

1                                   **MAPPING ROADSIDE NITROGEN DIOXIDE**  
2                                   **CONCENTRATIONS USING NON-STATIONARY**  
3                                   **KRIGING**

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5                                   Serge A. Séguret<sup>1</sup>, Laure Malherbe<sup>2\*</sup>, Gilles Perron<sup>3</sup>  
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7    (1) *Centre de Géostatistique, Ecole des Mines de Paris, 35, rue Saint-Honoré, 77300 Fontainebleau,*  
8        *France.*

9    (2) *Institut National de l'Environnement Industriel et des Risques, INERIS, Parc Technologique ALATA,*  
10       *B. P. N° 2, 60550 Verneuil-en-Halatte, France.*

11   (3) *Association pour la Surveillance et l'Etude de la Pollution atmosphérique en Alsace, ASPA, 5, rue de*  
12       *Madrid, 67300 Schiltigheim, France.*

13    \*Corresponding author. Tel.: +33 3 44 55 62 18; fax: + 33 3 44 55 68 99

14    E-mail address: [laure.malherbe@ineris.fr](mailto:laure.malherbe@ineris.fr)  
15

16    **Abstract**

17    Atmospheric nitrogen dioxide (NO<sub>2</sub>) concentrations around a major road in Alsace (France) are estimated  
18    on a fine grid using measurements given by passive samplers and a geostatistical approach. Data are  
19    referenced to a local coordinate system where (x, y) are respectively the distance from and along the road.  
20    They show a strong non-stationarity which does not allow ordinary kriging to be used in the estimation.  
21    Therefore a trend is modelled by a combination of exponential and polynomial functions. Experimental  
22    residuals are then computed as the differences between measurements and the trend. The idea is to  
23    interpolate the residuals at the nodes of the grid, applying kriging methods, and to add them to the trend  
24    estimate. Since their variance is not stationary either, an intermediary step is required. It consists in  
25    modelling the standard deviation of the residuals as a function of the drift and normalizing the residuals  
26    by this model. This defines a new regionalized variable which can be estimated in the framework of  
27    stationary geostatistics. Two possible kriging systems are tested, depending on the fitted variogram  
28    model: in the first one, a pure nugget effect (white noise) is used, in which case the best linear estimator  
29    of NO<sub>2</sub> concentration is the trend model; in the second one, a structured exponential variogram is  
30    adjusted. This case study shows that non-stationarity may not only characterize the raw variable but can

31 also affect the variance of a phenomenon. It illustrates the interest of modelling it so as to improve the  
32 experimental variogram, fit an acceptable variogram model and compute the variance of the estimation  
33 error even if the estimator is reduced to a simple regression function.

34

35 *Keywords:* geostatistics, non-stationarity, trend estimation, standard deviation model, universal kriging

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37

## 38 **1. Introduction**

39

40 Air pollution mapping is a valuable tool for assessing population long-term exposure and informing the  
41 public about the spatial distribution of outdoor concentrations. At the regulatory level, calculation of  
42 concentration maps is prompted by the European daughter directives on ambient air quality assessment  
43 (1999/30/ EC, 2000/69/ EC, 2002/3/ EC).

44 In France geostatistical methods have been receiving particular attention for several years and are now  
45 commonly used by the French air quality monitoring organizations. They are mostly applied for mapping  
46 background concentrations at the city or regional scale so as to fulfill the regulatory recommendations.  
47 Roadside pollution is equally a matter of concern as regards people exposure and has been the subject of  
48 many investigations (Gilbert, 2003, Kodama, 2001, Roorda-Knape, 1999). However, the difficulty in such  
49 cases is that concentration usually shows a strong drift (it decreases with distance from the source), which  
50 makes the classical methods of stationary geostatistics like ordinary kriging or cokriging unsuitable.

51 Deletraz and Dabos (2001) tested different interpolating methods in order to map the environmental  
52 impact (expressed as NO<sub>2</sub> deposition) of a road in a mountainous region and opted for a multiple  
53 regression estimator using the inverse distance from the road and the logarithm of roughness as  
54 explicative variables. The regression model explains 87% of the deposition variability. Unlike the  
55 geostatistical method explored hereafter, it does not take the residual variability into account. Briggs et al.  
56 (2000) also proposed a regression mapping technique to model the spatial patterns of traffic-related  
57 pollution. The developed model is a linear combination of variables derived from traffic counts, land  
58 cover and altitude.

59 Several studies show the relevance of geostatistics to treat non-stationary problems in the vicinity of  
60 chemical or contaminating sources. Figueira et al. (1999) applied kriging with external drift for estimating  
61 soil salinity in a coastal land strip. The logarithm of the distance from the coast was used as an auxiliary  
62 variable. The results were compared with those of ordinary kriging. They attested the efficiency of the

63 external drift. Saito and Goovaerts (2001) applied different techniques based on kriging to estimate lead  
64 soil concentrations around a former smelter in Dallas. In particular they modelled a trend by a linear  
65 combination of  $\log(d)$  and  $\Delta\theta$ ,  $d$  being the distance from the smelter and  $\Delta\theta$  the deviation from the main  
66 wind direction. Then they worked with residuals obtained from the difference between measurements and  
67 the trend estimates.

68

69 In this methodology-oriented study we propose to perform kriging of residuals from a global trend model  
70 in order to estimate  $\text{NO}_2$  roadside concentrations in the Thur Vosges Valley in Alsace, France.

