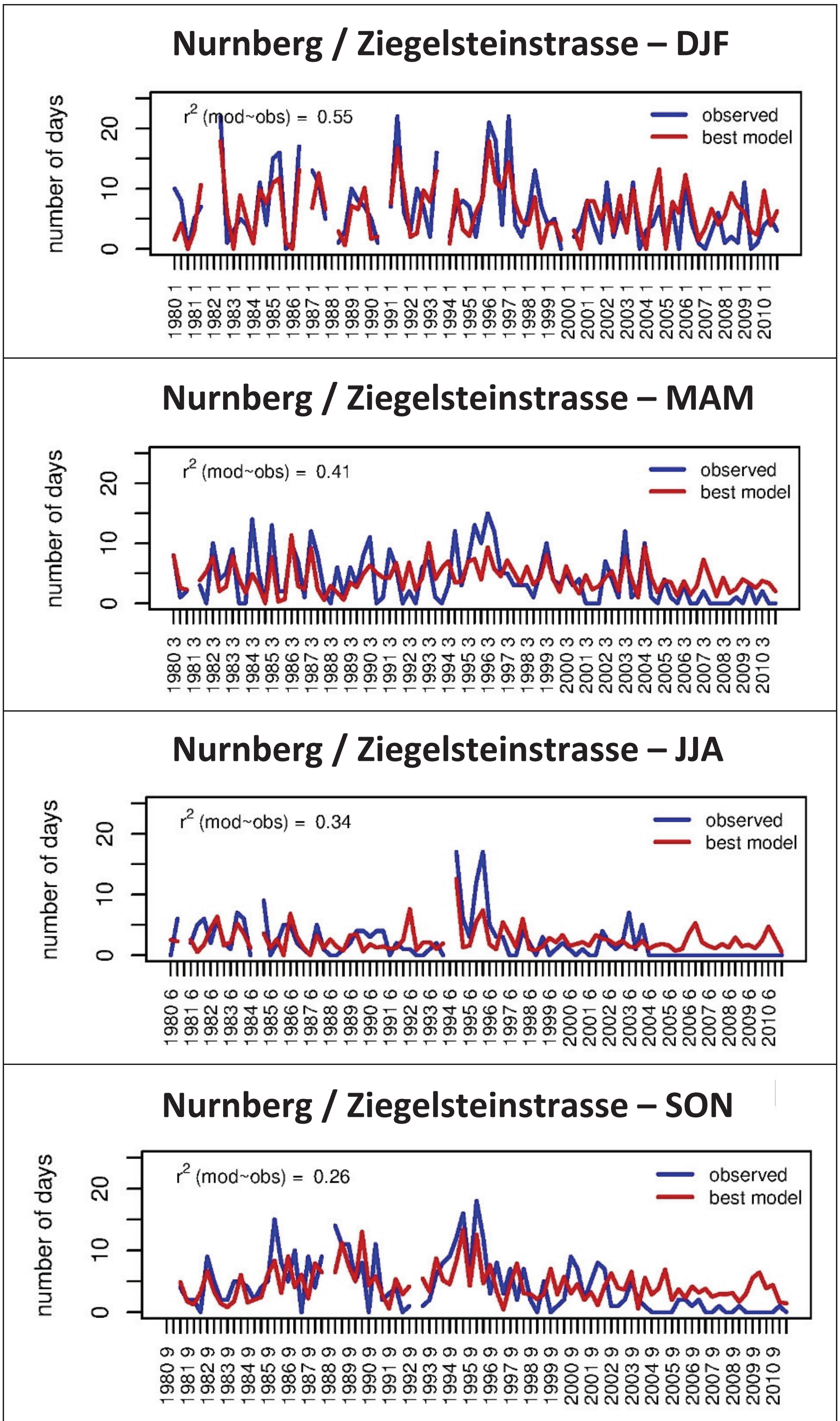

Figure 5. *Time series of observed and modeled (best seasonal models according to Table S2) PM_{e50} at the air quality monitoring station Nurnberg/Ziegelsteinstrasse for the four 3–month seasons (DJF, MAM, JJA and SON). The r^2 between observed and modeled series is given in the top left part of each plot.*

Figures 4 and 5 depict the variations in skill between seasons and between the predictand variables PM_{mean} and PM_{e50}. The classification–based downscaling approaches succeed in reproducing large parts of the inter–annual variations in PM_{10} indices. However, the modeled series do not reflect systematic longer term changes in PM_{10} concentration levels, for example the general decrease since the 1990s that is mainly due to reductions in particulate matter emissions in Germany (UBA, 2014).

