16th International Conference on
Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes
8-11 September 2014, Varna, Bulgaria

TRENDS OF DAILY VARIOGRAM PARAMETERS DERIVED BY GEOSTATISTICAL ANALYSIS OF PM_{10} TIME SERIES FROM THE AIRBASE AMBIENT AIR QUALITY DATABASE

Oliver Kracht and Michel Gerboles

European Commission - Joint Research Centre, Institute for Environment and Sustainability, Air and Climate Unit, Via E. Fermi, 21027 Ispra (VA), Italy

Abstract: We analyze the temporal variations which can be observed within time series of variogram parameters (nugget, sill and range). Datasets have been obtained from previous geostatistical analysis of country wide datasets of daily air quality data (PM_{10}) over a ten years time frame. Applying the Kolmogorov-Zurbenko filtering method, time series are being decomposed into their short-, mid-, and long-term component. Furthermore, the significance of a long term trend component is investigated by a block-bootstrap-based approach combined with linear regression. It is discussed if within these datasets the times series of nugget variance can provide information about the evolution of the mean measurement uncertainty of the related air pollutant, whereas the sill and the range parameter could contain information about the spatial representativeness of the monitoring stations.

Key words: *air quality monitoring, measurements, uncertainty, geostatistics, variogram, time series, model validation*

INTRODUCTION

A common step in the evaluation of model performance and in model validation is to compare the modelled results to observations obtained from air quality monitoring. In this context, statistical indicators which aim to determine whether model results have reached a sufficient level of agreement with the observations, require an assumption to be made about (i) the measurement uncertainties and (ii) the spatial representativeness of the air quality parameters being used. Quantitative values for the measurement uncertainty are usually derived from experimental work specifically addressing the individual measurement techniques. Quantification of spatial representativeness in conventional practice is frequently based on the evaluation of specific site characteristics and assumptions being made about similarities of those characteristics within the surrounding domains. However, in order to facilitate the thorough exploitation of comprehensive observation datasets, the use of geostatistical techniques can serve as an interesting alternative which can be immediately applied to the monitoring data without need to refer to secondary information. The underlying idea is that within such datasets the nugget variance can provide information about the measurement uncertainty and the micro-scale variability of the related air quality measurements (Gerboles and Reuter, 2010), whereas the sill and the range parameter contain information about their spatial representativeness. However, this information is assumed to be obfuscated by several atmospheric processes which are superimposing the signal of interest on a range of different time scales.

METHODOLOGY

In this exercise, we analyzed the temporal variations observed within time series of variogram parameters (nugget, sill and range) obtained from spherical variogram model fits. The underlying time series data used for this study were obtained within previous projects from the analysis of country wide datasets of hourly PM_{10} data from the AirBase ambient air quality database over a ten years time frame (Gerboles et al. 2014, in prep.). More details about these preceding geostatistical evaluations can be obtained from Gerboles and Reuter (2010). All simulation codes were developed in the R environment (R Development Core Team, 2014). For the scope of this assessment, it was decided to apply the evaluation to the sole stations of background type, but for all area types (urban, suburban and rural).

Table 1. Overview of daily variography sets (individual variogram models fitted to PM_{10} daily values) available.

Country	First Available Fit (year)	Last Available Fit (year)	Available Fits (count)	Accepted Fits (count)
FR	2001	2007	2051	1221
DE	1998	2007	3232	1555
GB	1997	2007	3932	2415
AT	2001	2007	2280	1189
IT	2003	2007	1737	890
NL	2003	2007	1258	1235

Constraints applied for accepting a valid variogram model fit have been:

(I) 1 < nugget < 150 $(\mu g/m^3)^2$, (II) 0 < sill < 10^4 $(\mu g/m^3)^2$, (III) 0 < range < 2 deg, (IV) 0.04 < sill / nugget < $5 \cdot 10^3$

In a first step, the time series of variography parameters were screened for internal consistency by applying a set of constraints (I, II, III, IV, see note in Table 1) for accepting a valid variogram model fit (Table 1). Using the Kolmogorov-Zurbenko (KZ) filtering method (Rao et al., 1997; Eskridge et al., 1997) we then aimed to separate the original time series *X(t)* into its different components (equation 1).

