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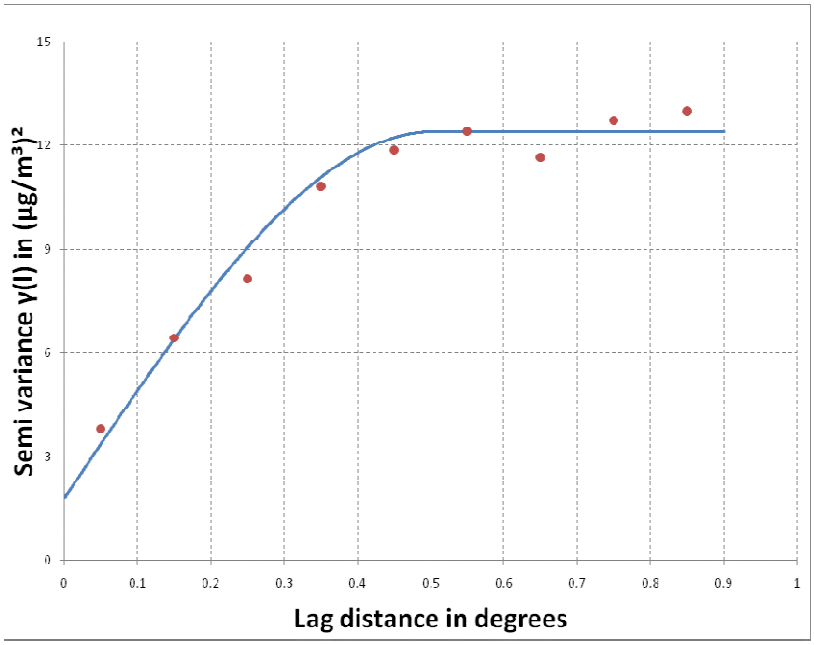
Estimation of the measurement uncertainty of ambient air pollution datasets using geostatistical analysis

Michel Gerboles* and Hannes I. Reuter+

*Joint Research Centre, Institute for Environment and Sustainability, Ispra, Italy
+Gisxperts gbr, Dessau, Germany



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Contact information
Address: Michel Gerboles, Via E. Fermi 2749, I - 21027 Ispra (VA)
E-mail: michel.gerboles@jrc.ec.europa.eu
Tel.: +39 0332 785652
Fax: +39 0332 789931

http://ies.jrc.ec.europa.eu/
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Abstract

We developed a methodology able to automatically estimate of measurement uncertainty in the air pollution data sets of AirBase. The figures produced with this method were consistent with expectations from laboratory and field estimation of uncertainty and with the Data Quality Objectives of European Air Quality Directives. The proposed method based on geostatistical analysis is not able to estimate directly the measurement uncertainty. It estimates the nugget effect from variogram modelling together with a micro-scale variability which must be minimized by accurate selection of the type of station. Based on the results obtained so far, it is likely that measurement uncertainty is best estimated using all background stations of whatever area type. So far the methodology has been used to estimate measurement uncertainty in datasets from 4 different countries independently. This work should be continued for the whole Europe or for background station without national borders. The method has been shown to be also useful to compare the spatial continuity of air pollution in different countries that seems to be influenced by the spatial distribution of the stations (e.g influenced by topography) of each country.

Moreover, the method may be used to quantify the trend of measurement uncertainty over long periods (decades) with the possibility to evidence improvement in the data quality of AirBase datasets.

The implemented outlier detection module would be of interest as the warning system when countries report their measurements to the European Environment Agency. The method could also provides a simple solution to investigate the assignment and accuracy of station classification in AirBase.

Acknowledgement

The authors would like to express their gratitude to Valentin Foltescu, Project Manager, of the Air and Climate Change Programme of the European Environment Agency in Copenhagen – Denmark for reviewing this report.

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Introduction

The European Commission has worked intensively on the implementation of a harmonized programme for the monitoring of air pollutants including arsenic (As), cadmium (Cd), nickel (Ni), benzo(a)pyrene, mercury (Hg), sulphur dioxide (SO_2), nitrogen oxides (NO/NO_2), ozone (O_3), benzene, carbon monoxide (CO), benzene and particulate matter ($PM_{10}/PM_{2.5}$) and lead (Pb) in ambient air. The harmonization program relies on the adopted European Directives 2008/50/EC and 2004/107/EC [1,2]. These directives defines limit and target values for air pollution that should not be exceeded if harmful effects on the population and the environment are to be avoided. Exceedances of these limits may have legal consequences that trigger measures aiming at reducing the exceeded limit values. To avoid those measurement artefacts triggering such measures, the Directives endeavour to improve the quality of the measurements by defining stringent protocols for the sampling/analysis/calibration methods and for the implementation of Quality Assurance/Quality Control programs (QA/QC). They also define data quality objectives (DQOs) that represent the highest allowed relative expanded uncertainty of measurements applied in the region of the Limit Values. The reference methods exhibiting the highest metrological quality of the Directives have been standardized by the European Committee for Standardization (CEN). These standards describe the methodology to be applied for the estimation of the measurement uncertainty. This estimation of the uncertainty of measurements is a long and tedious procedure that may require considerable experimental work. The European Directives allows that two methods of uncertainty estimation are applied following the guidance provided in a CEN report [3]:

- one is based on the Guide to the Expression of Uncertainty in Measurement [4], generally called the direct-approach or GUM method, in which the uncertainty of a measurement is described with

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[1] Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004 relating to arsenic, cadmium, mercury , nickel and polycyclic aromatic hydrocarbons in ambient air. Official Journal L 23, 26/01/2005.

[2] Directive 2008/50/EC of the European Parliament and the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe, Official Journal of the European Union L 152/1 of 11.6.2008

[3] Air Quality—Approach to Uncertainty Estimation for Ambient Air Reference Measurement Methods (CR14377:2002E)

[4] International Organisation for Standardisation, Guide to the expression of uncertainty in measurement, ISBN 92-67-10188-9, ISO, Geneva, 1995

a measurement model that includes several input quantities representing physical variables influencing the measurement. The standard uncertainty of all input quantities must be separately determined and are subsequently combined according to the law of propagation of errors to estimate the uncertainty of the measurement;

- the second method is based on the determination of "Accuracy (trueness and precision) of measurement methods and results" [5], the so called indirect approach, which is concerned exclusively with the uncertainty of measurement methods. The model explaining the measurement Y is based upon the sum of the overall mean, the laboratory bias and the random error. The laboratory bias and random error components are, in quantitative terms, obtained by a collaborative study consisting in an interlaboratory experiment run under reproducibility conditions whose results are treated using the analysis of variance (ANOVA) method.

Nowadays, the methods of estimation of the uncertainty of measurements of ambient air pollution made in Europe are well known. This estimation in carried out on a routine basis by the laboratories reporting their measurements in AirBase, the database maintained by the European Environment Agency (EEA).

From another perspective, it is possible to derive the uncertainty of spatially referenced measurements from the nugget effect of variogram analysis. The nugget effect represents fluctuations of the measurements on a very small scale (tending towards 0). It is often decomposed into the sum of micro-scale variations of the measurand under study and of the measurement errors [6].

In this report, we discuss about the possibility to automatically derive the uncertainty of measurements of ambient air pollutant using a new method based on geostatistical analysis of the spatially referenced datasets present in AirBase, using semi-variogram analysis. This report presents the results of a feasibility study in order to:

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[5] International Standards, 1994, Accuracy (trueness and precision) of measurement methods and results - Part 2: Basic method for the determination of repeatability and reproducibility of a standard measurement method, ISO 5725-2:1994, Geneva, Switzerland.

1. Develop a methodology for downloading geo-referenced air pollution data series of AirBase and for the automatic estimation of the parameters of their spherical variogram;
2. Discuss the uncertainty of measurement evaluated using the estimated nugget variance compared to the DQO for a chosen pollutant, PM_{10};
3. Identify trends over time in the nugget variance to show the variation of the uncertainty of measurement over the last ten years;
4. Create a warning system for assessing the quality of the classification of monitoring stations.

Methodology

AirBase

The European Environmental Agency (EEA) maintains a database on behalf of the participating countries throughout Europe, the EIONET network. Member states (MS) are due to report on the basis of the Council Decision 97/101/EC [7], with amendments 2001/752/EC [8]. Over 6738 stations are in this database, each providing different components of multi-annual time series of air quality measurements starting in 1981. Geographically, the stations are spread all over Europe as seen in Figure 1 with data collected in 36 different countries, including 27 European Union Member States.

The location of measuring stations of the EIONET network is clustered in general due to nature of the measuring network. About 155 parameters are reported in AirBase, ranging from the concentrations of inorganic/organic gases, particulate matter concentrations and wet and dry deposition with their speciation. IN 2008, about 66% of all values in AirBase comes from four different parameters: O_3 (21.2%), NO_2 (17.2%)/NO (8.2 %), SO_2 (18.8%), carbon monoxide (9.4%) and Particulate Matter (PM_{10} 9.0 %, $PM_{2,5}$ 0.5 %, black smoke 1.1 % Total Suspended Particulate – 2.9 % and Pb/Cd/As/Ni 1.5 %).

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[6] Statistics for Spatial Data, Noel A. C. Cressie,, John Wiley & Sons, P. 59.

[7] Council Decision 97/101/EC of 27 January 1997 establishing a reciprocal exchange of information and data from networks and individual stations measuring ambient air pollution within the Member States, Official Journal L 035 , 05/02/1997 P. 0014 - 0022

[8] Commission Decision 2001/752/EC of 17 October 2001 amending the Annexes to Council Decision 97/101/EC establishing a reciprocal exchange of information and data from networks and individual stations measuring ambient air pollution within the Member States.

The quality of the data depends on the chosen measurement method and QA/QC procedures applied by each country. The data in AirBase has undergone additional quality control performed during the upload of the data from the MS to EEAs database using a specifically designed software called DEM (Data Exchange Module). The European Topic Centre on Air and Climate Change (ETC/ACC) is also involved in data quality checking.

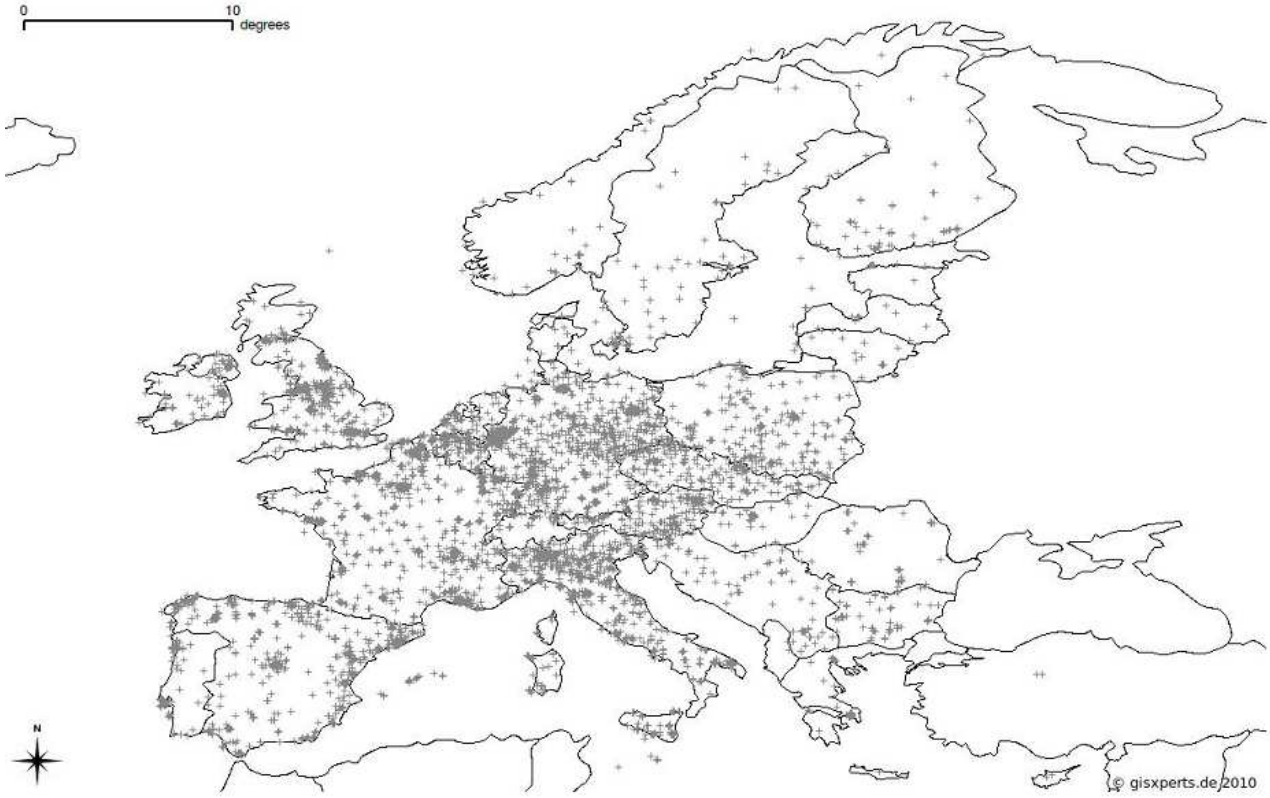

Figure 1: Location of sampling sites reporting data to EEAs Air Quality Database – AirBase - in Europe