71

72 Concentration data are derived from passive diffusion tube measurements performed 3 meters above the  
73 ground at 39 sites around a major road, (Fig. 1-A). The aim of the measurement campaign, which took  
74 place in summer 2001, was to assess the environmental impact of traffic. Therefore the samplers were  
75 located in places supposed to be strictly under the influence of road emissions. Most tubes were  
76 positioned on transects and situated 1 m, 2 m, 50 m, 200 m, 400 m far from the road. Similar  
77 measurements were also conducted in winter. However, the present article focuses on summer average  
78 concentrations, calculated from 6 fortnightly records. Results for winter are mentioned only if they  
79 present significant differences with summer. The road is 20 km long. To simplify its width has been set  
80 to zero as it is not precisely known.

81

82 Among all the auxiliary variables (land use, emission inventory, population density ...) distance to the  
83 road and elevation appear to be explanatory of  $\text{NO}_2$  concentration, as indicates a Factorial Component  
84 Analysis. Now, the elevation is strongly correlated with distance along the road (curvilinear abscissa)  
85 (see Fig.1-B). Consequently  $\text{NO}_2$  concentrations are expressed in a coordinate system where  $(x, y)$  are  
86 distances from and along the road, respectively (Fig. 1-C). By this way elevation is implicitly taken into  
87 account.

88

89 Data show a clear tendency of  $\text{NO}_2$  concentration to decrease with distance from the road, indicating the  
90 non-stationary nature of the pollution phenomenon. In that context a mapping methodology based on  
91 estimating the trend is suggested.

92 In what follows, concentration measurements are considered as the realization of a random function  
93  $\text{NO}_2(x,y)$ . The estimation involves four stages.

- 94 1. At any point (x,y), NO<sub>2</sub> is written as the sum of a trend m(x,y) providing the general pattern of  
 95 pollution along and across the road, and a random residual:  $NO_2(x,y)=m(x,y)+R(x,y)$   
 96 Therefore this first step consists in adjusting a trend model on the concentration data.
- 97 2. As the variance of the residuals depends on their value (non stationary variance), those residuals have  
 98 to be normalized so that classical kriging methods can be applied. For this purpose their standard  
 99 deviation is modelled by a function  $\sigma(x,y)$ .
- 100 3. The experimental variogram of the normalized residuals  $NR(x,y)=R(x,y)/\sigma(x,y)$  is calculated to  
 101 describe the spatial variability of concentration fluctuations around the trend and a variogram model is  
 102 fitted.
- 103 4. A simple kriging system is solved to estimate NR(x,y) at the nodes of a 250m x 250m mesh grid.  
 104 Finally the estimator of NO<sub>2</sub>(x,y) is built :

$$NO_2^*(x, y) = \sigma(x, y)NR^*(x, y) + m(x, y)$$

105  
 106 After presenting each stage, we conclude by a discussion where :

- 107 - the results are interpreted;  
 108 - other possible kriging systems are considered, depending on the confidence given to the trend  
 109 estimate.

110

## 111 2. Modelling the trend

112

113 As a preamble, let us define the notion of non-stationarity : a regionalized phenomenon, namely a  
 114 physical phenomenon stretching out in space and represented by a function Z, is stationary if its mean  
 115  $m(p)$  and its covariance  $cov[Z(p),Z(p+h)]$  are independent of the location of point p.

116 Fig. 2-A represents the projection of concentrations on a plane perpendicular to the road. Two main  
 117 observations can be made: 1) NO<sub>2</sub> concentration decreases sharply with distance from the road, indicating  
 118 the non-stationary nature of the phenomenon; 2) there is no significant dissymmetry between the left and  
 119 right sides of the road. The first remark involves trend modelling, the second one allows us to handle the  
 120 left and right sides together, increasing the total number of available data in a class and hence, the  
 121 accuracy of the model.

122 A difficult task is now to give a proper analytical form to the trend.

123

124 • For a given  $y = y_j$ , i.e. when concentration variations are examined perpendicularly to the road,  
 125 optimality in the least square sense is obtained in the frame of a bi-exponential function of  $x$  :

$$m(x, y_j) = a(y_j) e^{-\frac{|x|}{s_1}} + b(y_j) e^{-\frac{|x|}{s_2}} \quad [1]$$

126 with  $(s_1, s_2) = (18, 550)$ . For distances from the road greater than 1600 meters the drift can be considered  
 127 as zero. The average concentrations and their interpolated values by function (1) are represented in Fig. 2-  
 128 B.

129 In winter  $(s_1, s_2) = (14, 940)$ . Concentrations are distributed following a different pattern, and become  
 130 zero at more than 3000 meters from the road. Such a difference with summer can be explained by  
 131 climatology, changes in the emissions and photochemical consumption of nitrogen oxide in summer.  
 132 Such observations are consistent with what Monn et al. (1997) already noticed.

133

134 • For a given  $x = x_i$ , i.e. when concentration variations are examined parallel to the road, optimality in  
 135 the least square sense is obtained in the frame of a linear function of  $y$  (Fig. 2-C) :

$$m(x_i, y) = c(x_i) y + d(x_i) \quad [2]$$

136

137 • Combining formulas (1) and (2) leads to the trend model :

$$m(x, y) = c_0 + (c_1 y + c_2) e^{-\frac{|x|}{s_1}} + (c_3 y + c_4) e^{-\frac{|x|}{s_2}} \quad [3]$$

138

139 The constant  $c_0$  is introduced so that the trend is not necessarily zero for distances from the road greater  
 140 than 1600 meters ; the factors  $s_1$  and  $s_2$  are fixed once and for all; the coefficients  $c_0$  to  $c_4$  are obtained by  
 141 a global regression on the 39 measurements. The trend is represented in three dimensions (Fig. 3-A) and  
 142 by isovalues (Fig 3-B).