$$X(t) = e(t) + S(t) + W(t) \tag{1}$$

In this conceptualization, the short-term component *W(t)* is attributable to variations of weather and to short-term fluctuations in precursor emissions. The mid-term (seasonal) component *S(t)* can be interpreted as a result of changes in the solar angle (induced variations of emissions and temperature dependencies). The long-term signal *e(t)* can be interpreted to result from long-term changes in overall emissions, pollutant transport, climate, economics, and environmental policies (Wise and Comrie, 2005). *e(t)* is as well supposed to be influenced by evolutions in the operational principles of the monitoring network. In addition, *Baseline(t)* is defined as the sum of the long-term and seasonal component.

Furthermore, the significances of long term trend components of the nugget and the sill effect were investigated from the slope of linear regression lines fitted to each set of the unfiltered time series *X(t)*. This required specific care to be taken because of the serial autocorrelation inherent in these data, which became evident from systematic patterns observed in *S(t)*. A simple linear regression would have all observations within an individual time series of variogram parameter values assumed to be independent. This is likely to give reasonable estimates of the regression coefficients, but to overstate their significance. To obtain more accurate confidence intervals, we chose to combine linear regression with a block-bootstrap-based approach (Künsch, 1989). In this way, the slope parameter was estimated by ordinary linear regression applied to bootstrap replicates chosen randomly from the original time series. We used a variable block length following a geometric distribution with a mean value of 30 days (CF-Interval 1) and 365 days (CF-Interval 2), respectively. Confidence intervals on the 95% level were then estimated by applying a coverage factor of 1.96 to the empirical standard deviation of the bootstrapped slope estimates. After some initial tests, it was considered suitable to perform 10´000 resamples per time series to obtain good stability in the significance level estimates.

RESULTS AND DISCUSSION

Figure 1 and Figure 2 are illustrating the decomposition and trend analysis being applied to the nugget and sill datasets of background stations from six different countries (FR, DE, GB, AT, IT and NL). Note that for the convenience of the discussion the nugget and sill values have been recalculated from variance to 2-times standard deviations and are thus presented in units of $\mu g/m^3$. As a first observation, several data series are revealing a pronounced cyclic behaviour in their seasonal component *S(t)*, and a non-stationarity (change of variance over time) in their short-term component *W(t)*. These effects clearly indicate the presence of temporal variations in the macroscale spatial correlation structures (sill). Furthermore, for the examples of Austria, Italy, Germany and France a phase relationship of the *S(t)* component consisting in a winter increase of the sill effect is observed. It is likely that spatial variability increases in winter time because of local emission caused by heating, sanding and particulate matter re-suspension, as well as by limited air mixing increasing the discontinuity of PM_{10} concentration levels. However, for Austria and Italy this phase relationship is also observed in the nugget effect time series, which might indicate the influence of increasing small scale variability in winter time, too.