Geostatistical method

Geo-statistics is a branch of applied statistics that quantify the spatial dependence and the spatial structure of a measured property. It is based on the regionalised variable theory by which spatial correlation of some properties can be treated [[9]]. Commonly, the geo-statistical analysis includes two phases: the spatial modelling called variography followed by spatial interpolation, the most common one being the Kriging interpolation. In this study, we looked at the first step, focussing on the

[9] Matheron, G., 1963. Principles of geostatistics. Econ. Geol. 58, 1246–1266.

modelling of the semi-variogram (also simply called variogram) that describes the spatial correlation between observations described by the semi variance. The semi variance γ(l) is expressed by equation 1.

$$\gamma(l) = \frac{1}{n(l)} \sum_{i=1}^{n-l} \left[z(x_{i+l}) - z(x_i) \right]^2 \qquad \textit{Equation 1}$$

where n(l) is the number of sample pairs at each distance l (called lags) and $z(x_i)$ and $z(x_{i+l})$ are the values of x, the pollutant of interest, at the locations i and i+l.

The graphical representation of the semi-variance γ(l) as a function of the distance is the semi-variogram or variogram (see an example of variogram in Figure 2). Its main parameters are: nugget, sill and range.

The semi-variograms obtained from experimental data often have a positive value of intersection with the semi-variance axis called the nugget variance or nugget. From this point, the semi variance increases until the variances of the data, called sill, is reached. Up to this point, the regionalized variables in the sampling locations are correlated. They must be considered to be spatially independent at higher distances than this point, called range. The sill is the variogram value at distances beyond the range and, generally, it equals or approaches the population variance. The range provides the distance beyond which variogram values remain constant.

An experimental semi variogram is modelled by fitting a simple function to the data pairs l_i, $\gamma(l_i)$. Linear, spherical or exponential models are often used [10]: The spherical model is the most commonly used one (see Equation 2). For example, a spherical model is fitted to the experimental data of the variogram shown in Figure 2.

$$\begin{cases} if \quad l \le a \quad \gamma(l) = C_0 + C_1 \left[1.5\, l/a - 0.5 (l/a)^3 \right] \\ if \quad l > a \quad \gamma(l) = C_0 + C_1 \end{cases} \qquad \textit{Equation 2}$$

Where C_0 is the nugget variance, C_1 is the difference between the Sill C and the nugget variance C_0 ($C = C_0 + C_1$), l is the lag distance and a is the range.

The nugget effect is the value of the theoretical variogram C_0 at the origin of the variogram ($h \rightarrow 0$) and is thus unknown. The empirical nugget is estimated by extrapolating the empirical variogram towards h=0. It consists of the short-scale gradient of concentrations in the pollutant at distances much shorter than the sampling distance or called micro-scale variation and of a stochastic measurement

uncertainty mainly the sampling and analytical variability, which should be true uncorrelated random noise [10]. The nugget variance s^2_{nugget} can be expressed using Equation 3:

$$s^2_{nugget} = s^2_{meas} + s^2_{sc} \qquad \textit{Equation 3}$$

where s^2_{meas} is the variance associated with the sampling and analytical variability and s^2_{sc} is the variance due to micro-scale variability. Equation 3 is based on the assumption that s^2_{meas} and s^2_{sc} are not correlated. In fact, for some atmospheric parameters, small changes in location can cause significant changes in the concentration level of the pollutant. For example, if one moves from a ridge to a valley, pollution may change quickly and at a scale at which we cannot predict because of sparse observations.

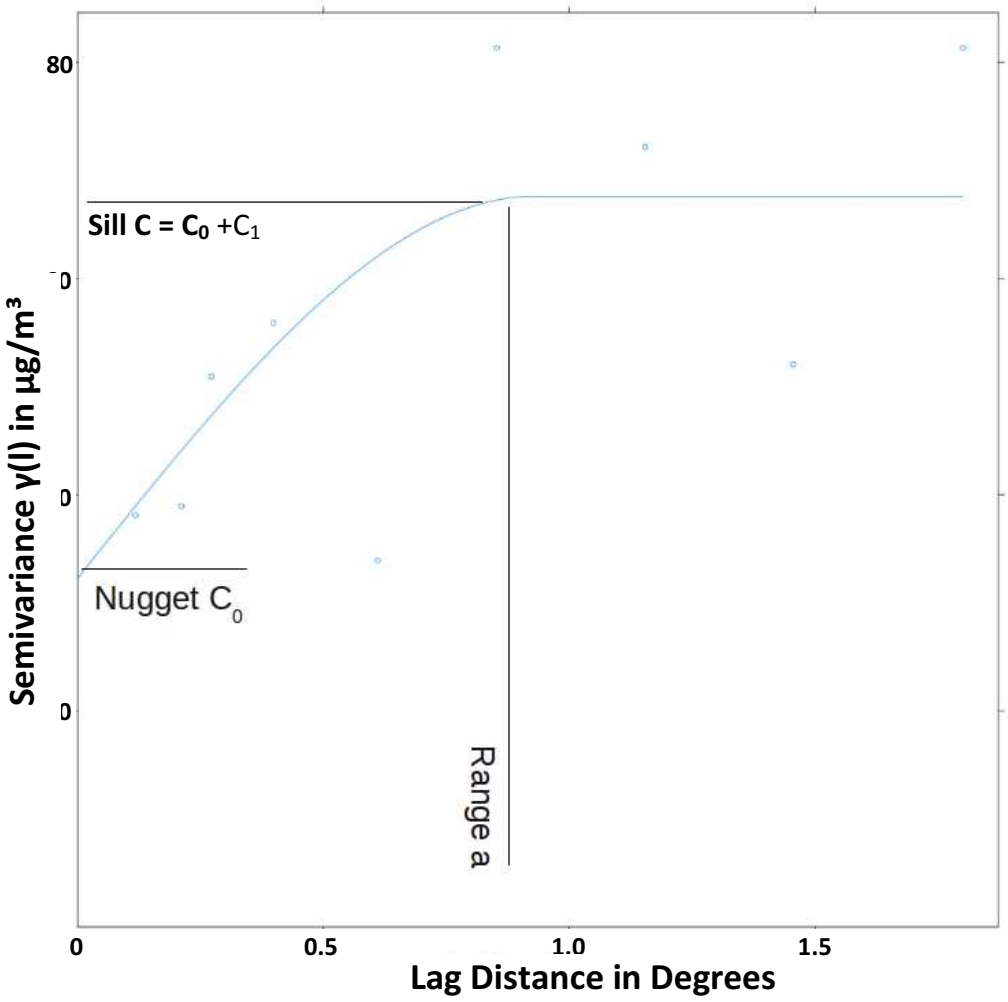

Figure 2: Example of a semi-variogram for PM10 stations in Europe, showing the nugget (local spatial variability and measurement uncertainty), the range (the extent of spatial variability) and the sill (the total variability in the dataset or the given extent). The gray line represents a example of a fitted semi-variogram function.

[10] Isaak, E. H., Srivastava, R. M., 1989, An Introduction to Applied Geostatistics. Oxford University Press, New York.

The estimated value of s^2_{nugget} cannot be decomposed in measurement error variance s^2_{meas} and micro-scale variance s^2_{sc} without further information or prior belief. However, the square root of s^2_{nugget} overestimates the uncertainty of measurement according to the extent of s^2_{sc}, the micro-scale variance. It is then necessary to control the micro-scale variance so that the nugget variance, used as a surrogate of the uncertainty of measurement, will only slightly overestimate the nugget variance. In this study, the micro-scale variance is minimized by determining the nugget variance of subsets of all available sampling sites selected according of their classification: background, traffic and industrial stations in order to minimize the micro-scale variance.

The nugget variance is estimated by the intersection of the fitted model with the Y-axis of the variogram. However, fitting different model (linear, spherical, exponential, power or a combination of these) to the same experimental data set would have resulted in estimating different values for the same nugget variance. Even more, fitting different type of models to different data sets would have ended up in nugget variances that would have not been comparable anymore. In order to be consistent in the method used to estimated the nugget variance of several data sets and hence be able to compare them, it was decided to always fit a spherical model to all the prepared variograms.

When the uncertainty of measurement is determined using the direct approach, one starts by listing all the possible contribution arising from different parameters (sampling, calibration ...) to be able to combine them afterwards. One nice feature of estimating uncertainty using the nugget variance is that all these contributions can be included by selecting appropriate sets of sampling sites that would include different type of sampling lines, method of measurements/maintenance/calibration, equipment brand, etc. The simple fact to select different sets of the sampling sites results in a wider estimation of all parameters contributing to the uncertainty of measurements.

However, one should always keep in mind that there is a risk to attribute some contribution of the micro-scale variation to the uncertainty of measurements and that this micro-scale variance might be magnified by the heterogeneity of the sampling sites.

Notably one type of parameter contributing to the uncertainty of measurement that cannot be detected by the nugget Variance consists in systematic bias that would be present at all sampling sites e.g. a bias of the measurement methods or chemical interference. The presence of this type of systematic bias in all the selected stations is nevertheless highly unlikely because of the very diverse implementation of sampling, analytical and calibration methods managed by different laboratories implementing different QA/QC procedures for a whole set of monitoring stations.

Finally, the method of estimation of the uncertainty of measurement proposed in this study relies on the modelling of the variogram based on the data pairs consisting of lags and semi variances. The

nugget variance will depend on the semi-variance at the smallest lag distance of each variogram. When the nugget variance is estimated per country using data sets whose smallest lag are different per country, one cannot exclude a lack of homogeneity of the extrapolation of the spherical model on the Y-axis.

Development of a methodology for downloading geo-referenced air pollution data series of AirBase and automatic estimation of the nugget variance of their variogram;

A PostgresSQL DB V8.3 was installed on a 64bit Ubuntu 9.4 distribution (http://www.ubuntu.com/) with 8 GB RAM and 4 processors. Data were loaded using a self developed shell script (see Appendix II: Developed Shell Script for Data import into the PostgresSQL Database), which automatically loaded data if their measurement quality flag available in AirBase was set to 1. Each data record was characterized using sample date, measurement value, station code and component code. For performance issues each component was indexed on time and station code. Station data locations were converted into a shapefile and loaded directly into Postgress. Indexing was performed on station code and the geometry column. Several iterations where performed to determine which combination of index / requests delivered the fastest return of data.