In order to find out in how far model skill may be increased and to detect variations in model skill between months, all downscaling models have additionally been applied to single months. Thereby the sample size for calibration and validation decreases from 93 cases to 31 cases. It has to be kept in mind that this may affect the reliability of the skill estimates for the monthly models. The comparison of the skill of the models for single months (Tables S3 and S4 in the SM) and seasons (Table 2 and Table S2) reveals generally higher skill (averaged over all months and stations) of the monthly models with r^2 values reaching 0.54 and 0.51 for PM_{mean} and PM_{e50}, respectively. Partly distinct variations in model skill appear among months assigned to the same 3–month seasons utilized for the seasonal models. This indicates that differences in the relationship between circulation and PM_{10} exist not only between seasons (see Figure 3) but as well within the 3–month seasons.

3.2. Characteristic features of the best performing downscaling approaches

A wide variety of downscaling approaches to estimate monthly PM_{10} indices have been applied. It is further investigated in how far the best performing downscaling models show characteristic features, in order to derive hints at the most appropriate starting point for further methodical developments.

Considering only the seasonal models listed in Table 2 and Table S2 (see the SM), it turns out that the LND and the DKM classifications appear most often (44% and 38%, respectively) in the best performing models while GWT (18%) is chosen only in a minor number of cases. The low frequency of occurrence of GWT among the best performing models is due to the fact that GWT has not been applied to the classification of 3–day sequences.

CTCs comprising 27 or 18 CTs are far more prevalent in the best performing models than CTCs having 10 CTs. This is in accordance with several other studies (e.g. Beck and Philipp, 2010; Beck et al., 2014) that have shown a distinct dependence of synoptic skill of CTCs (the ability of CTCs to discriminate between varying states of an environmental target variable) on the number of CTs. The concurrent increase in the number of CTs and synoptic skill may be explained by the fact that for higher numbers of types the probability for deriving CTs that are related to clearly disparate states of the environmental target variable (PM_{10}) is higher as well. With respect to the MRA approach, it has to be mentioned in this context that the preferred utilization of CTCs with higher numbers of CTs does not lead to a systematically increased number of final predictors in the best performing downscaling models.

From the varying sizes of the spatial domain to which the CTCs are applied, the smaller domains 1–4 appear more frequently among the best performing approaches than the larger domains 5–8. Apart from respective variations between seasons, this is in general accordance with findings concerning the preferred domain size of CTCs featuring maximum synoptic skill for temperature and precipitation (Beck et al., 2013).

The classification of 3–day sequences instead of single days takes into account the day to be classified and the two preceding days. This CTC variant can be thought of as a reasonable modification to increase the synoptic skill of CTCs for PM_{10}. For instance, it can be assumed that a central high–pressure pattern over Central Europe (see CT 10 in Figure 2) that persists over 3 days leads to a distinct accumulation of PM_{10} due to reduced vertical and horizontal mixing and thus should result in higher PM_{10} concentration levels compared to a central high–pressure pattern that is preceded by two days featuring advective synoptic dynamics. From Tables 2 and S2, it can be deduced that CTCs of 3–day sequences were chosen for roughly 41% of the best performing approaches, whereas one–day CTC variants appear in around 59% of the best models.

Finally, we compared the frequencies of occurrence of the 3 different downscaling approaches in the best performing models. SD is chosen only for roughly 9% of the best models and almost exclusively for DJF. This very low percentage is due to the fact that the very simple SD approach only differentiates as many realizations of the target variable PM_{10} as CTs are derived by the respective CTC. Accordingly SD appears more or less only in models for DJF when connections between CTs and PM_{10} are most clear–cut. MRA and RF appear with approximately comparable relative frequencies of 42% and 49%, respectively, however, RF exhibits a distinct maximum in frequency of appearance in JJA.

4. Summary and Conclusions

In this study the relationship between synoptic circulation and local PM_{10} concentrations at different sites in Bavaria (Germany) has been investigated. Varying CTCs have been applied to daily gridded SLP for the period 1980–2010 to derive disjunct CTs representing main characteristics of the atmospheric circulation over the European domain. Monthly occurrence frequencies of these CTs have been related to monthly indices of PM_{10} (PM_{mean} and PM_{e50}) at 16 Bavarian air quality monitoring stations utilizing 3 different downscaling approaches. A leave–one–out cross validation framework has been used to reliably determine the performance of each downscaling model.