143 Results are different for each season. For exemple,  $c_0$  is equal to  $0.3 \mu\text{g m}^{-3}$  in summer and  $3.5 \mu\text{g m}^{-3}$  in  
 144 winter.

145

146 Most measurements are located at distances to the road less than 400 meters. Beyond that, the trend  
 147 model is not conditioned by experimental data and only determined by the mathematical properties of the  
 148 exponential functions. To prevent improper extrapolation, the use of  $m(x, y)$  is restricted to  $x$  values below  
 149 500 meters on both sides of the road ( $|x| < 500$  m).

150

### 151 3. Non-stationarity of the variance

152

153 Let  $u$  be the coordinate vector  $(x,y)$ . Using the trend model (3) the residuals  $R(u)$  are calculated at the  
154 measurement points :

$$R(u) = \text{NO}_2(u) - m(u) \quad [4]$$

155 Those residuals are plotted as a function of  $m(u)$  in Fig. 4-A. As expected they fluctuate around 0 but the  
156 amplitude of the fluctuations increases with  $m(u)$ , suggesting that the variance of the residuals is not  
157 stationary. Therefore this variance has to be modelled too. The trend values calculated at the sampling  
158 points are gathered in six different classes and for each class the standard deviation of the residuals is  
159 computed. A power function of  $m(u)$  is then adjusted (Fig. 4-B) :

$$\sigma(u) = d m(u)^p \quad [5]$$

160 The parameters  $d$  and  $p$  are equal to 0.261 and 0.879 respectively ( $d=0.015$  and  $p=1.57$  in winter).

161

### 162 4. Variograms

163

164 To get stationarity the residuals are normalized :

$$\text{NR}(u) = \frac{R(u)}{\sigma(u)} \quad [6]$$

165 Such an operation is possible because  $\sigma(u)$  is strictly positive in the domain under study.

166 Is this normalization (6) really necessary ? The transverse semivariogram (i.e. perpendicular to the road)  
167 of  $R(u)$  is plotted in Fig. 5-A : the large fluctuations are mainly explained by the contrast of variability  
168 between the measurements close to the road and the other concentration measurements. If the non-  
169 stationarity of the variance is ignored and the residuals are not normalized then the variance assigned to  
170 the first set of measurements is too low and the variance assigned to the second one is too high.  
171 Normalization makes it possible to homogenize residuals and is essential to go further in the analysis.

172 When calculated on  $\text{NR}(u)$ , the transverse variogram is significantly improved (Fig. 5-B). The  
173 longitudinal variogram (i.e. along the road, Fig. 5-C) has a cyclic aspect due to the alternation of higher  
174 (about  $50 \mu\text{g m}^{-3}$ ) and lower (about  $40 \mu\text{g m}^{-3}$ ) values for samplers located at distances less than 2 meters  
175 from the road. The housing type may explain such concentration variations but additional information  
176 need to be collected to confirm such an hypothesis. The first point of the longitudinal variogram is  
177 calculated with only ten pairs of data whereas the second one is computed with 118 pairs. Whether or not

178 the first point is taken into account and the transversal or longitudinal variogram is assigned with a  
 179 structure, two approaches are examined :

- 180 - the variogram is considered as not being structured in any of the directions (white noise) : it is  
 181 modelled by a pure nugget effect equal to the variance of NR(u) : C(0)=1.13.
- 182 - a structure is detected and described by the sum of a nugget effect and an exponential model with a  
 183 geometrical anisotropy of ranges 1450 m in the x-direction and 10000 m in the y-direction  
 184 (respectively 80 m and 2200 m in winter).

185

## 186 5. Estimation

187

188 Whatever the approach, let C(h) be the covariance of NR(u). It is linked to the stationary variogram by  
 189 the relationship :

$$190 \quad C(h) = C(0) - \gamma(h)$$

191 The normalized residuals are estimated at the nodes  $u_0 = (x_0, y_0)$  of a grid by a linear combination of the n  
 192 = 39 concentration measurements :

$$NR^*(u_0) = \sum_{\alpha=1}^n \lambda_{\alpha} NR(u_{\alpha}) \quad [7-a]$$

193 The kriging weights  $\lambda_{\alpha}$  are the solution of a simple kriging system :

$$\sum_{\alpha=1}^n \lambda_{\alpha} C_{\alpha\beta} = C_{\beta 0} \quad \forall \beta : 1, n \quad [7-b]$$

194 where  $C_{\alpha\beta}$  (resp.  $C_{\beta 0}$ ) stands for the covariance between measurements taken in  $u_{\alpha}$  and  $u_{\beta}$  (resp.  $u_{\beta}$  and  
 195  $u_0$ ).

196 The estimator of  $NO_2(u_0)$  is obtained setting :

$$NO_2^*(u_0) = \sigma(u_0) NR^*(u_0) + m(u_0) \quad [7-c]$$

197 where  $m(u_0)$  and  $\sigma(u_0)$  are given by (3) and (5).