For further data analysis, the open source software R V 2.8.1 (www.r-project.org) with several extensions (Rdbi + RdbiPgSQL for Database access, gstat, sp, automap for semi-variogram calculations) has been used. Out of this analysis, a whole toolbox of algorithms (see Appendix I: Developed R- Routines) has been developed which allow to process and calculate different kinds of analysis (e.g. raw and fitted semi-variograms, outlier calculation, general statistics and fitted functions to the time series), all with respect to the analysis of the air quality datasets. In general, the data analysis consisted of three different steps: loading the data from the DB into memory, performing the necessary calculations, writing semi-variogram results into ASCII files for further analysis.

Data connections between R and the Postgress DB have been established using the RDBI driver. The time of this driver delivering data is approx 2% compared to the time the ODBC Database drivers would deliver data. Stations were selected, joined with the corresponding location data table and imported into R. All datum data were converted into Julian day to be able to perform temporal and spatial selections. If data needed to be normally distributed for applying the outlier test (see below), a natural logarithmic transformation was performed. Additionally, to improve mathematical stability, an offset has been added before the log transformation derived by the absolute minimum value of the dataset + 0.5 to avoid undefined values (a log of zero or of negative values is not defined).

Semi-variogram analysis was performed using an automatic semi-variogram fitting provided by the automap toolbox (Hiemstra et al, 2008) [11]. This toolbox was adapted by limiting the maximum lag distance to a total search radius of 2° of latitude and longitude corresponding to ~ 220 km long. This method automatically could test/fit different semi-variogram models and fits semi-variogram parameters based on a given dataset. Usually, the algorithm determines the boundaries for the lags by determining the spatial boundary and dividing it by the size of the area. However, as the distribution of stations is clustered, this algorithm delivered at times lag boundaries, which were not corresponding to the spatial autocorrelation of the underlying data. For example, fitting lag distances to Spanish data without limits resulted in a maximum lag distance of ~10° due to some stations on the Canary Islands as well as on the mainland. By introducing a limiting factor, the spatial variability of the mainland which should have been up to a maximum of ~2° for Spain could be maintained. Another typical example is given by stations placed behind a mountain range, while all other stations form a cluster. Therefore, we let the algorithm estimated the boundaries, while limiting the maximum distance to preset value of 2°. Thereby we effectively ensured that the lag boundaries where always within the autocorrelation range.

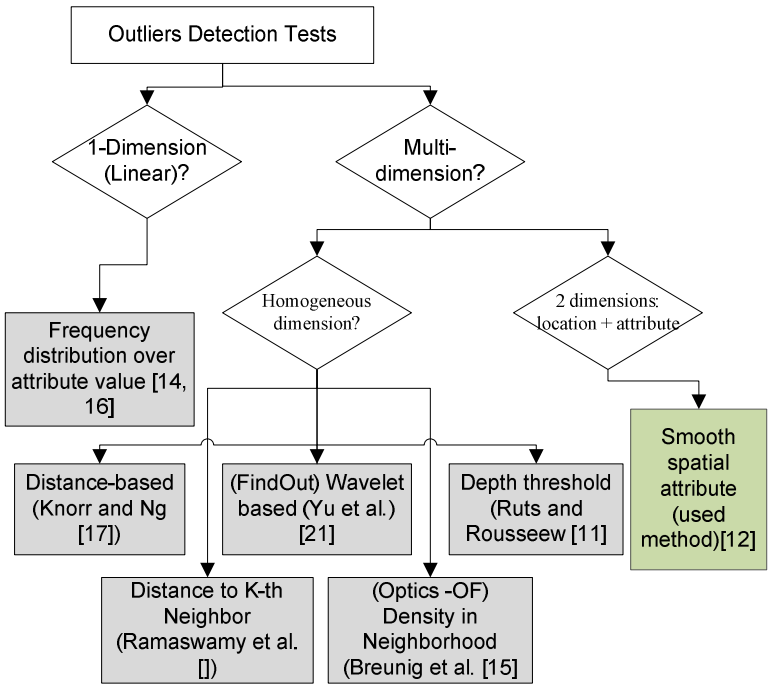

Figure 3: Several methods to detect outliers in 3D datasets (Figure taken from Chang-Tien Lu[12])

In the beginning of the analysis, a couple of hundred semi-variograms were computed: It became clear that outliers influenced the semi-variogram calculation, which rendered analysis of spatial dependency

[11] Hiemstra, P.H., Pebesma, E.J., Twenhöfel, C.J.W and G.B.M. Heuvelink (2008). Automatic real-time interpolation of radiation hazards: a prototype and system architecture considerations. International Journal of Spatial Data Infrastructures Research, vol 3, p 58-72

using semi-variance analysis questionable. The outliers influenced the fitting of the semi-variogram function and led to an artificial increase of the nugget effect.

Therefore an outlier procedure was implemented based on already existing literature. Chang-Tien Lu [[12]] have outlined and classified several algorithms [13,14,15,1617,18,19,20,21] as seen in Figure 3. Two families of outlier detection methods can be distinguished. First the ones which calculates statistic of the distribution of pollutant in one dimension and ignore geographical location [14, 16]. The second family, the spatial-set outlier detection methods, consider both attribute values and spatial relationships. Within this family we used the "Smooth Spatial Attribute method" [12] that was developed for the identification of outliers in traffic sensors. This method is thought to be fit for the identification of outliers in a given homogeneous dataset of air quality data that represents in a similar way a quantity measured in time and space.

The Smooth Spatial Attribute method relies on the definition of a neighbourhood for each air pollutant measurement. It corresponds to a spatio-temporal domain limited in time (+/- 1 day) and distance (+/- 1 degree) around location *x*. The neighbourhood is better understood by observing the diagram in Figure 4. The objective of the method is that within a given spatio-temporal domain in which the value of the attribute values of neighbours have a relationship due to the distribution/transport/emission and reaction of air pollution, outliers will be detected by extreme value of their attribute value compared to the attribute value of their neighbours. The main computation cost of the method is dominated by disk Input/Output cost and the main constrain of the method is the normality of the distribution of the attribute values of neighbours.

In the following text, we called *x* the concentration of a pollutant or its location. Within each neighbourhood, several measurements of the same compounds at different locations and time $x_{xi,yi}$ are

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[12] Chang-Tien Lu, Dechang Chen, Yufeng Kou, "Detecting Spatial Outliers with Multiple Attributes," ictai, pp.122, 15th IEEE International Conference on Tools with Artificial Intelligence (ICTAI'03), 2003.

[13] M. Ankerst, M. Breuning, H. Kriegel and J. Sander. Optics: Ordering points to identify the clustering structure in Proceedings of the 1999 ACM SIGMOD International Conference on Management of Data, Philadelphia, Pennsylvania, USA, pages 49-60, 1999.

[14] V. Barnett and T. Lewis. Outliers in Statistical Data. John Wiley, New York, 3rd Ed. 1994.

[15] M. Breuning, H. Kriegel, R. T. Ng and J. Sander. OPTICS-OF: Identifying Local Outliers in Proc. Of PKDD '99, Prague, Czech Republic, Lectures Notes in Computer Science (LNAI 1704), pp 262-270, Springer Verlag, 1999.

[16] R. Johnson. Applied Multivariate Statistical Analysis, Prentice Hall, 1992.

[17] E. Knorr and R. Ng. Algorithms for Mining Distance-Based Outliers in Large Datasets in Pric. 24th VLDB Conference, 1998.

[18] M. Kraak and F. Ormeling. Cartographer: Visualization of Spatial Data. Longman, 1996

[19] F. Preparata and M. Shamos. Computational Geometry: An Introduction. Springer Verlag, 1998.

[20] I. Ruts ans P. Rousseeuw. Computing Depth Contours Of Bivariate Point Clouds. In Computational Statistics and Data Analysis, 23:153-168, 1996.

[21] D. Yu, G. Shekholeslami and A. Zhang. Findout: Finding Outliers in Very Large Datasets. In Department of Computer Science and Engineering State University of New York at Buffalo Buffalo, Technical report 99-03, http://www.cse.buffalo.edu/tech-reports/, 1999.

available. Equation 4 allows computing a weighted average of all available measurements $x_{xi,yi}$ within each neighbourhood where the weights correspond to the inverse spatial and time distance between $x_{xi,yi}$ and x.

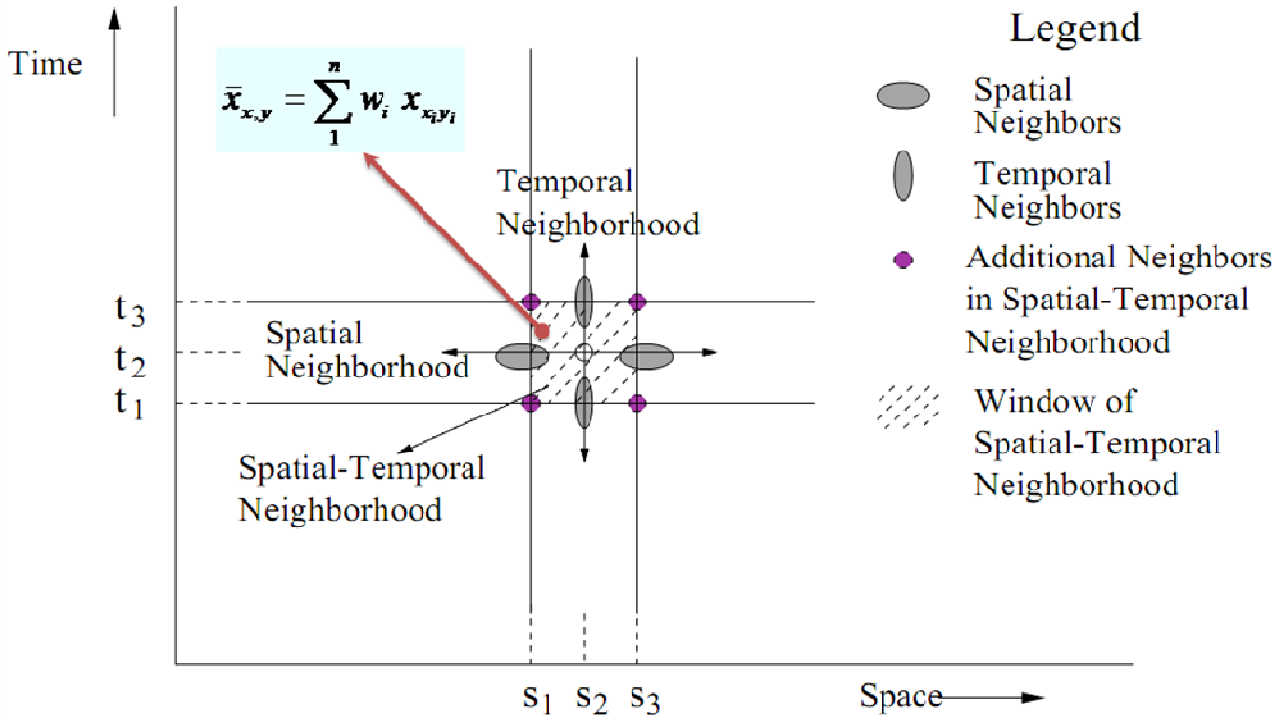

Figure 4: Spatial and temporal outliers – definition of neighborhood [12]

After a log-transformation of non - Gaussian data within any neighbourhood, we computed the differences *Sx* between value at *x* and the average of its neighbourhood for each measurement according to Equation 5.

$$\bar{x}_{x,y} = \sum_{1}^{n} w_i \, x_{x_i y_i} \qquad \textit{Equation 4}$$

$$Sx = fx - \bar{x}_{xi,yi} \qquad \textit{Equation 5}$$

$$z_i = \frac{x_i - \bar{x}}{s} \qquad \textit{Equation 6}$$

$$z_i > \theta \qquad \textit{Equation 7}$$

Then within each neighbourhood, the *Sx* values were normalised to center data at 0 with a standard deviation of 1 using Equation 6 in which $\bar{x}$ and s are the weighted average and weighted standard deviation of all possible *Sx* values within any neighbourhood. Finally, the test for detecting an outlier, given in Equation 7, searches for z_i values exceeding a threshold value consisting in the moving average of five consecutive z_i values plus a threshold value of 2 corresponding to a confidence interval

in which 95 % of z_i values would lay. In contrast to the paper by Lu [12], we did not use an absolute value of the z-transformation due to the fact that the sign of the outlier is of interest to us as we want to understand if a station is measuring to low quantities or to high quantities compared to the its neighbourhood stations with the same classification (urban, background, traffic ..). By plotting the result of the zi against the moving average of the z- plus the threshold value, outliers were identified. An example is given in Figure 5 for an Austrian station monitoring PM_{10} with the identification of 19 outliers of daily values in 2007.