Focusing on those models that perform best (in terms of r^2 between observed and modeled PM_{10}) at each location in each season, the main conclusions can be specified as follows.

The most distinct connection between CTs and local PM_{10} exists in DJF. Relationships were less pronounced in JJA and the transitional seasons MAM and SON. A general decrease in model skill also appears for the target variable PM_{e50} compared to PM_{mean}. In addition, variations in the strength of the connection between circulation and PM_{10} have also been detected between locations.

With respect to characteristic features of the best performing models, no clearly superior general classification approach for generating the CTCs (DKM, GWT, LND) nor one generally best temporal sequence–length (1–day or 3–day) for the CTCs can be identified. Concerning the size of the spatial domain and the number of CTs, a preference of relatively small spatial domains and higher numbers of classes, respectively, became obvious. Finally MRA and RF clearly outperformed SD as tool for estimating PM_{10} indices from CTs.

As noted above, some hints concerning the preferred configurations of classification–based approaches for downscaling of monthly PM_{10} can be derived. However, the first main conclusion arising from our study is that no generally optimal classification–based downscaling approach exists. It is rather necessary to ascertain the best approach separately for each location and each season.

From the here presented results it can furthermore be concluded that the CTC–based statistical downscaling of monthly PM_{10} indices at the majority of stations exhibits promising skill, particularly in DJF. Given the fact that highest PM_{10} concentration levels in Bavaria usually appear in late winter – particularly in February – this finding has relevance for the intended application of further advanced classification–based downscaling approaches to Global Climate Model projections for the 21st century.

The development of such advanced approaches will particularly comprise enhancements of CTCs with respect to their synoptic skill for PM_{10}. An increase in synoptic skill for PM_{10} may be achieved by applying CTCs to alternative variables (e.g. geopotential height, relative humidity, vorticity for varying pressure levels) or to multiple variables (e.g. geopotential height and relative humidity) or by incorporating the target variable PM_{10} into the classification.

Beside such efforts to increase the synoptic skill of CTCs, a further improvement in model skill may be achieved by applying downscaling models to seasonal subsets differing from the traditional 3–month seasons used in this contribution. Because of the considerably reduced sample sizes for calibration and validation, the application of downscaling models to single months does not appear to be feasible. Instead the implementation of models for 2–month seasons or alternatively defined 3–month seasons may lead to further improvements in the skill of classification–based downscaling of PM_{10}.

Acknowledgments

The authors gratefully acknowledge the Bavarian Environment Ageny (LfU) for the provision of the PM_{10} concentration data from the Bavarian air quality monitoring network and Dr. Andreas Philipp for providing extensive support related to the software package cost733class (http://cost733.geo.uni–augsburg.de/cost733class–1.2). Several of the analyses and plots in this paper were made using GNU's R language (http://www.R–project.org/). This work is funded by the German Research Foundation under contract BE 2406/2–1.

Supporting Material Available

Circulation type composite patterns can be found in Figures S1–S2. Figure S3 provides information on model skill dependent on the site environment. An overview of the utilized variants of circulation type classifications and supporting information on the features of the downscaling approaches are given in Table S1 and Tables S2 to S4, respectively. This information is available free of charge via the internet at http://www.atmospolres.com.

References

Akaike, H., 1974. A new look at the statistical model identification. *IEEE Transactions on Automatic Control* 19, 716–723.

Arlot, S., Celisse, A., 2010. A survey of cross–validation procedures for model selection. *Statistics Surveys* 4, 40–79.

Beck C., Philipp, A., Jacobeit, J., 2014. Interannual drought index variations in Central Europe related to large–scale atmospheric circulation. Submitted to*Theoretical and Applied Climatology*.

Beck C., Philipp, A., Streicher, F., 2013. The effect of domain size on the relationship between circulation type classifications and surface climate. *International Journal of Climatology*, in press, doi: 10.1002/joc.3688.

Beck, C., Philipp, A., 2010. Evaluation and comparison of circulation type classifications for the European domain. *Physics and Chemistry of the Earth* 35, 374–387.

Beck, C., Jacobeit, J., Jones, P.D., 2007. Frequency and within–type variations of large–scale circulation types and their effects on low–frequency climate variability in central Europe since 1780. *International Journal of Climatology* 27, 473–491.

Bernard, S.M., Samet, J.M., Grambsch, A., Ebi, K.L., Romieu, I., 2001. The potential impacts of climate variability and change on air pollution–related health effects in the United States. *Environmental Health Perspectives* 109, 199–209.