198 The variance of the estimation error is :

$$\sigma_{NO_2}^2(u_0) = \sigma^2(u_0) (C(0) - \sum_{\alpha=1}^n \lambda_{\alpha} C_{\alpha 0}) \quad [7-d]$$

199 When the covariance C(h) is described by a pure nugget effect, the weights minimizing (7-d) are all zero  
 200 and the estimation is reduced to :

$$\text{NO}_2^*(\mathbf{u}_0) = \mathbf{m}(\mathbf{u}_0) \quad [8-a]$$

201 This result is actually well-known : kriging with a nugget covariance is strictly equivalent to estimating  
202 the trend. Here the trend has been obtained by a mean-square regression. At each node of the grid, the  
203 estimation variance is then :

$$\sigma_{\text{NO}_2}^2(\mathbf{u}_0) = \sigma^2(\mathbf{u}_0) C(0) \quad [8-b]$$

204 As  $C(0)$  is close to 1,  $\sigma_{\text{NO}_2}$  is nearly equal to the non-normalized residues standard deviation model  $\sigma$   
205 given by (5).

206 Fig. 6-A shows the estimation of concentrations using an exponential covariance. Comparing it with the  
207 trend representation (Fig. 3-B) makes it evident how the isolines get distorted to fit the experimental  
208 values. Such a comparison points up the advantage of kriging over a mean-square regression and the  
209 interest of modelling the variogram when possible. The estimation standard deviation for that kriging is  
210 displayed in Fig. 6-B. Compared to the one associated with regression (Fig. 6-C) it is lower around the  
211 data points.

212

## 213 **6. Conclusion and discussion**

214

215 Through the analysis of a case example, a methodology is proposed for elaborating concentration maps in  
216 presence of a spatial trend. Though the limited number of data did not facilitate the interpretation of the  
217 variogram (see section 4), our aim was to point out the interest of modelling non-stationarity when it  
218 affects a variable and its variance. Here we discuss the choices we made for calculating the variogram and  
219 defining a model.

220

### 221 **6.1 Interpretation of the results**

222 The variogram  $\gamma(h)$  is computed with residuals derived from a trend estimate, which necessarily  
223 introduces a bias (Matheron, 1970). As a consequence, the real underlying variogram, assuming it is  
224 structured, can be either masked or deformed, depending on the representativity of the data. Quantifying  
225 the impact of the number of samplers on the quality of the trend estimate is not easy. However the  
226 estimation standard deviation map for a nugget covariance model may be a useful tool for evaluating  
227 kriging results (Fig. 6-C). On the road axis the standard deviation is  $8 \mu\text{g m}^{-3}$  at  $y=5$  km. Then it decreases  
228 sharply till it reaches 1 at  $y = 20$  km and  $x = \pm 500$ m. If the standard deviation is not to be larger than  $3.5$   
229  $\mu\text{g m}^{-3}$  for example, then only the area where this condition is met should be selected for representing

230 concentrations, whereas estimates near the road are ignored (hatched region in Fig. 6-C). This  
231 interpretation of the kriging standard deviation map is strengthened by sensitivity tests consisting in  
232 estimating  $m(x, y)$  with restricted data sets. According to such tests, the trend estimate was sensitive to  
233 data located less than 2 m from the road. For longer distances the coefficients  $c_0$  to  $c_4$  of formula (3) did  
234 not change significantly if 4 or 5 measurements were removed randomly. The poorer quality of estimation  
235 near the road is to be expected since concentrations, and hence the estimation variances, are higher there.  
236 To improve it, more measurements are required.

237

238 The kriging standard deviation map for an exponential covariance model (Fig. 6-B) can be studied in the  
239 same way. Regions respecting the quality criterion, expressed as a maximal value for kriging standard  
240 deviation, can be selected as previously.

241

## 242 **6.2 Using the IRF-k theory ?**

243 A drawback of the proposed methodology is that the final  $\text{NO}_2$  concentration estimate closely depends on  
244 the mean-square trend estimate. To weaken this dependency relationship a more elaborated kriging  
245 system might be used. Actually two different systems could be envisaged :

- 246 - universal kriging of  $\text{NO}_2$  (Appendix 1) : an affine function of the trend  $m(u)$  is reestimated;
- 247 - a more general universal kriging of  $\text{NO}_2$  (Appendix 2): all the coefficients of the trend are  
248 reestimated.

249 Whatever the chosen method, all the systems proceed from the non stationary approach called Universal  
250 Kriging (Matheron, 1969) and do not solve the problem of the bias of the variogram which has been  
251 previously mentioned.

252 The theory of the Intrinsic Random Functions of order  $k$  (IRF- $k$ ) (Matheron, 1971) was developed to  
253 answer that problem. A synthetic description can be found in Chilès and Delfiner (1999). To summarize,  
254 we can say that this theory implies fitting most part of the phenomenon by all possible linear combination  
255 of polynomials having a given degree  $k$ , the variability of the residuals being modelled by a Generalized  
256 Covariance. This method is useful when polynomials can explain the large-scale spatial behaviour of the  
257 phenomenon under study. Is it the case here? Fig. 7 represents the averaged concentrations projected on a  
258 plane perpendicular to the road, and their regression by polynomials. To obtain an acceptable  
259 approximation, we need to use at least all the degrees of  $x$  up to 4. This implies modelling the trend using  
260 a term in  $yx^4$  relevant of an IRF-5. As the set of polynomials must be complete, in order to ensure the

261 regularity of the kriging system, the trend must then be developed on 21 monomials with as much  
 262 multiplicative coefficients to be calculated. This is not reasonable.