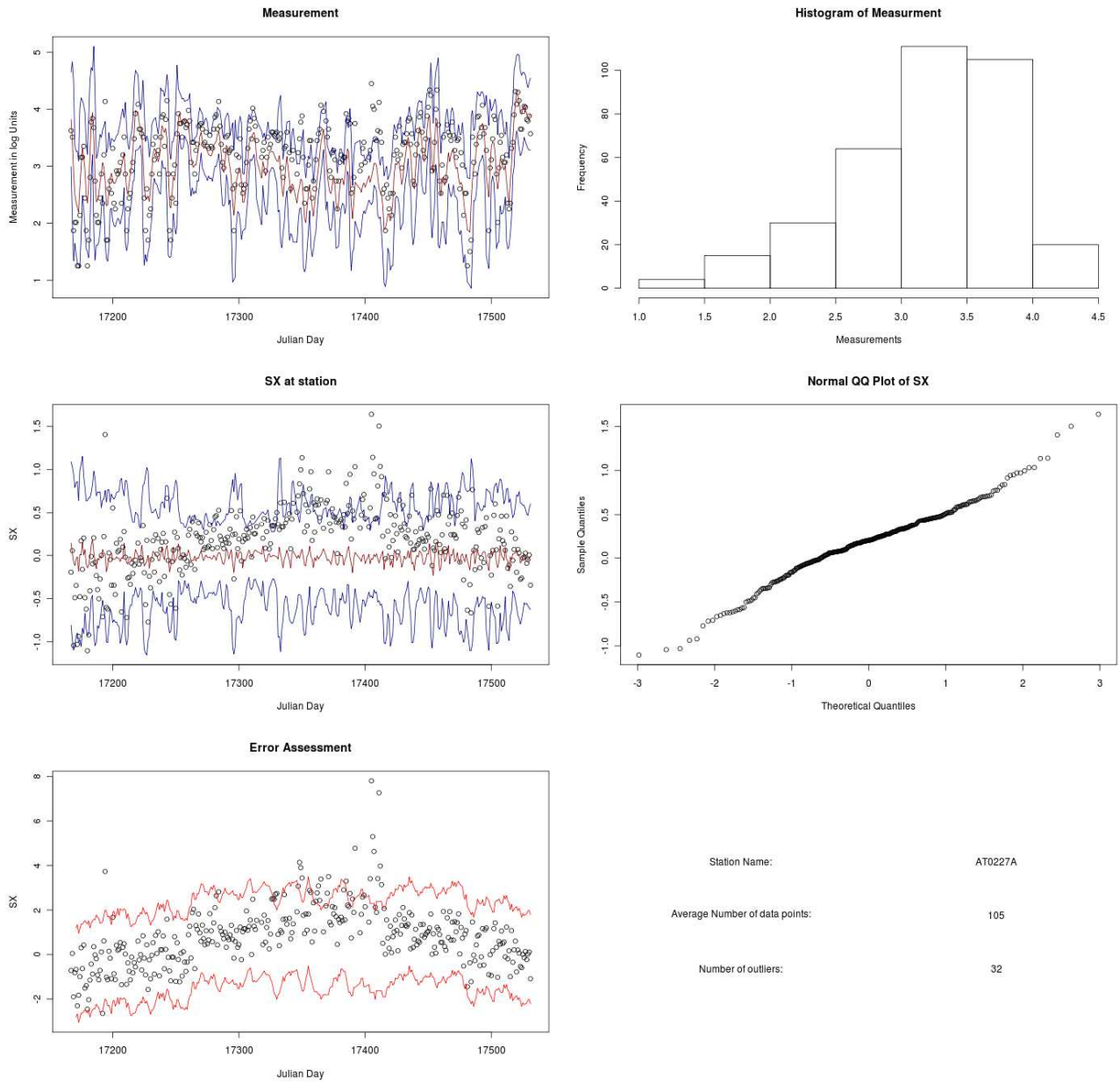

Figure 5: Outlier analysis for Austrian Station AT0227A (Großenzersdorf/Glinzendorf) using the developed method. The Station Values (black circles) are shown with the average of the surrounding stations circle(red line) +/- 2 standard deviation (SD) (Top Left), the histogram of the (log transformed) measurement value for normality (Top Right); the Sx values for the station (black) with respect to the surrounding Mean (Red) and SD(blue)(Middle Left); the Quantile distribution of the Sx values (Middle Right) to see if the distribution contains any large deviations; the z_i values of the station (black) plotted against the specified threshold (Lower Left); and in the lower right corner the average number of stations in the surrounding used for the calculations and the number of identified outliers out of one year .

Finally, the methodology developed for the estimation of the uncertainty of measurement based on the nugget variance can be described by the flow chart given in Figure 6.

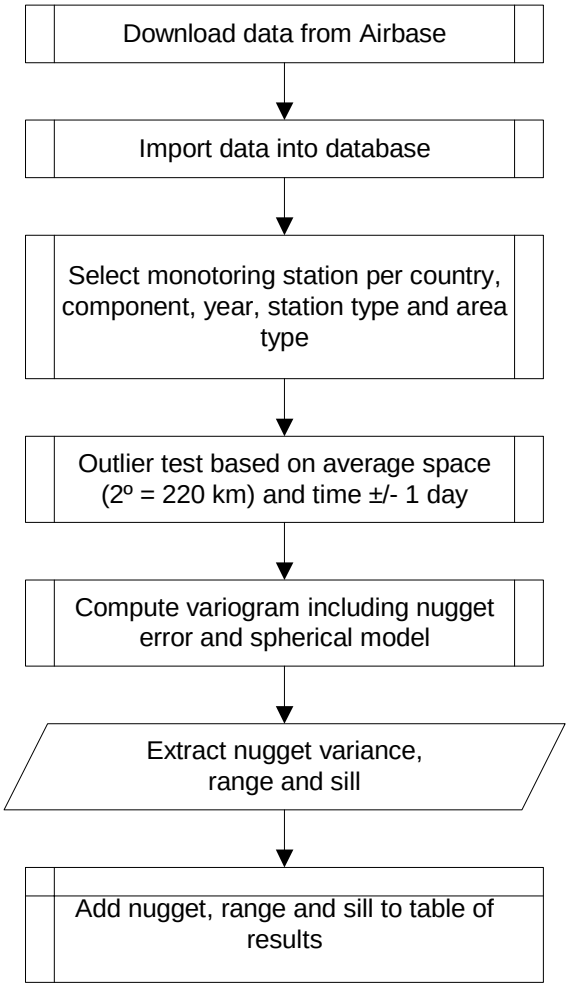

Figure 6: Flow chart representing the different steps of the developed methodology for the estimation of uncertainty of measurements

If more than 20 stations were available at any given time step (e.g. day), a semi-variogram analysis was performed consisting of a nugget effect and a spherical model. An example of the effect of discarding outliers producing a decrease of the nugget effect and sill is presented in Figure 7 for the rural background station in Germany for PM_{10} in 2007.

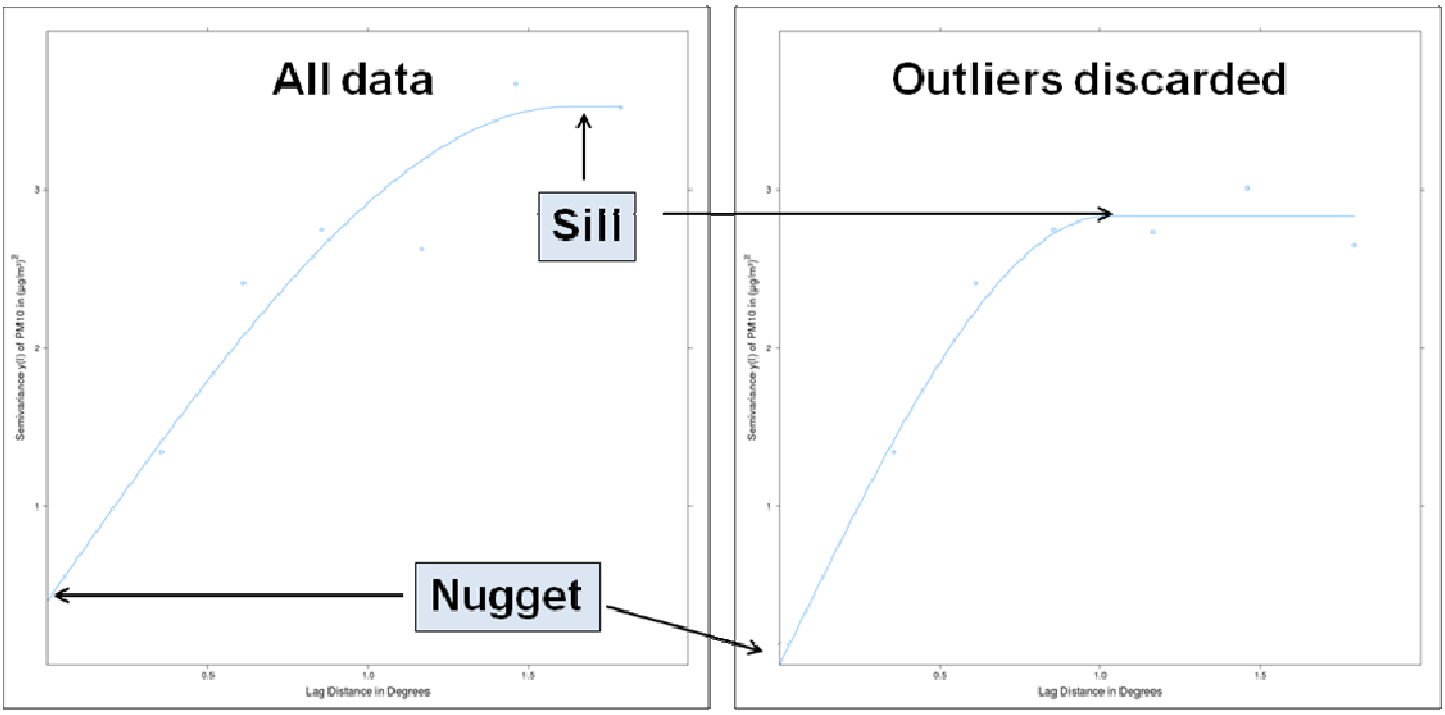

Figure 7: Decrease of the nugget effect and sill of the semi variogram for Julian Day 17176 in Germany on rural background stations by discarding outliers (left all monitoring stations are used, right outliers are discarded)

Estimation of the measurement uncertainty using the nugget variance

For PM_{10}, our pollutant of interest, the number of monitoring stations increased in all countries whatever station or area type as shown in Table 1. In 2007, Germany had a total of 358 stations (among which 155 traffic stations in urban areas), France had 238 stations (among which 126 background stations in urban areas), Italy had 141 stations (among which 86 traffic stations in urban areas) and Austria had 87 stations with 59 background stations in urban areas. The number of stations included for which data are present in AirBase for all possible combination of station and area type is given in Table 2. As the micro micro-scale variability estimated form the geostatistical analysis of Industrial stations was expected to be higher than with urban and industrial stations, it was decided not to select this type of station in the analysis.