Breiman, L., 2001. Random forests. *Machine Learning* 45, 5–32.

Breiman, L., Friedmann, J.H., Ohlsen, R.A., Stone, C.J., 1984. Classification and Regression Trees. Wadsworth. Belmont, pp. 368.

Brunekreef, B., Holgate, S.T., 2002. Air pollution and health. *Lancet* 360, 1233–1242.

Buchanan, C.M., Beverland, I.J., Heal, M.R., 2002. The influence of weather–type and long–range transport on airborne particle concentrations in Edinburgh, UK. *Atmospheric Environment* 36, 5343–5354.

Cheng, C.S.Q., Campbell, M., Li, Q., Li, G.L., Auld, H., Day, N., Pengelly, D., Gingrich, S., Yap, D., 2007. A synoptic climatological approach to assess climatic impact on air quality in south–central Canada. Part I: Historical analysis. *Water Air and Soil Pollution* 182, 131–148.

Dayan, U., Levy, I., 2005. The influence of meteorological conditions and atmospheric circulation types on PM_{10} and visibility in Tel Aviv. *Journal of Applied Meteorology* 44, 606–619.

Demuzere, M., Trigo, R.M., de Arellano, J.V.G., van Lipzig, N.P.M., 2009. The impact of weather and atmospheric circulation on O_3 and PM_{10} levels at a rural mid–latitude site. *Atmospheric Chemistry and Physics* 9, 2695–2714.

Dominici, F., McDermott, A., Daniels, M., Zeger, S.L., Samet, J.M., 2005. Revised analyses of the national morbidity, mortality, and air pollution study: Mortality among residents of 90 cities. *Journal of Toxicology and Environmental Health–Part A–Current Issues* 68, 1071–1092.

Ebi, K.L., McGregor, G., 2008. Climate change, tropospheric ozone and particulate matter, and health impacts. *Environmental Health Perspectives* 116, 1449–1455.

Enke, W., Spekat, A., 1997. Downscaling climate model outputs into local and regional weather elements by classification and regression. *Climate Research* 8, 195–207.

EU (European Union), 2008. http://eur-lex.europa.eu/legal-content/en/ALL/?uri=CELEX:32008L0050, accessed in January 2014.

EU (European Union), 1999. http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:31999L0030, accessed in January 2014.

Flocas, H., Kelessis, A., Helmis, C., Petrakakis, M., Zoumakis, M., Pappas, K., 2009. Synoptic and local scale atmospheric circulation associated with air pollution episodes in an urban Mediterranean area. *Theoretical and Applied Climatology* 95, 265–277.

Gietl, J.K., Klemm, O., 2009. Analysis of traffic and meteorology on airborne particulate matter in Munster, Northwest Germany. *Journal of the Air & Waste Management Association* 59, 809–818.

Hartigan, J.A., 1975. *Clustering Algorithms*, Wiley, New York, 351 pages.

Harrison, R.M., Yin, J.X., 2000. Particulate matter in the atmosphere: Which particle properties are important for its effects on health? *Science of the Total Environment* 249, 85–101.

Holst, J., Mayer, H., Holst, T., 2008. Effect of meteorological exchange conditions on PM_{10} concentration. *Meteorologische Zeitschrift* 17, 273–282.

Hooyberghs, J., Mensink, C., Dumont, G., Fierens, F., Brasseur, O., 2005. A neural network forecast for daily average PM_{10} concentrations in Belgium. *Atmospheric Environment* 39, 3279–3289.

Huth, R., Beck, C., Tveito, O.E., 2010. Classifications of atmospheric circulation patterns – theory and applications – preface. *Physics and Chemistry of the Earth* 35, 307–308.

Huth, R., Beck, C., Philipp, A., Demuzere, M., Ustrnul, Z., Cahynova, M., Kysely, J., Tveito, O.E., 2008. Classifications of atmospheric circulation patterns recent advances and applications. *Trends and Directions in Climate Research* 1146, 105–152.

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K.C., Ropelewski, C., Wang, J., Leetmaa, A., Reynolds, R., Jenne, R., Joseph, D., 1996. The NCEP/NCAR 40–year reanalysis project. *Bulletin of the American Meteorological Society* 77, 437–471.

Keim, B.D., Meeker, L.D., Slater, J.F., 2005. Manual synoptic climate classification for the east coast of New England (USA) with an application to $PM_{2.5}$ concentration. *Climate Research* 28, 143–154.