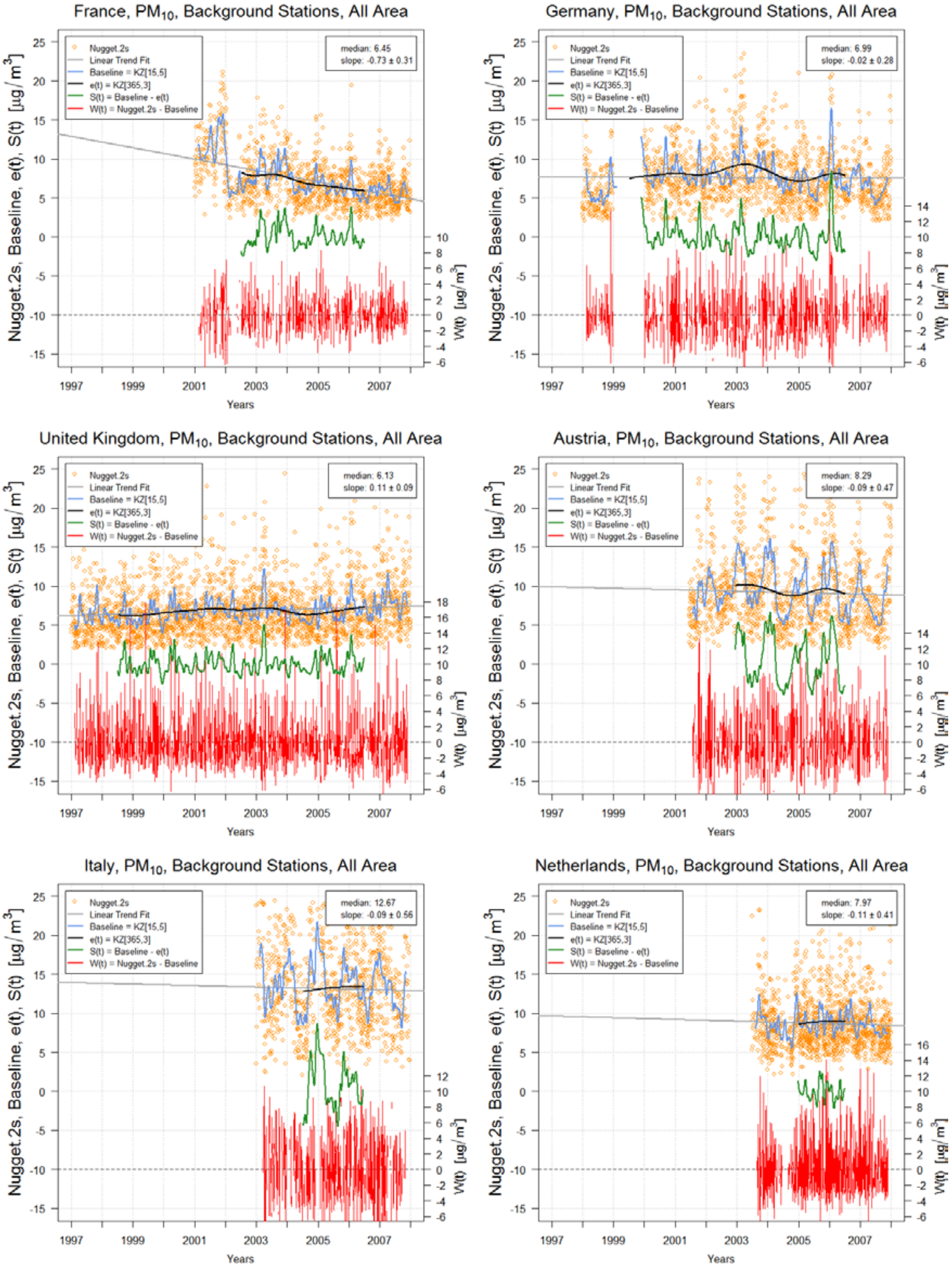

Figure 1. Time series of estimated nugget parameter values from spherical variogram models fitted to PM_{10} daily values of AirBase v.4 background stations. Nugget values have been recalculated from variance to 2-times standard deviations and are presented in units of [µg/m^3]. Note that *KZ[15,5]* has a low pass periodicity of 34 days that gives the baseline air quality. *KZ[365,3]* has a low pass periodicity of 632 days. *W(t)* can be used to characterize the short-term component, *S(t)* for the seasonal component, and *e(t)* reflects the long-term signal and trend. *Baseline(t)* is defined as the sum of the long-term and seasonal component.