As mentioned in Introduction, the European Directives defines data quality objectives (DQOs) that represent the highest allowed expanded uncertainty of measurements in percentage of the Limit Value. Table 4 gives for each pollutant the combined uncertainty in µg/m³ corresponding to this percentage. For PM10, we will remember that the combined uncertainty corresponding to the European Data Quality Objective is 5 µg/m³.

For Year 2007, we calculated the averages of all daily measurement uncertainties estimated by the square root of the nugget variance and ranges of the semi-variograms for different station and area types for Austria, (AT), Germany (DE), France (FR) and Italy (IT). The values are given in Table 4.

Table 1: Number of Stations for the most abundant parameters of AIRBASE in 2002 and 2007 for traffic (TR) and background (BG) type stations for urban (UR), rural (RU) and suburban (SU) areas.

		SO_2				O_3				NO_2				PM10			
		AT	DE	FR	IT	AT	DE	FR	IT	AT	DE	FR	IT	AT	DE	FR	IT
2007	TR-UR	31			94	7	27	2	47	29	26	44	196	28	155	27	86
	BG-RU	87		1	21	50	69	62	41	34	11	30	37	19	52	11	8
	BG-UR	49	1		42	17	82	164	69	22	14	183	90	20	86	126	31
	BG-SU	52		5	25	23	72	147	41	24	27	115	48	20	65	74	16
2002	TR-UR	58			29	6	41	6	18	27	19	39	41	15	84	26	17
	BG-RU	103		2	6	53	72	53	15	37	9	36	13	12	35	11	-
	BG-UR	55	2		15	18	92	130	21	23	19	147	21	9	79	101	10
	BG-SU	54	1		10	21	84	136	19	22	36	119	12	11	52	70	-

Table 2: Classification of station and area in Austria, Germany, France and Italy.

Station type	Area type	Number of station	Station type	Area type	Number of station
Industrial	suburban	524	Background	urban	1658
Industrial	urban	263	Background	suburban	1213
Industrial	*	1	Background	unknown	35
Industrial	unknown	17	Background	rural	814
Industrial	rural	298	Unknown	rural	2
Traffic	unknown	5	*	*	4
Traffic	suburban	248	Unknown	unknown	32
Traffic	urban	1493	*	suburban	1
Traffic	rural	44	Unknown	urban	67

Stars denote missing station types.

First of all, they are consistent with the expected uncertainty in field. They are generally lower than the data quality objective expressed as combined uncertainty of the Limit Value (5 µg/m³). It is likely that the estimation of the measurement uncertainty using traffic type station is overestimated by a micro-scale variation included in this type of station. For urban traffic, Austria shows the lowest values for nugget, followed by Germany and France with double the amount effect. Italy shows the highest amount for the nugget.

Background stations do not show such clear patterns. The nugget values of Austria are double compared to the observed values for Germany and France; while Italy shows nearly three fold values of these nuggets. The reason for the nugget differences is unclear. A clear attribution to different station networks or different traceability of standard strategies seems not be possible but should be investigated. A second factor could be the different spatial distributions of the station networks for background stations influencing the computed results. We believe that the best estimated of the

measurement uncertainty is the one found for “all background stations” with enough stations including different factors in the estimation of nugget effect. For Austria, France and Germany the nugget effect is lower than the Limit Value while the Limit Value is slightly exceeded in Italy.

Table 3 Data quality objectives for ambient air quality assessment and its corresponding combined uncertainty at the limit/target values (Directive2004/107/EC and 2008/50/EC)

	Data Quality Objectives (relative expanded uncertainty)	**Limit/Target value**	**Corresponding combined uncertainty**
Sulphur dioxide, SO_2	*15 %*	Yearly :350 µg/m³ *Daily : 125 µg/m³*	26,2 µg/m³ *7,8 µg/m³*
Nitrogen dioxide NO_2	*15 %*	Yearly: 40 µg/m³ *Daily: 200 µg/m³*	3 µg/m³ *15 µg/m³*
Ozone, O_3	*15 %*	*8 hr mean: 120 µg/m³*	*9 µg/m³*
Carbon monoxide, CO	*15 %*	*8 hr mean: 10 mg/m³*	*0,75 mg/m³*
Benzene	*25 %*	*Yearly: 5 µg/m³*	*0,63 µg/m³*
PM_{10}	*25 %*	***Yearly: 40 µg/m³*** Daily: 50 µg/m³	**** 5 µg/m³*** 6,3 µg/m³
$PM_{2,5}$	*25 %*	*Yearly: 25 µg/m³*	*3,1 µg/m³*
Lead, P	*25 %*	*Yearly: 0,5 µg/m³*	*0,063 µg/m³*
Benzo(a)pyrene	*50 %*	*1 ng/m³*	*0,25 ng/m³*
Arsenic, As	*40 %*	*6 ng/m³*	*1,2 ng/m³*
Cadmium, Cd	*40 %*	*5 ng/m³*	*1 ng/m³*
Nickel, Ni	*40 %*	*20 ng/m³*	*4 ng/m³*

** expanded uncertainty for PM_{10} that will be used in this study*

Unfortunately, not all combinations delivered enough data points (e.g. less than 300 days or less than 20 stations), therefore no average values could be reported e.g. background-urban (BG-UR). For comparison we also report the values for all stations type – station area type in Table 2.

The range values increase in a different manner than the nugget effect. Observing the ranges for the “All background” stations, we can see that Austria and Italy have a range about 0.9º while the one of Germany and France is about 1.2º which would imply that the long range spatial dependency also increase. In fact, it was expected that the spatial continuity of the PM_{10} concentrations would be higher for Germany/France as a result of the higher number of stations in low land areas compared to Austria and Italy.

A conclusion drawn from the different results are:

> Austria as a country with medium background and low urban traffic nugget also shows the lowest overall nugget as well as range effect.

Italy, shows the largest nugget parameters across all combinations. The reason for this is still unclear.

Station classification appears to influence the nugget and range results in different MS in various degrees - a more precise classification delivers an increased spatial dependency

The spatial distribution of the station network (e.g. the Austrian/Italian Mountain Valley situation versus a German/France lowland situation) might have influenced the quantified nugget and range values as well. However the size of this effect is uncertain.

Table 4 Averaged Daily Nugget and Range Values for the year 2007 for the EU Member states Austria (AT), Germany (DE),France (FR) and Italy (IT) for the Station Type - Station Area Type Combinations Traffic-Urban (TR-UR), Background-rural (BG-RU), Background-Urban (BG-UR), Background-Suburban (BG-SU), all Background stations (BG-ALL), and all stations (ALL-ALL).

Station Combination		**Nugget and Range Values for the Year 2007 for PM_{10}**			
	St - Iso	AT	DE	FR	IT
Nugget in µg/m³	TR-UR	3.8	5.5	5.3	9.3
	BG-RU		1.9		
	BG-UR		2.8	1.6	7.5
	BG-SU		1.9	1.9	
	BG-ALL	4.0	2.7	3.1	7.0
	ALL-ALL	4.2	6.1	6.2	8.9
Range in spherical degrees	TR-UR	0.93	1.22	1.06	1.20
	BG-RU		1.19		
	BG-UR		1.06	0.91	1.10
	BG-SU		0.94	1.16	
	BG-ALL	0.9	1.17	1.2	0.89
	ALL-ALL	0.91	1.01	1.16	1.11

Trend over time of the measurement uncertainty indicated by the nugget effect

While the results for year 2007 give only a short snapshot in time, we investigated how the nugget and range change over time. The same geostatistical analysis as for year 2007 was performed over the timeframe 1997-2007.

By plotting the nugget effect of PM_{10}versus time, it is possible to observe that the slope of regression line (-0,002) for Germany (see Figure 8) shows a decrease that indicates a slight improvement of the measurement uncertainty between 1997-2007. The same decrease of measurement uncertainty is stronger for Austria as shows the slope of the nugget variance versus time (-0,010, see Figure 9). However one should note that the initial nugget variance of Austria in 1997 was higher than the one of Germany. This is evidenced by the intercept of Austria of 213, compared to 45 for Germany (see Figure 8 and Figure 9). Figure 8 also shows an annual effect of changes in spatial distribution in Austria that is clearly visible in the yearly increase in small scale variability (i. e. nugget effect) during the winter months.

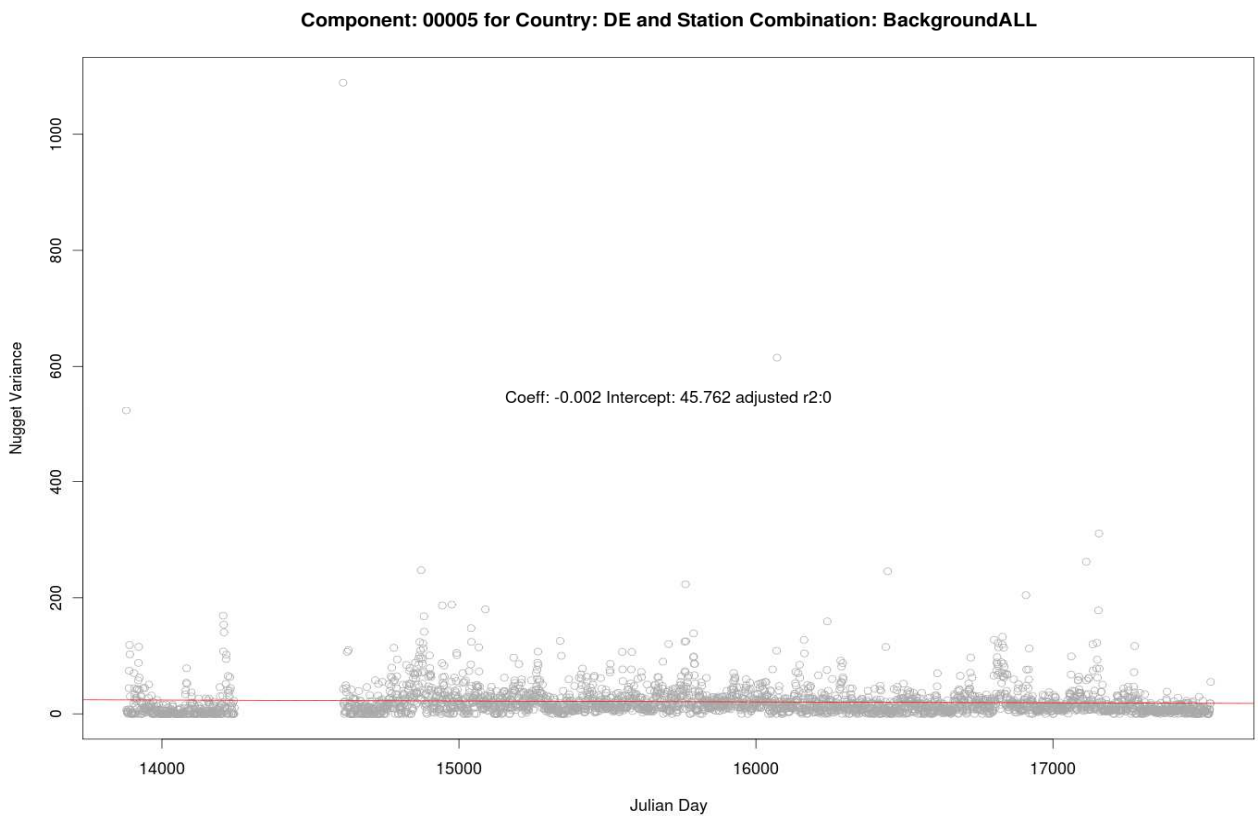

Figure 8: Nugget Semi-variance (gray circles) plotted versus Time (Julian Day) for all German Background Stations over the timeframe 1997-2007. The red line indicates a linear model fit for the plotted data; the parameters shown in the centre of the figure.