263

264 Universal kriging seems therefore to be an effective way of handling the problem of double non-  
 265 stationarity in mean and variance. It is based on hypotheses consistent with the spatial pollution pattern.

266 Besides this methodology does not involve complex mathematical developments and is suitable for  
 267 regular use, in compliance with regulatory requirements.

268

## 269 **Appendix 1 : Universal Kriging of NO<sub>2</sub>**

270 A linear system is built where a linear function of the trend  $m(u)$  is locally estimated by kriging.  $NO_2(u)$   
 271 is consequently expressed as :

$$NO_2(u) = \sigma(u) NR(u) + a_0 m(u) + a_1 \quad [A1-1]$$

272 where  $a_0$  and  $a_1$  are unknown and  $m(u)$  is the trend previously obtained by least square regression. The  
 273 specificity of the present application is a non stationary covariance :

$$Cov(NO_2(u), NO_2(v)) = \sigma(u) \sigma(v) C(v-u) \quad [A1-2]$$

274

275 At any node  $u_0$  of the estimation grid, the estimator is a linear combination of measurements taken at the  
 276 points  $u_\alpha$  :

$$NO_2^*(u_0) = \sum_{\alpha=1}^n \lambda_\alpha NO_2(u_\alpha) \quad [A1-3]$$

277 The weights  $\lambda_\alpha$  are solutions of the following system :

$$\sum_{\alpha=1}^n \lambda_\alpha \sigma(u_\alpha) \sigma(u_\beta) C_{\alpha\beta} + \mu_0 + \mu_1 m(u_\beta) = \sigma(u_\beta) \sigma(u_0) C_{\beta 0} \quad \forall \beta : 1, n \quad [A1-4]$$

$$\sum_{\alpha=1}^n \lambda_\alpha = 1 \quad [A1-5]$$

$$\sum_{\alpha=1}^n \lambda_\alpha m(u_\alpha) = m(u_0) \quad [A1-6]$$

278 The estimation error is defined as the difference  $NO_2^*(u_0) - NO_2(u_0)$ . Its variance is given by :

$$\sigma_{NO_2}^2(u_0) = \sigma^2(u_0) C(0) - \sum_{\alpha=1}^n \lambda_\alpha \sigma(u_\alpha) \sigma(u_0) C_{\alpha 0} - \mu_0 - \mu_1 m(u_0) \quad [A1-7]$$

279 Coefficients  $\mu_0$  et  $\mu_1$  are Lagrange parameters imposed by the universality conditions (A1-5) and (A1-6).  
 280 Equations (A1-4) to (A1-6) make a classical Universal Kriging system which has the distinctive feature of  
 281 using a non-stationary covariance  $\sigma(u)\sigma(u+h)C(h)$ . This system is reversible and has a unique solution  
 282 since the covariance  $\sigma(u)\sigma(u+h)C(h)$  is positive definite like  $C(h)$ . The data are  $\text{NO}_2$  measurements (and  
 283 not the normalized residuals NR). It must be useful to use a moving neighborhood with at least 10  
 284 measurements around the node  $u_0$  where the estimator is calculated.

285

## 286 **Appendix 2 : more general Universal Kriging of $\text{NO}_2$**

287 A linear system is built where the coefficients  $c_0$  to  $c_4$  of the trend  $m(u)$  are locally estimated by kriging.

288 If we put :

$$289 \{X^l, l=1 \text{ to } 5\} = \{1, e^{-\frac{|x|}{s_1}}, e^{-\frac{|x|}{s_2}}, y e^{-\frac{|x|}{s_1}}, y e^{-\frac{|x|}{s_2}}\}$$

290 the kriging system becomes :

$$\sum_{\alpha=1}^n \lambda_{\alpha} \sigma(u_{\alpha}) \sigma(u_{\beta}) C_{\alpha\beta} + \sum_{l=1}^5 \mu_l X^l(u_{\beta}) = \sigma(u_{\beta}) \sigma(u_0) C_{\beta 0} \quad \forall \beta : 1, n \quad [\text{A2-1}]$$

$$\sum_{\alpha=1}^n \lambda_{\alpha} X^l(u_{\alpha}) = X^l(u_0) \quad \forall l : 1, 5 \quad [\text{A2-2}]$$

291 and the variance of the error is :

$$\sigma_{\text{NO}_2}^2(u_0) = \sigma^2(u_0) C(0) - \sum_{\alpha=1}^n \lambda_{\alpha} \sigma(u_{\alpha}) \sigma(u_0) C_{\alpha 0} - \sum_{l=1}^5 \mu_l X^l(u_0) \quad [\text{A2-2}]$$

292 Those expressions use five Lagrange multipliers  $\mu_l$ . As previously, it is advised to use a moving  
 293 neighborhood. The selected data must not lie on a line.

294

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303

304 **References**

305 Briggs, D.J., Hoogh (de), C., Gulliver, J., Wills, J., Elliott, P., Kingham, S., Smallbone, K., 2000. A  
306 regression-based method for mapping traffic-related air pollution: application and testing in four  
307 contrasting urban environments. *The Science of the Total Environment*, 253, 151-167.

308

309 Chilès, J.-P., Delfiner, P. 1999. *Geostatistics. Modeling Spatial Uncertainty*. Wiley, New York.

310

311 Deletraz, G., Dabos, P. 2001. Modélisation statistique de la pollution azotée à proximité d'un axe routier  
312 et évaluation des incidences sur l'environnement. Colloque Risques – octobre 2001, Besançon.