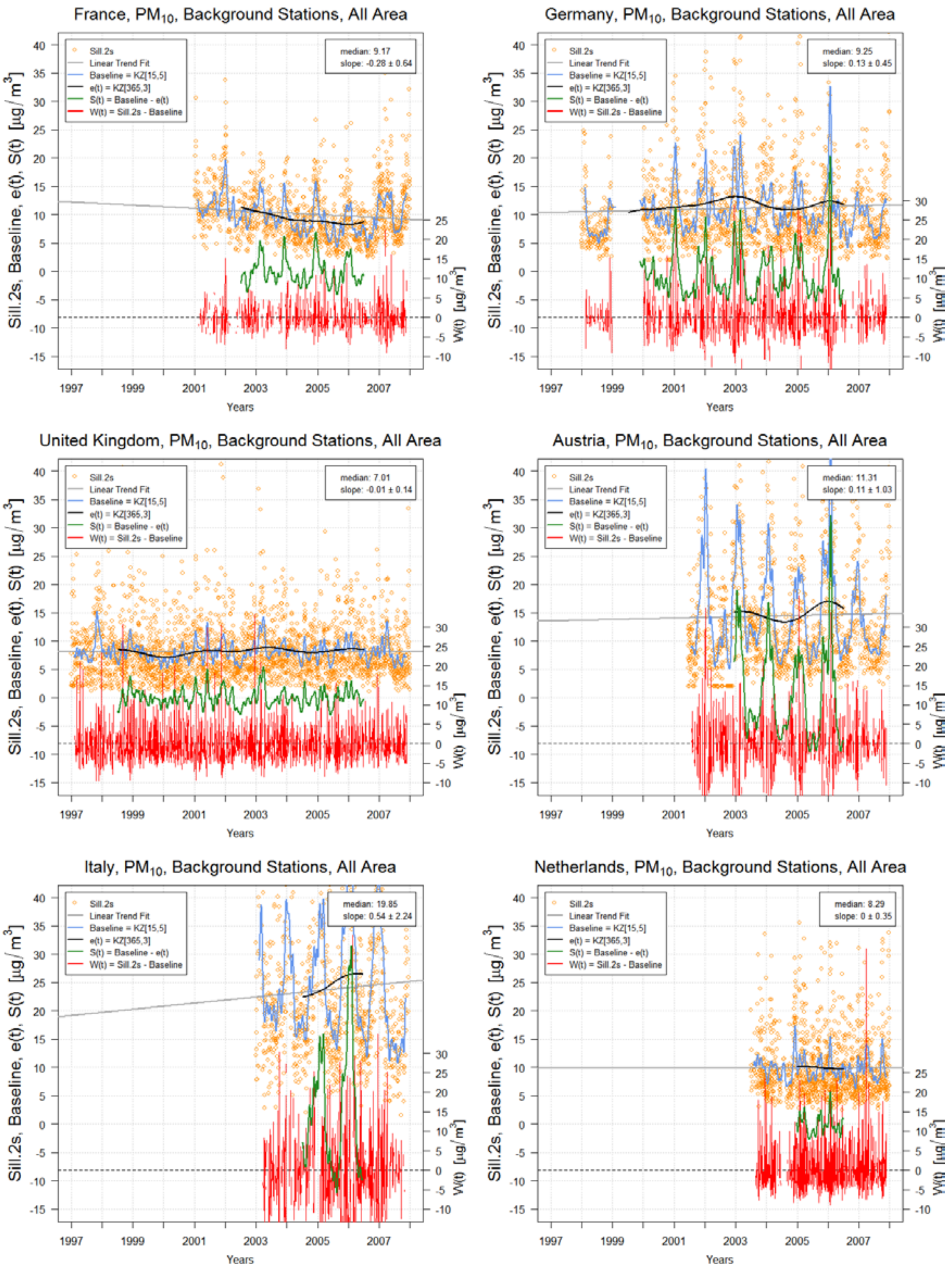

Figure 2. Time series of estimated sill parameter values from spherical variogram models fitted to PM_{10} daily values of AirBase v.4 background stations. Sill values have been recalculated from variance to 2-times standard deviations and are presented in units of [µg/m^3]. Note that *KZ[15,5]* has a low pass periodicity of 34 days that gives the baseline air quality. *KZ[365,3]* has a low pass periodicity of 632 days. *W(t)* can be used to characterize the short-term component, *S(t)* for the seasonal component, and *e(t)* reflects the long-term signal and trend. *Baseline(t)* is defined as the sum of the long-term and seasonal component.