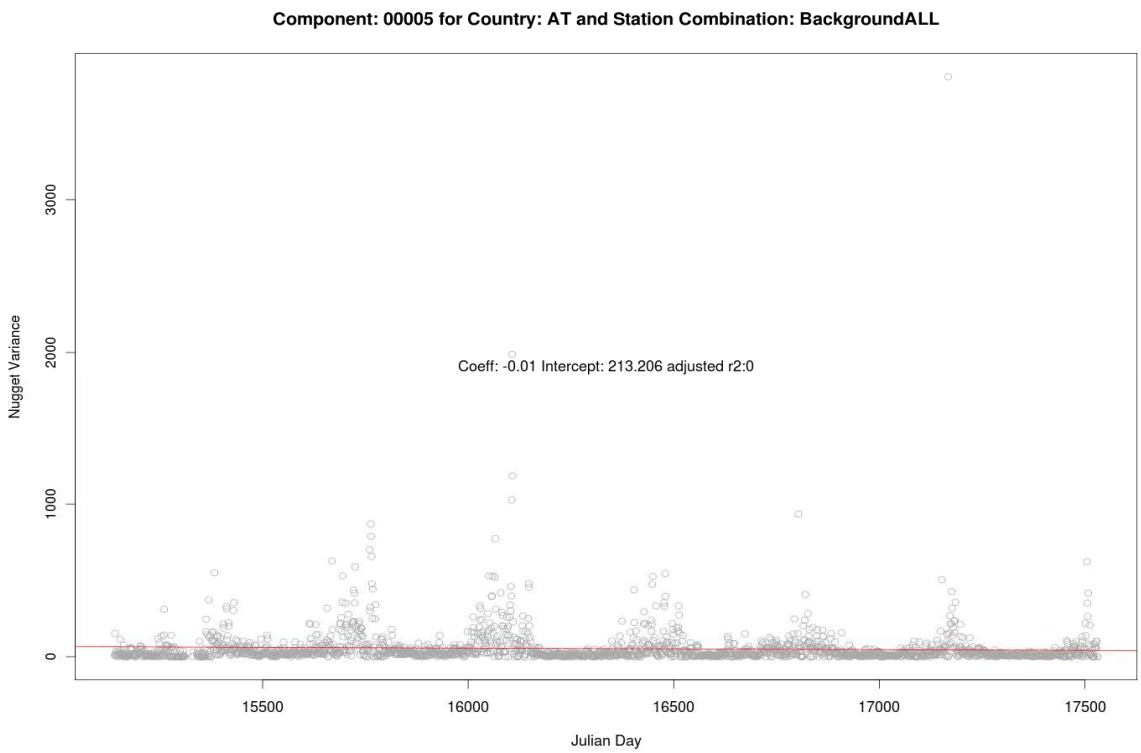

Figure 9: Nugget Semi-variance (gray circles) plotted versus Time (Julian Day) for all Austrian Background Stations over the timeframe 1997-2007. The red line indicates a linear model fit for the plotted data; the parameters shown in the centre of the figure

A more general picture for the different stations types can be found using simple linear regression analysis as shown in the Table 5. In general we observe a decrease in small scale variability across all years. For the "All background stations (line BG-ALL)", a negative coefficient can be observed. Certainly, the values are rather small considering the fact that 10 years of observations are taken into account. While Germany has the smallest decrease in the nugget effect with time (e.g. the least reduction in small scale variability), we could observe Austria and France had similar decreasing values. The largest decrease could be observed for Italy (fourfold over that from Austria).

For the Urban Traffic combination the results are different. We see the strongest decrease in the Austrian dataset, followed by Germany and Italy. Interestingly, France showed an increase in small scale variability with years which need further investigation.

Range effects for "All background stations" (e.g. the length of the spatial dependency) are actually increasing for Germany and slightly increasing for France, while decreasing for Austria and slightly decreasing for Italy. This might indicate that the stations with an increasing range show a more homogeneous picture of the air quality situation surrounding it. In fact, it might be due to an increase of QA/QC actions performed over the years. It could also be the result of a change in the nature or quantity of air pollution emissions/transport or reactions over the year. Another reason might be the

increase/decrease of number of monitoring stations or change in the station classifications. For traffic stations, such statement cannot be made. We observed increase in ranges for Austria, Italy and Germany ans significant decrease for France.

Table 5 Coefficients for fitted linear models for nugget and Range Values for Austria (AT), Germany (DE), France (FR) and Italy (IT) for the Station Type - Station Area Type - Combinations Traffic-Urban(TR-UR), Background-rural(BG-RU), Background-Urban(BG-UR), Background-Suburban(BG-SU), All Background stations (BG-ALL).

		Slope of regression lines over 10 years			
	St - Iso	AT	DE	FR	IT
Slope in % for nugget	TR-UR	-1,8 %	-0,2 %	0,6 %	0,0 %
	BG-RU		-0,8 %		
	BG-UR		-0,6 %	-1,6 %	-1.9 %
	BG-SU		-0,2 %	-1,7 %	
	BG-ALL	-1,0 %	-0,2 %	-1,2 %	-4,3 %
Slope in % for Range	TR-UR	12,2 %	2,8 %	-6,0 %	4,0 %
	BG-RU		2,0%		
	BG-UR		3,3%	-5,9 %	5,8 %
	BG-SU		1,7%	-1,4 %	
	BG-ALL	-2,8 %	2,6%	1,0 %	-1,0 %

All the semi-variograms of Germany between 1997 and 2007 are shown in Figure 10. Along the x-axis the Julian Day is shown, while in y direction the lag distances are plotted. The height of each cross displays the semi-variance for background stations in rural area. Additionally, a surface is plotted inside the figure that fits all points in the semi-variograms. This surface gives a linear model with respect to the time and the distance. For Germany and rural background stations, a clear decreasing trend can be observed for the nugget effect as well as for the semi-variogram range.

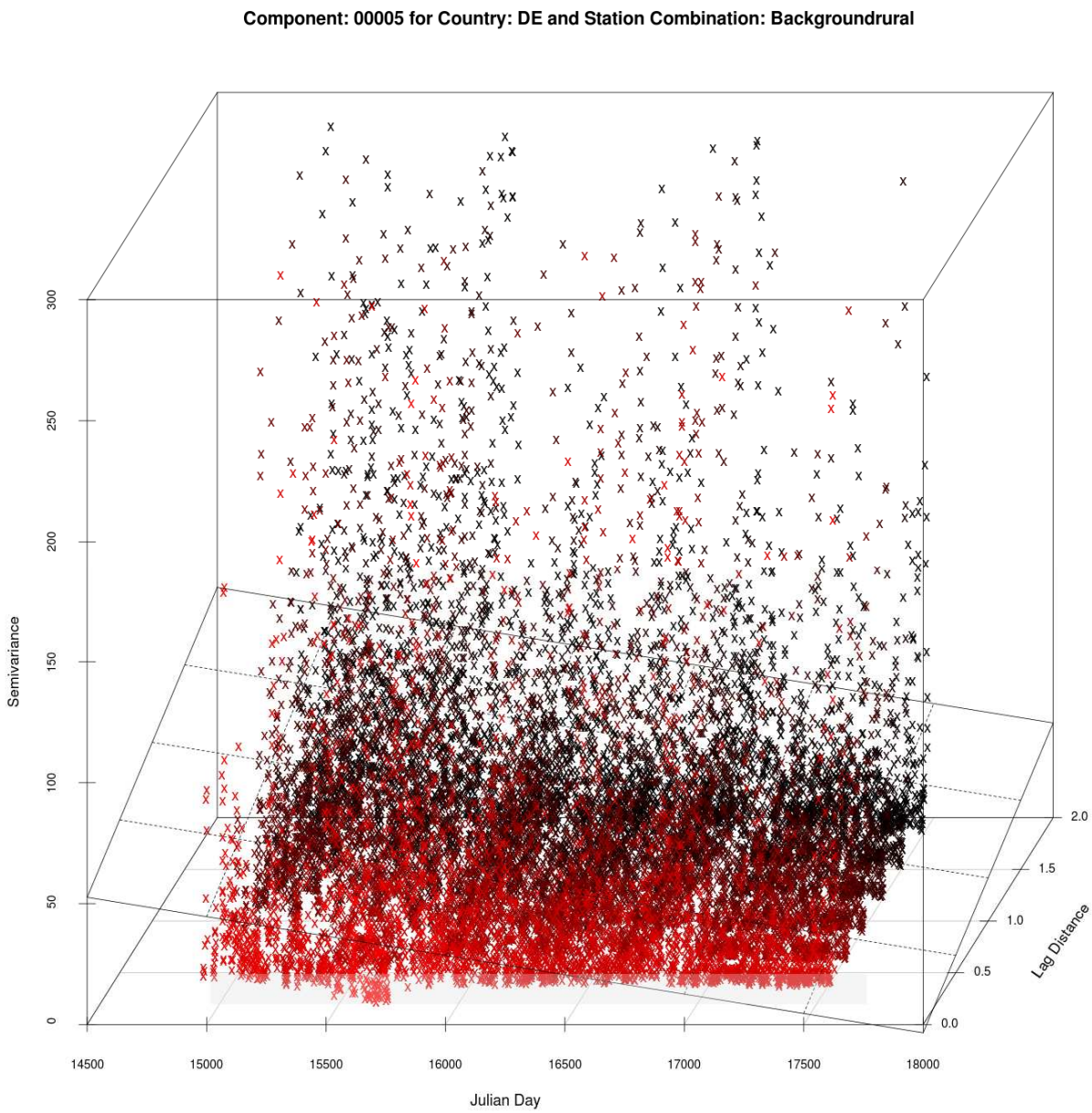

Figure 10: The raw semi-variogram plotted versus Julian day. The x-axis shows the Julian day; the y-axis shows the lag distance; the z-axis the calculated semi-variance. The surface inside the 3D plot represents a fitted model of the semi-variance versus julian day and lag distance; while parameters of these equation are shown at the bottom part of the figure. Black crosses indicates higher values than black crosses.

Create a warning system for classification of monitoring stations

For the identification of environments responsible for population exposure we applied the functions developed in the outlier detection methodology. An example from the results is shown in Table 6. We classified every single measurement that exceeded our conservative threshold of 2. The identified outliers as well as the given percentages are similar for urban and rural areas in Austria. For Germany, quite some significant difference can be observed as almost 2 % of the urban stations measurements

are detected as outliers, similar to the Austrian data. However, Germany's rural background station data show a very low number of outliers.

Table 6 General Statistics of number of records, identified outliers and percentages of outliers identified for four different station type - station area type combinations

Station Type	Number of Records	Identified Outliers	Percentage Outliers
DE Background rural	38480	27	0.07
DE Background urban	63906	1259	1.97
AT Background rural	13339	331	2.48
AT Background urban	13990	352	2.52

Based on these data we divided the stations with respect to their average zi values in 4 classes. For example, for the background data shown in the table above, the four classes are delimited by: low-level stations (bg, with $z < -1$), stations below average (ba, with $-1<z<0$), above average(aa, with $0<z<1$), and high level stations stations(nb with $z>1$). Examples for the four cases are shown in Figure 11 to Figure 14. For the rural background type, stations which are classified as high level stations should be examined further and a reclassification of the station type and of the station area type should be considered if appropriate. It should be stressed that the proposed methodology is a first preliminary assessment, which needs expert validation from the local station managers to see if the assignment needs to be changed. The same is valid for urban background stations which are classified as bg stations – a reclassification as rural background stations might be appropriate. However, more investigations have to be performed to include actual population density data as well as more in depth investigations to quantify the differences in population exposure measurements and the ambient air measurements to come to a sound scientific assessment.