313

314 Council Directive 1999/30/EC of 22 April 1999 relating to limit values for sulphur dioxide, nitrogen  
315 dioxide and oxides of nitrogen, particulate matter and lead in ambient air.

316

317 Directive 2000/69/EC of the European Parliament and of the Council of 16 November 2000 relating to  
318 limit values for benzene and carbon monoxide in ambient air.

319

320 Directive 2002/3/EC of the European Parliament and of the Council of 12 February 2002 relating to  
321 ozone in ambient air.

322

323 Figueira, R., Sousa, A.J., Pacheco, A.M.G., Catarino, F. 1999. Saline variability at ground level after  
324 kriging data from *Ramalina* app. *Biomonitoring*. *The Science of the Total Environment*, 232, 3-11.

325

326 Gilbert, N.L., Woodhouse, S., Stieb, D.M., Brook, J.R., 2003. Ambient nitrogen dioxide and distance  
327 from a major highway. *The Science of the Total Environment*, 312, 43-46.

328

329 Kodama, Y., Arashidani, K., Tokui, N., Kawamoto, T., Matsuno, K., Kunugita, N., Minakawa, N., 2002.  
330 Environmental NO<sub>2</sub> concentration and exposure in daily life along main roads in Tokyo. *Environmental*  
331 *Research, Section A*, 89, 236-244.

332

333 Matheron, G., 1969. *Le Krigeage Universel*. Cahiers du Centre de Morphologie Mathématique de  
334 Fontainebleau, Fascicule 1, Ecole des Mines de Paris.

335

336 Matheron, G. 1970. La Théorie des Variables Régionalisées et ses Applications. Cahiers du Centre de  
337 Morphologie Mathématique de Fontainebleau, Fasc. 5, Ecole des Mines de Paris. Translation (1971) :  
338 The Theory of Regionalized Variables and Its applications.  
339  
340 Matheron, G. 1971. La Théorie des Fonctions Aléatoires Intrinsèques Généralisées. Note Géostatistique  
341 No 117. Technical Report N-252, Centre de Géostatistique, Fontainebleau, France.  
342  
343 Monn, Ch., Carabias, V., Junker, M., Waeber, R., Karrer, M., Wanner, H.U. 1997. Small-scale spatial  
344 variability of particulate matter <math><10 \mu\text{m}</math> ( $\text{PM}_{10}$ ) and nitrogen dioxide. Atmospheric Environment, 31 (15),  
345 2243-2247.  
346  
347 Roorda-Knape, M., Janssen, N.A.H., Hartog (de), J., Van Vliet, P.H.N., Harssema, H., Brunekreef, B.,  
348 1999. Traffic related air pollution in city districts near motorways, The Science of the Total Environment,  
349 235, 339-341.  
350  
351 Saito, H., Goovaerts, P., 2001. Accounting for source location and transport direction into geostatistical  
352 prediction of contaminants. Environmental Science and Technology, 35, 4823-4829.  
353  
354 Séguret, S.A., 2003. Estimation du dioxyde d'azote routier dans la vallée de la Thur. Technical Report N-  
355 1/03/G available on pdf file at <http://www.lcsqa.org>.

356 **Figure captions**

357

358 Fig. 1 : (A) Road representation in the original geographical system. The red points stand for the 39  
359 measurement points, 33 of which lie on transepts T1 to T6.

360 (B) Correlation cloud between altitude and measurement distance along the road (curvilinear abscissa).

361 (C) Location of measurements in the local reference system.

362

363

364 Fig. 2 : (A) Projection of the measurement data onto a plane perpendicular to the road (only data located  
365 on transepts have been projected). Black points represent the average concentrations for different distance  
366 classes ( $|x|=1\text{m}, 2\text{m}, 50\text{m}, 200\text{m}$  and  $400\text{m}$  apart from the road).

367 (B) Average concentrations for the considered distance classes and adjustment of a combination of two  
368 exponential functions.

369 (C) Concentration variations along the road (i.e. along  $y$ ) for each  $|x|$ -distance class and their  
370 approximation by linear functions.

371

372

373 Fig. 3 : (A, B) Graphic representation of the trend  $m(x,y)$  in perspective (A) and in isolines (B). The red  
374 and black isolines are  $10 \mu\text{g m}^{-3}$  and  $2 \mu\text{g m}^{-3}$  distant from each other, respectively.

375

376

377 Fig. 4 : (A) Residuals  $R(u) = \text{NO}_2(u) - m(u)$  as a function of the trend  $m(u)$ .  $u = (x,y)$  is the coordinate  
378 vector in the local reference frame.  $R(u)$  fluctuations around 0 grow with  $m(u)$ . (B)  $R(u)$  standard  
379 deviations for each  $m(u)$  value class and adjustment of a model written as  $d m(u)^p$ , with  $d=0.261$  and  
380  $p=0.879$ .

381

382

383 Fig. 5 : (A) Experimental variogram of the residuals  $R(u)$  in the transverse direction before  
384 standardization. (B) Experimental variogram of the standardized residuals  $R(u)$  in the transverse  
385 direction. The number of pairs participating to the calculation of each point are indicated on the graph.  
386 Two types of models have been fitted: a pure nugget model (in blue) and a nugget+exponential

387 anisotropic model with a range of 1.45 km along x and 10 km along y. (C) Variogram of the standardized  
388 residuals along the road.