A closer look at the nugget effect time series is indeed of high practical interest, as it can provide information about the evolution of the mean measurement uncertainty of the related air pollutant. From a conceptual point of view, the two dominant causes of the nugget effect should consist of (i) the uncertainty of the measurements, and (ii) the microscale variability. Nevertheless, for the generalized case it is actually difficult to distinguish the proportions contributed by these two components, by using information obtained from geostatistics and time series analysis only. Interpretations of seasonal effects on the spatial correlation length have been discusses in the previous paragraph. However, as the composition and properties of particulate matter and ambient parameters (like temperature and relative humidity) vary in time, they do as well influence the uncertainty of different PM_{10} mass measurements principles (Pernigotti et al., 2013). Thus, the observed short term influences (non-stationarities) and seasonal variations in the nugget time series are as well considered compatible with the assumption of related short term influences and seasonal fluctuations of measurement uncertainty. Note that in order to minimize the influence of small scale variability the data sets used in this exercise have been preselected to comprise background type stations only.

The magnitude of the individual spectral signal contributions to the total variation of the nugget and sill parameter time series can be compared from the overall variance calculated for each of the filter-separated time series. Table 2 provides an overview of the observed total variances. In addition, it also summarizes normalized values (given in parentheses), for which the total values of the different filter-separated spectral components have been adjusted by division with the variance of the raw values. Example given for interpretation, the seasonal component *S(t)* of absolute values is less expressed in both the nugget and the sill data series from France, but more strongly pronounced within the data series from Italy and Austria. Also the short term component *W(t)* has its largest expression in the datasets of Italy and Austria. We consider that this observation can be attributable a lack of continuity due to the stronger topographic roughness and dissection of these two countries. In contrary, within the comparison of the absolute values, Great Britain and the Netherlands are characterized by the least strong total seasonal variation of both the nugget and the sill parameter values. On the other hand, because of this weakly pronounced *S(t)* component, the relative importance of *W(t)* is most prominent for these two countries (normalized values in Table 2).

Table 2. Overall variance of the raw variogram parameter data and of the filtered time series. Note that data have not been de-trended for these calculations. Values in parentheses are normalized by the variance of the raw values series.

Country	Parameter	Raw Values ((µg/m^3)^2)		Baseline ((µg/m^3)^2)		e(t) ((µg/m^3)^2)		S(t) ((µg/m^3)^2)		W(t) ((µg/m^3)^2)	
FR	Nugget (2s)	2.51	(1.00)	1.48	(0.59)	0.14	(0.06)	0.46	(0.18)	1.16	(0.46)
DE	Nugget (2s)	3.28	(1.00)	0.94	(0.29)	0.08	(0.02)	0.73	(0.22)	1.96	(0.60)
GB	Nugget (2s)	2.87	(1.00)	0.43	(0.15)	0.03	(0.01)	0.34	(0.12)	2.16	(0.75)
AT	Nugget (2s)	5.41	(1.00)	1.95	(0.36)	0.06	(0.01)	2.28	(0.42)	3.17	(0.59)
IT	Nugget (2s)	6.50	(1.00)	2.09	(0.32)	0.01	(0.00)	2.34	(0.36)	4.28	(0.66)
NL	Nugget (2s)	2.92	(1.00)	0.47	(0.16)	0.00	(0.00)	0.30	(0.10)	2.21	(0.75)
FR	Sill (2s)	5.98	(1.00)	1.99	(0.33)	0.19	(0.03)	1.33	(0.22)	2.76	(0.46)
DE	Sill (2s)	13.95	(1.00)	3.86	(0.28)	0.14	(0.01)	4.00	(0.29)	7.32	(0.52)
GB	Sill (2s)	6.50	(1.00)	0.73	(0.11)	0.05	(0.01)	0.57	(0.09)	5.04	(0.78)
AT	Sill (2s)	30.47	(1.00)	15.29	(0.50)	0.34	(0.01)	16.44	(0.54)	13.58	(0.45)
IT	Sill (2s)	53.66	(1.00)	26.47	(0.49)	0.54	(0.01)	29.76	(0.55)	26.16	(0.49)
NL	Sill (2s)	8.38	(1.00)	0.94	(0.11)	0.01	(0.00)	0.74	(0.09)	6.63	(0.79)

Results of the trend analysis are presented in Table 3. A significant trend component could only be found in the nugget time series of France (a negative slope of -0.73 ± 0.31 µg/m^3/year) and Great Britain (a weakly positive slope of 0.11 ± 0.09 µg/m^3/year). For all other series the slope of the linear trend component was not significant in consideration of the bootstrapped confidence intervals. However, the expressiveness of trend analysis was partially limited by the relatively short range of valid variogram time series for some of the countries. For future work, an improvement of the robustness of the variogram fit procedures is needed, in order to obtain more comprehensive sets of time series for the trend analysis.