Lenschow, P., Abraham, H.J., Kutzner, K., Lutz, M., Preuss, J.D., Reichenbacher, W., 2001. Some ideas about the sources of PM_{10}. *Atmospheric Environment* 35, S23–S33.

LfU (Bavarian Environment Agency), 2014. http://www.lfu.bayern.de/luft/immissionsmessungen/dokumentation/index.htm, accessed in January 2014.

Lund, I.A., 1963. Map–pattern classification by statistical methods. *Journal* of *Applied Meteorology* 2, 56–65.

Makra, L., Mika, J., Bartzokas, A., Sumeghy, Z., 2007. Relationship between the peczely's large–scale weather types and air pollution levels in Szeged, Southern Hungary. *Fresenius Environmental Bulletin* 16, 660–673.

Medina, S., Plasencia, A., Ballester, F., Mucke, H.G., Schwartz, J., grp, A., 2004. Apheis: Public health impact of PM_{10} in 19 European cities. *Journal of Epidemiology and Community Health* 58, 831–836.

Philipp, A., Beck, C., Huth, R., Jacobeit, J., 2014. Development and comparison of circulation type classifications using the COST 733 dataset and software. *International Journal of Climatology*, DOI: 10.1002/joc.3920.

Philipp, A., Bartholy, J., Beck, C., Erpicum, M., Esteban, P., Fettweis, X., Huth, R., James, P., Jourdain, S., Kreienkamp, F., Krennert, T., Lykoudis, S., Michalides, S.C., Pianko–Kluczynska, K., Post, P., Alvarez, D.R., Schiemann, R., Spekat, A., Tymvios, F.S., 2010. Cost733cat–A database of weather and circulation type classifications. *Physics and Chemistry of the Earth* 35, 360–373.

Pitz, M., Schmid, O., Heinrich, J., Birmili, W., Maguhn, J., Zimmermann, R., Wichmann, H.E., Peters, A., Cyrys, J., 2008. Seasonal and diurnal variation of $PM_{2.5}$ apparent particle density in urban air in Augsburg, Germany. *Environmental Science & Technology* 42, 5087–5093.

Querol, X., Alastuey, A., Ruiz, C.R., Artinano, B., Hansson, H.C., Harrison, R.M., Buringh, E., ten Brink, H.M., Lutz, M., Bruckmann, P., Straehl, P., Schneider, J., 2004. Speciation and origin of PM_{10} and $PM_{2.5}$ in selected European cities. *Atmospheric Environment* 38, 6547–6555.

Rost, J., Holst, T., Sahn, E., Klingner, M., Anke, K., Ahrens, D., Mayer, H., 2009. Variability of PM_{10} concentrations dependent on meteorological conditions. *International Journal of Environment and Pollution* 36, 3–18.

Sanchez–Reyna, G., Wang, K.Y., Gallardo, J.C., Shallcross, D. E., 2006. Association between PM_{10} mass concentration and wind direction in London. *Atmospheric Science Letters* 6, 204–210.

Schiemann, R., Frei, C., 2010. How to quantify the resolution of surface climate by circulation types: An example for alpine precipitation. *Physics and Chemistry of the Earth* 35, 403–410.

Seinfeld, J. H., Pandis, S. N., 2006. *Atmospheric Chemistry and Physics from Air Pollution to Climate Change*, John Wiley & Sons, New York, 1232 pages.

Smith, S., Stribley, F.T., Milligan, P., Barratt, B., 2001. Factors influencing measurements of PM_{10} during 1995–1997 in London. *Atmospheric Environment* 35, 4651–4662.

Stadlober, E., Hormann, S., Pfeiler, B., 2008. Quality and performance of a PM_{10} daily forecasting model. *Atmospheric Environment* 42, 1098–1109.

Triantafyllou, A.G., 2001. PM_{10} pollution episodes as a function of synoptic climatology in a mountainous industrial area. *Environmental Pollution* 112, 491–500.

UBA (Federal Environment Agency), 2014.http:// www.umweltbundesamt. de/publikationen/trends–in–air–quality–in–germany, accessed in April 2014.

Weijers, E.P., Schaap, M., Nguyen, L., Matthijsen, J., van der Gon, H.A.C.D., ten Brink, H.M., Hoogerbrugge, R., 2011. Anthropogenic and natural constituents in particulate matter in the Netherlands. *Atmospheric Chemistry and Physics* 11, 2281–2294.

Wise, E.K., Comrie, A.C., 2005. Meteorologically adjusted urban air quality trends in the Southwestern United States. *Atmospheric Environment* 39, 2969–2980.