389

390

391 Fig. 6 : (A) Nitrogen dioxide concentration obtained by simple kriging of the standardized residuals,  
392 using a structured variogram model. Size of the data points is proportional to the measurement values. (B)  
393 Kriging standard deviation for the above calculated map. (C) Kriging standard deviation for a pure nugget  
394 variogram model. It is almost equal to  $\sigma(x,y)$  and can be regarded as the error standard deviation for the  
395 trend estimation. In the red hatched area the standard deviation is greater than 3.5.

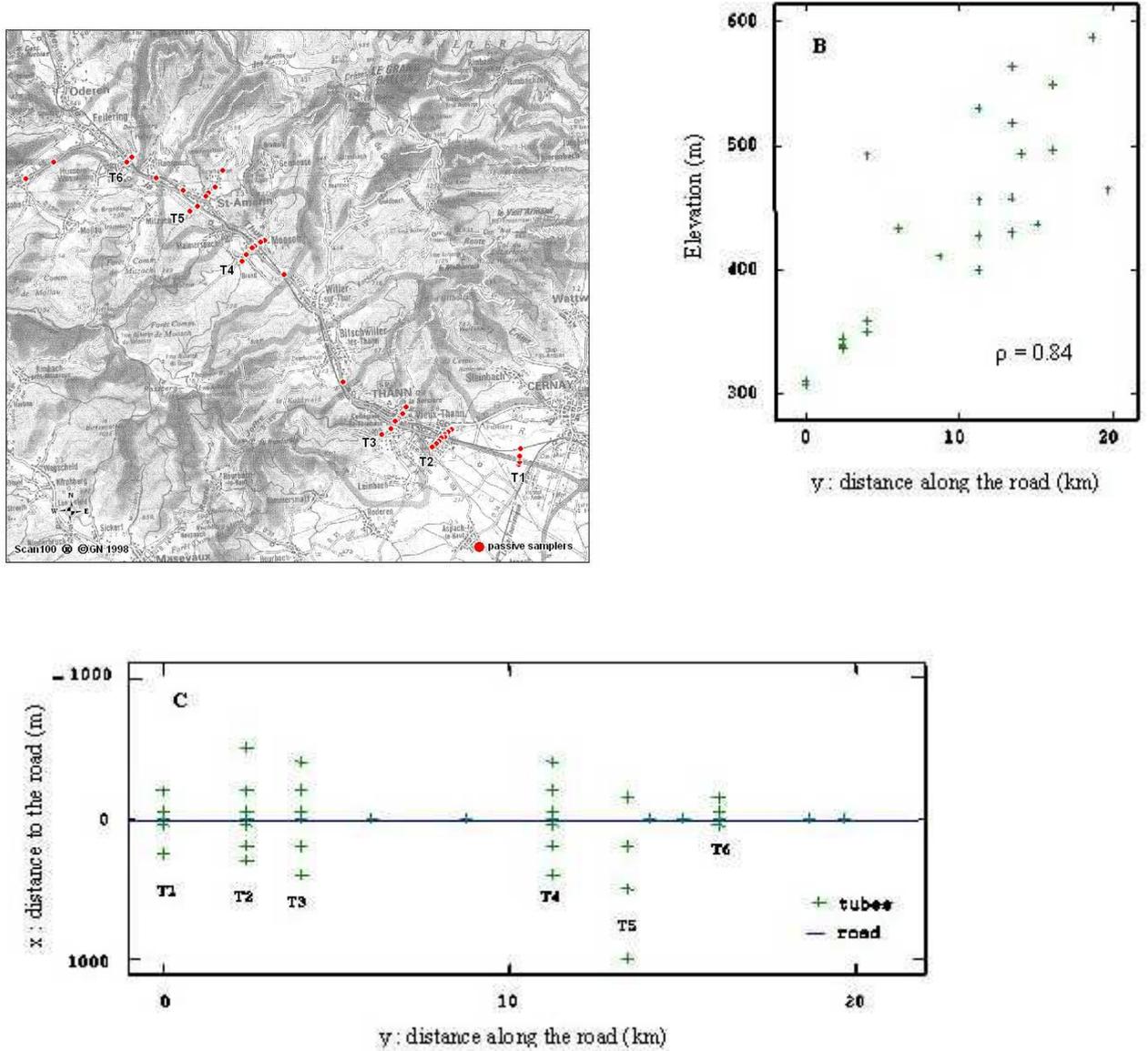
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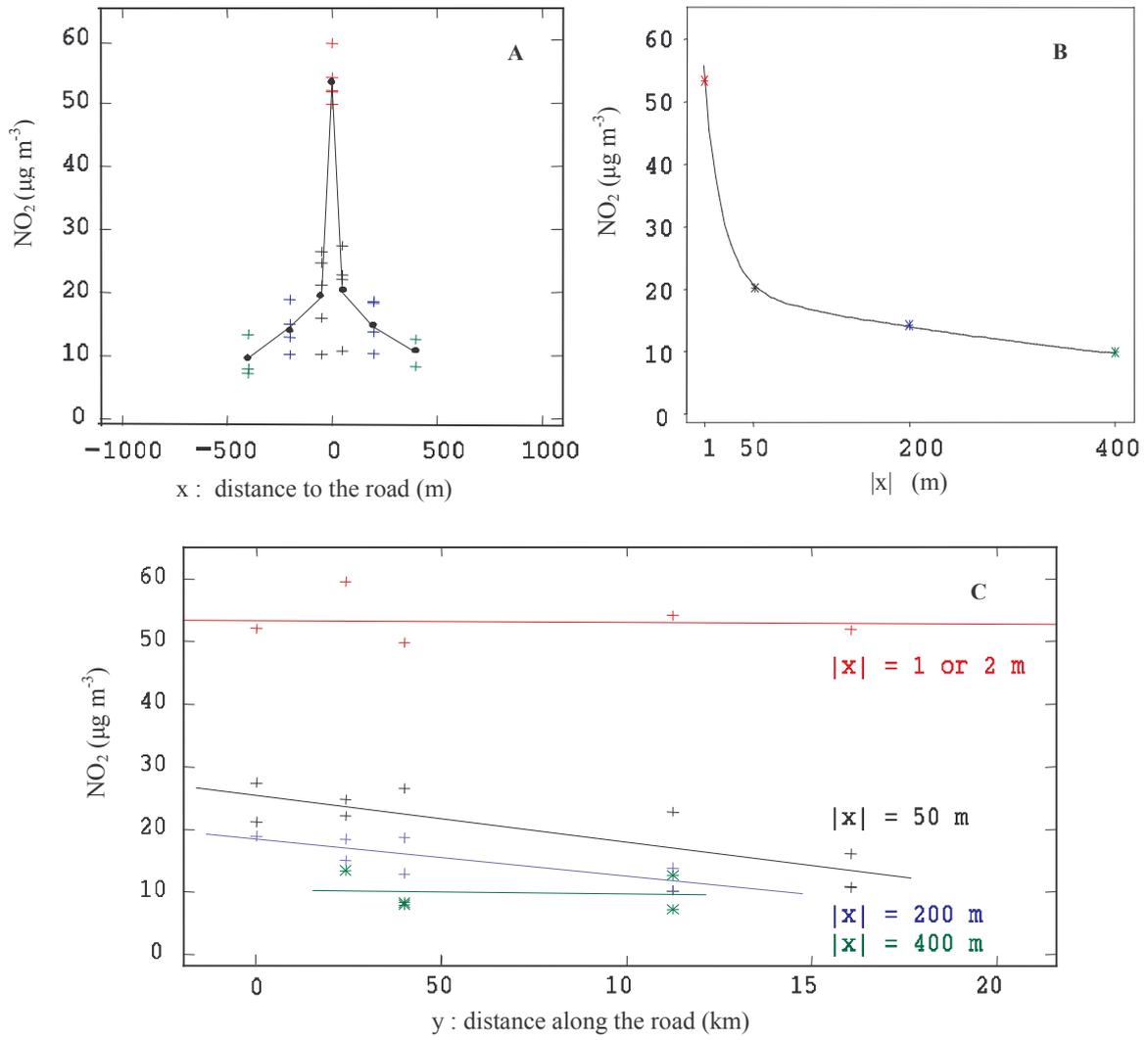
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398 Fig. 7 : NO<sub>2</sub> concentrations across the road (green dashed curve) and their approximation by 2<sup>nd</sup> to 4<sup>th</sup>  
399 order polynomial functions (solid curves).