A negative trend in the nugget time series can be interpreted by an improvement of the measurement uncertainty of the monitoring stations over the years, but also by a reduction in small scale variability (change in the nature or quantity of emissions, transported pollution, or atmospheric reactions). Other reasons causing either negative or positive trends might be the increase / decrease of the number of monitoring stations or a change in the station classifications. In conclusion, further investigations are needed to determine if the trends of nugget variance are caused by a decrease / increase of the measurement uncertainty or by long term variations of air pollution and / or meteorological factors.

Table 3. Summary of bootstrap based trend analyses performed on the nugget and sill parameter time series. CF-Interval 1 and CF-Interval 2 are corresponding to resampling with mean block lengths of 30 days and 365 days.

Country	Parameter	Median Value ($\mu g/m^3$)	Trend Slope ($\mu g/m^3$/year)	95% CF-Interval 1 ($\mu g/m^3$/year)	95% CF-Interval 2 ($\mu g/m^3$/year)
FR	Nugget (2s)	6.45	-0.73	± 0.24	± 0.31
DE	Nugget (2s)	6.99	-0.02	± 0.18	± 0.28
GB	Nugget (2s)	6.13	0.11	± 0.07	± 0.09
AT	Nugget (2s)	8.29	-0.09	± 0.41	± 0.47
IT	Nugget (2s)	12.67	-0.09	± 0.51	± 0.56
NL	Nugget (2s)	7.97	-0.11	± 0.30	± 0.41
FR	Sill (2s)	9.17	-0.28	± 0.44	± 0.64
DE	Sill (2s)	9.25	0.13	± 0.36	± 0.45
GB	Sill (2s)	7.01	-0.01	± 0.11	± 0.14
AT	Sill (2s)	11.31	0.11	± 1.04	± 1.03
IT	Sill (2s)	19.85	0.54	± 1.89	± 2.24
NL	Sill (2s)	8.29	0.00	± 0.45	± 0.35

REFERENCES

Eskridge, R. E., J. Y. Ku, S. T. Rao, P. S. Porter and I. G. Zurbenko, 1997: Separating Different Scales of Motion in Time Series of Meteorological Variables. Bulletin of the American Meteorological Society, 78, 1473 - 1483.

Gerboles, M. and H. I. Reuter, 2010: Estimation of the measurement uncertainty of ambient air pollution datasets using geostatistical analysis. JRC Technical Reports 59441, EUR 24475 EN, DOI 10.2788/44902, 38 pp.

Gerboles, M., H. I. Reuter and O. Kracht, in prep. 2014: Feasibility of automatic estimations of measurement uncertainty, detection of abnormal values and check of site classification in large air pollution datasets of AirBase. In preparation for Atmospheric Environment.

Kracht, O., H. I. Reuter and M. Gerboles, 2013: A Tool for the Spatio-Temporal Screening of AirBase Datasets for Abnormal Values. JRC Technical Reports 78437, EUR 25787 EN, DOI 10.2788/81552, 209 pp.

Künsch, H. R., 1989: The jackknife and the bootstrap for general stationary observations. Annals of Statistics, **17**, 1217–1241.

Pernigotti, D., M. Gerboles, C. A. Belis and P.Thunis, 2013: Model quality objectives based on measurement uncertainty. Part II: NO_2 and PM_{10}. Atmospheric Environment, **79**, 869–878.

Rao, S. T., I. G. Zurbenko, R. Neagu, P. S. Porter, J. Y. Ku and R. F. Henry, 1997: Space and Time Scales in Ambient Ozone Data. Bulletin of the American Meteorological Society, 78, 2153 - 2166.

R Development Core Team, 2014: R: A language and environment for statistical computing. http://www.R-project.org/

Wise, E. K. and A. C. Comrie, 2005: Extending the Kolmogorov–Zurbenko Filter: Application to Ozone, Particulate Matter, and Meteorological Trends. Journal of the Air and Waste Management Association, **55:8**, 1208-1216.