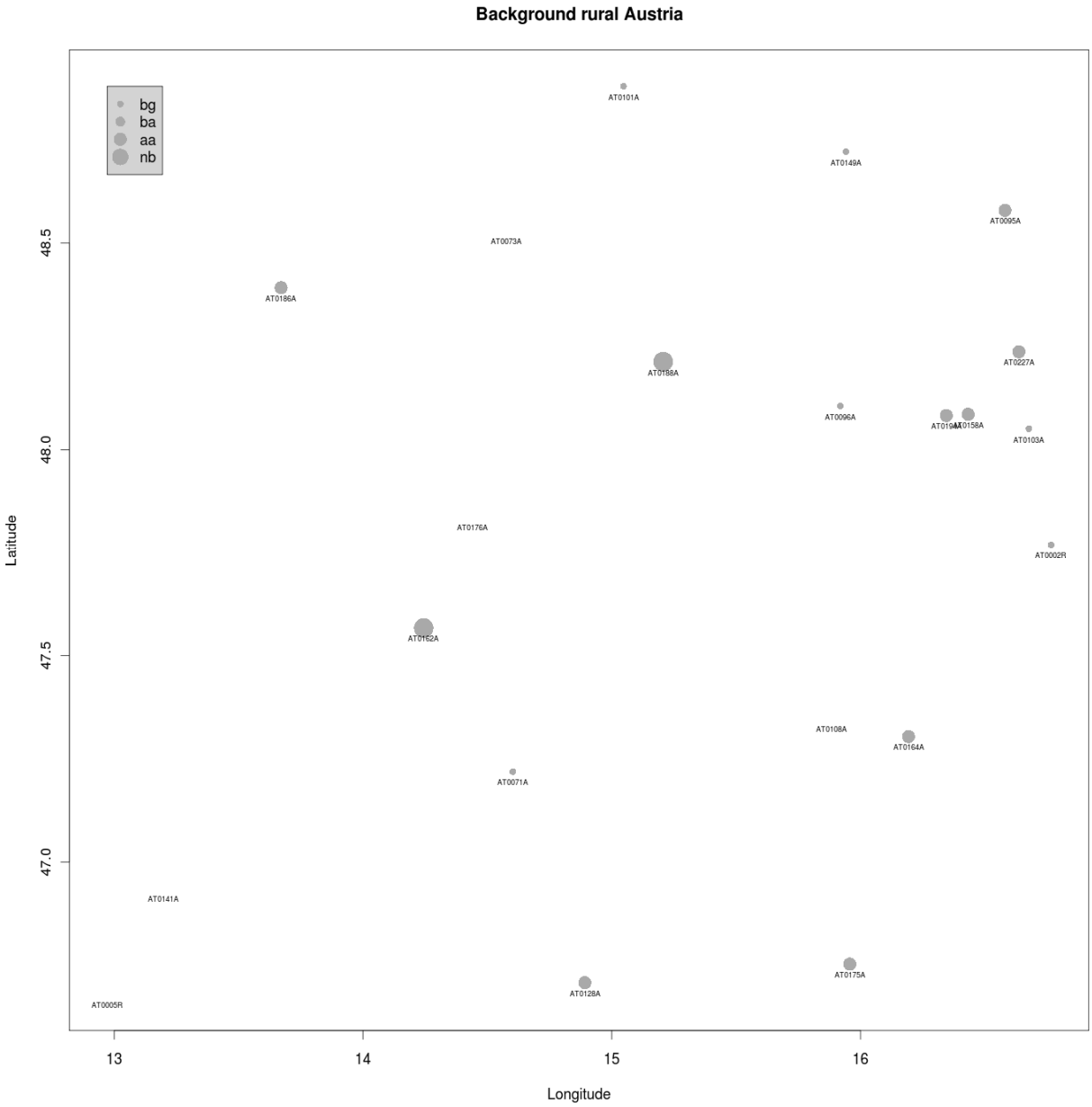

Figure 11: Classification of station for Austria for the rural station type for PM_{10}. Station Labels without points are positions where no classification has been performed

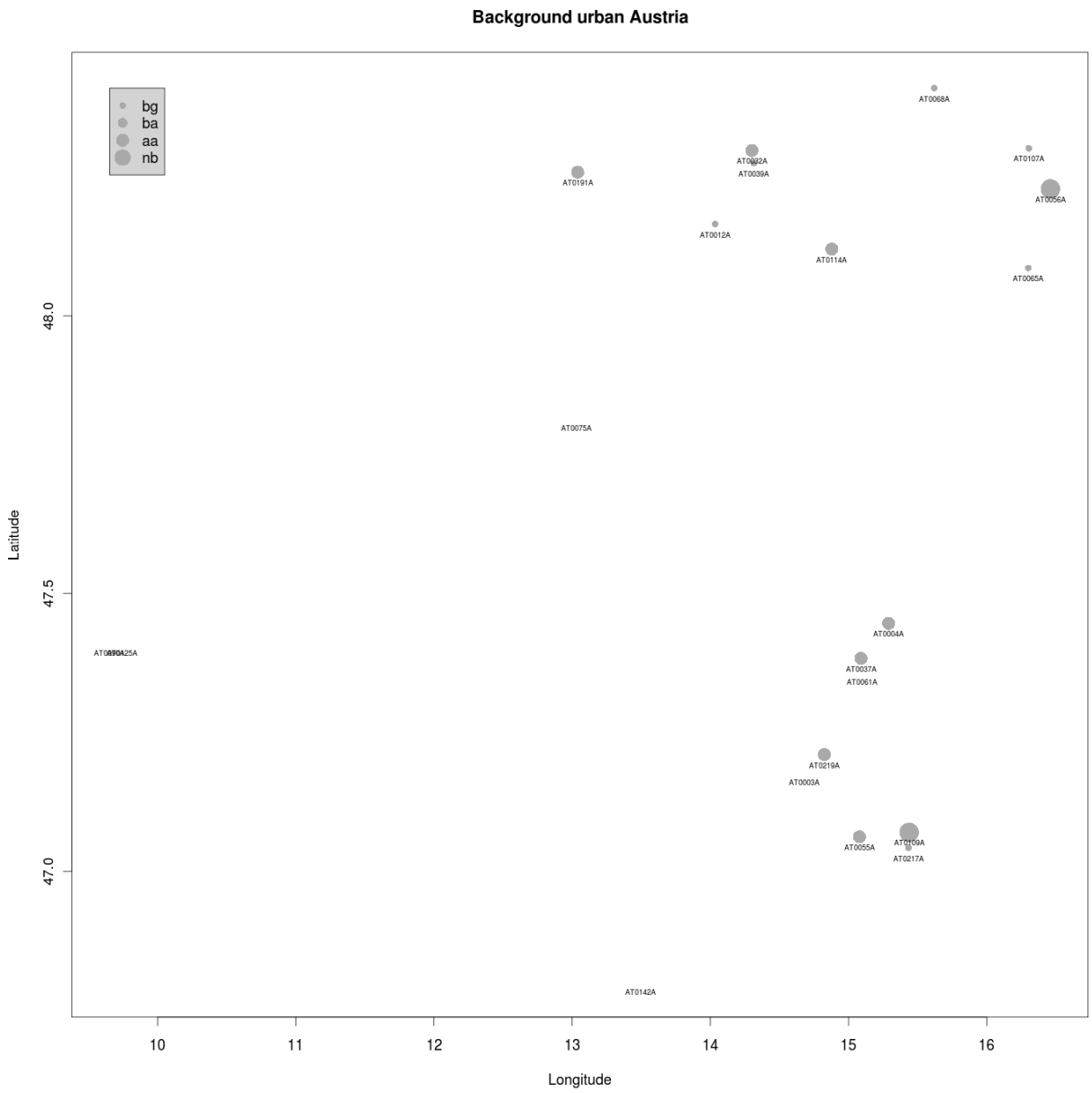

Figure 12: Classification of station for Austria for the urban station type for PM_{10}. Station Labels without points are positions where no classification has been performed.

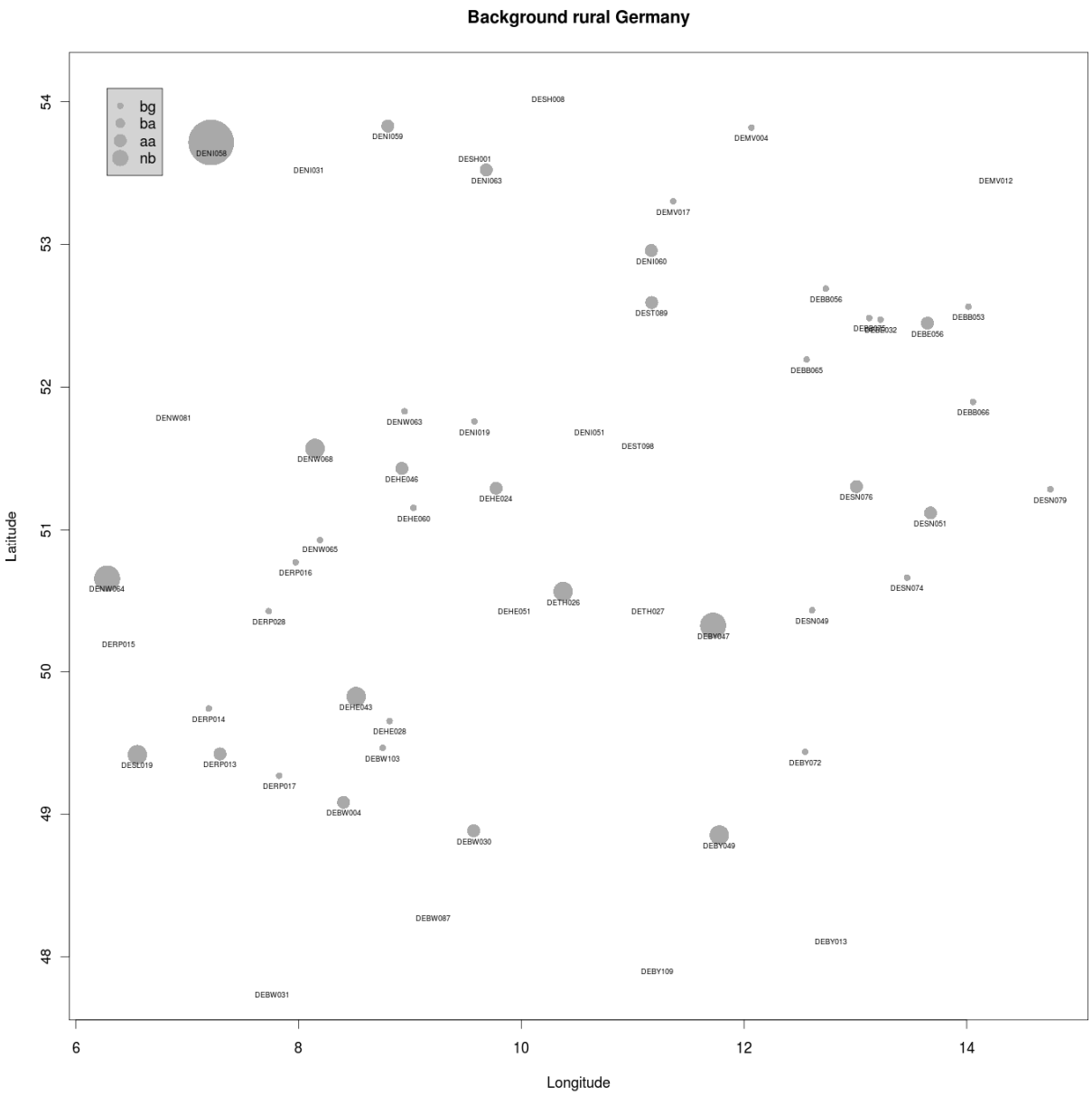

Figure 13: Classification of station for Germany for the rural station type for PM_{10}. Station Labels without points are positions where no classification has been performed

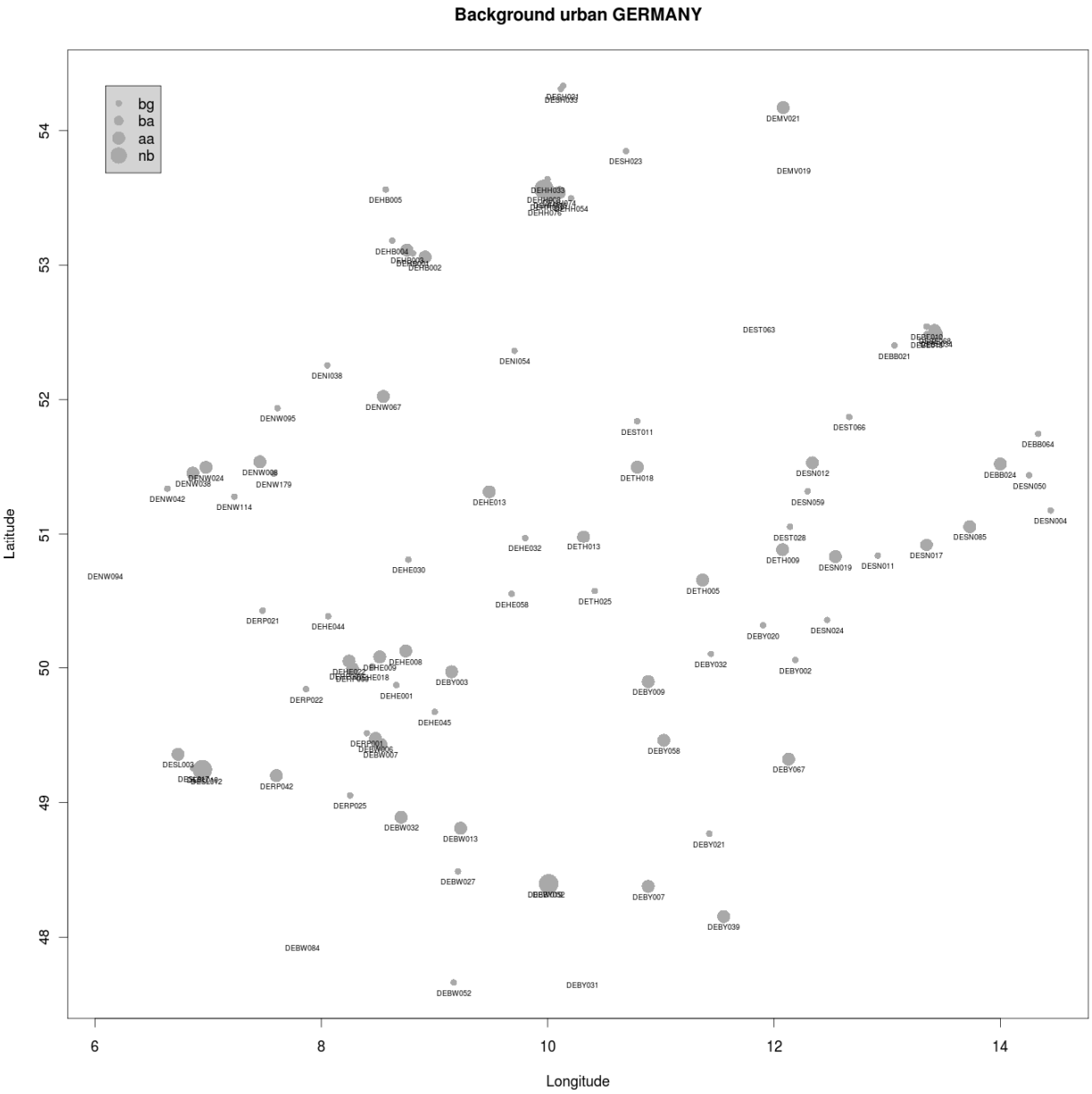

Figure 14: Classification of station for Germany for the urban station type for PM_{10}. Station Labels without points are positions where no classification has been performed

Conclusions

We developed a methodology able to automatically estimate the measurement uncertainty in the air pollution data sets of AirBase. The figures estimated with this method were consistent with expectations from laboratory and field estimation of uncertainty and with the Data Quality Objectives of the European Directives.

The proposed method based on geostatistical analysis is not able to estimate directly the measurement uncertainty. It estimates the nugget effect together with a micro-scale variability that must be minimized by accurate selection of the type of station. Based on the results obtained so far, it is likely that measurement uncertainty is best estimated using all background stations of whatever area type.

So far the methodology has been used to estimate uncertainty in 4 different countries independently. This work should be continued for the whole Europe or for background station without national borders. The method has been shown to be also useful to compare the spatial continuity of air pollution in different countries that seems to be influenced by the topography of each country.

Moreover, it may be used to quantify the trend of measurement uncertainty over long periods like decades with the possibility to evidence improvement in the data quality of AirBase datasets. Over the last 10 years for Austria, Germany, France and Italy a decrease in the nugget effect can be observed, while the change in range (long range spatial dependency) was not significant. Further investigations are needed to determine if this decrease of nugget variance is caused by a decrease of the measurement uncertainty or by long term variations of air pollution or other meteorological factors. We showed that the nugget and range for PM_{10} in 2007 differed significantly between traffic stations while being more or less consistent for all background station types sited in whatever area type. Traffic situations showed up to twice higher nugget effects compared to background station scenarios. Data for different seasons are computed. However more analysis is needed to clarify the results.

Thanks to the implemented outlier detection module, that could also be of interest as a warning system when countries report their measurements to EEA, we have proposed a simple solution to investigate

station classifications in AirBase. We tested the method on the German and Austrian background stations. For several stations, differences in classification could be identified which appeared with respect to the inherent data properties of the selected dataset. However, validation of the outcome of this module has to be performed thoroughly.

The developed method presents a number of shortcomings:

1. The nugget variance overestimates the uncertainty of measurement because of the micro-scale variations and in case of lack of spatial continuity of the pollutant (river, island, mountains ...)
2. The micro-scale variance might be magnified/decreased by the heterogeneity/homogeneity of the sampling sites.
3. The nugget variance cannot detect systematic bias e.g. bias of the measurement methods or chemical interference. This type of systematic bias is unlikely if the selected sufficient sampling sites have different sampling systems, analytical and calibration methods and QA/QC.
4. The nugget variance will depend on the semi-variance of the smallest lag distance of each variogram. When the nugget variance is estimated per country using data sets whose smallest lag are different, one cannot exclude a lack of comparability with the extrapolation of the spherical model on the Y-axis.

Seen the number of shortcomings of the method, validation of the method by comparison to direct approach is needed. For now, this method can be used as a confirmation tool or a ranking tool.

Future study:

Some points of the method need subsequent validation or modification:

- Optimization of the maximum lag distance of the variogram in order to strengthen the estimation of the nugget effect, range and sill. Currently, we preset the maximum extent of the boundaries for the semi-variogram analysis to effectively ensured that the lag boundaries were always within the autocorrelation range. Further research has to investigate how the boundaries

could be fitted automatically also for different area size dataset.

- Optimization and validation of the parameters used for the outliers test (the limit of the neighbourhood ± 1 day and ± 2º and the criteria for the z test: z average over 5 days ± 2). It might be that the test threshold should be different for different components or location.

- Should the semi-variogram be plotted in absolute or in relative values on its y-axis? This is an evaluation of the effect of local mean that may have an effect on the nugget, range and sill. Study whether the uncertainty has a constant value for the whole range of concentration of pollutant (i.e. like in our estimation) or is dependent of the level of concentration (i.e a percentage of the concentration). The latter case is more likely, the variogram should thus be built using the percentage of the concentration of pollutant versus the limit value.

- To diminish the contribution of the micro-scale variability to the nugget effect, explicative variables known on the whole dominium with a high density should be included in multivariable geostatistics like co-Kriging or Kriging with external drift.

- Setup a system to be able to spike air pollution data sets with signal noise (error), quantify the effect on the nugget effect, range and sill in order to validate the whole methodology of uncertainty estimation.

- Validation of the method by comparing its estimation of uncertainty with estimation carried out with laboratory or field experiments. Another solution could be chosen by selecting variograms with pure nugget error to estimate the measurement uncertainty and compare this value with the one only estimated from background stations or against direct estimates of uncertainty with the direct approach.

- Determine which subset of station type and area type to estimate these metrics. The actual hypothesis is that the nugget variance should be estimated using all background stations which lead to a low sill, long range and nugget variance near pure measurement error.

- Look for variables with high density values that are correlated with the concentrations of pollutants (emissions, population density, number of buildings, models outputs ...). By developing variogram of the detrended variables, the influence of the micro-scale variation on the nugget variance might be deleted.

- Optimization of the outlier procedure in terms of computing speed to reach a near-to-real time detection method that might be useful when countries report their data.

The computation of statistics and their evaluation needs be continued:

- Carry out the assessment of nugget variance, range and sill for other pollutants with sufficient monitoring stations (eg. O_3, NO_2…) and for the averaging time of the monitoring for regulatory purposes defined in the European Directives.

- The values of nugget variance should be investigated according to the type calibration chain of standards and other QA/QC and sampling procedures that is implemented by each country or in relation to the implemented inter-comparison exercises to check if these factors may influence the nugget variance.

- The spatial continuity estimated using the range of variograms (the longer the range the more stable the spatial distribution) should be investigated to evidence which compounds are more affected by local emissions, reaction or log-range transport of pollutants.

- Evaluate the trend of nugget variance, sill and range of spatial continuity e.g. over the last 10 years.

- Investigate effect of season. While we have performed a time series analysis to establish how the nugget and range effect changes over a ten year time frame, we already could see from our analysis the influence of seasons. Still the question remains about how the seasonality influences these results in a quantitative way. We should split up the 10 year dataset in steps of 3-4 month each (maybe using a cluster analysis) and analyze them separately. This is important to evidence effects of the station density across different years and for a better understanding of the uncertainty of the different contributing measurement networks of the AirBase Database.

- Estimate the sill, range and nugget variance by selecting monitoring stations belonging to more than one country to detect the presence of possible clusters with borders.

- Map of number of outliers: by performing this in a consistent way across several components, countries might be able to further streamline and improve their station monitoring network. Based on the analysis performed in the classification of sampling sites for the year 2007, we observed that different stations with respect to their station area type or their station type would have to be reclassified. However, what is currently missing is the temporal domain. We urgently need to re evaluate this kind of classification over a range of years to see if a consistent pattern can be detected, otherwise no sound scientific advice can be given to reclassify these stations.

Appendix I: Developed R- Routines

Appendix II: Developed Shell Script for Data import into the PostgresSQL Database

Please contact the Authors

European Commission

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Abstract

We developed a methodology able to automatically estimate of measurement uncertainty in the air pollution data sets of AirBase. The figures produced with this method were consistent with expectations from laboratory and field estimation of uncertainty and with the Data Quality Objectives of European Air Quality Directives. The proposed method based on geostatistical analysis is not able to estimate directly the measurement uncertainty. It estimates the nugget effect from variogram modelling together with a micro-scale variability which must be minimized by accurate selection of the type of station. Based on the results obtained so far, it is likely that measurement uncertainty is best estimated using all background stations of whatever area type. So far the methodology has been used to estimate measurement uncertainty in datasets from 4 different countries independently. This work should be continued for the whole Europe or for background station without national borders. The method has been shown to be also useful to compare the spatial continuity of air pollution in different countries that seems to be influenced by the spatial distribution of the stations (e.g influenced by topography) of each country.

Moreover, the method may be used to quantify the trend of measurement uncertainty over long periods (decades) with the possibility to evidence improvement in the data quality of AirBase datasets.

The implemented outlier detection module would be of interest as the warning system when countries report their measurements to the European Environment Agency. The method could also provides a simple solution to investigate the assignment and accuracy of station classification in AirBase.

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