400

A

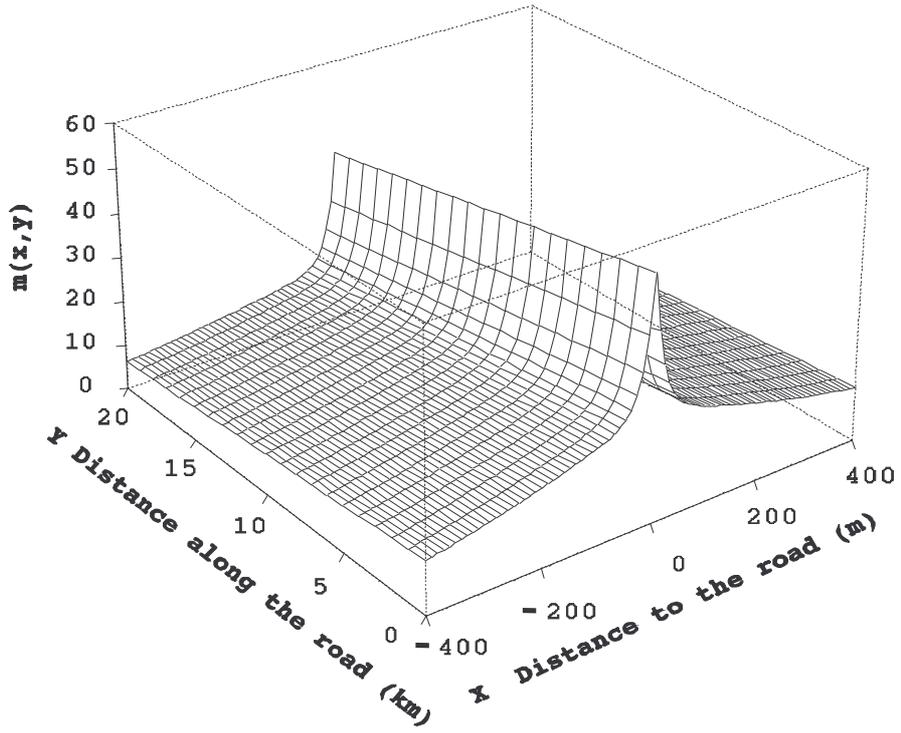




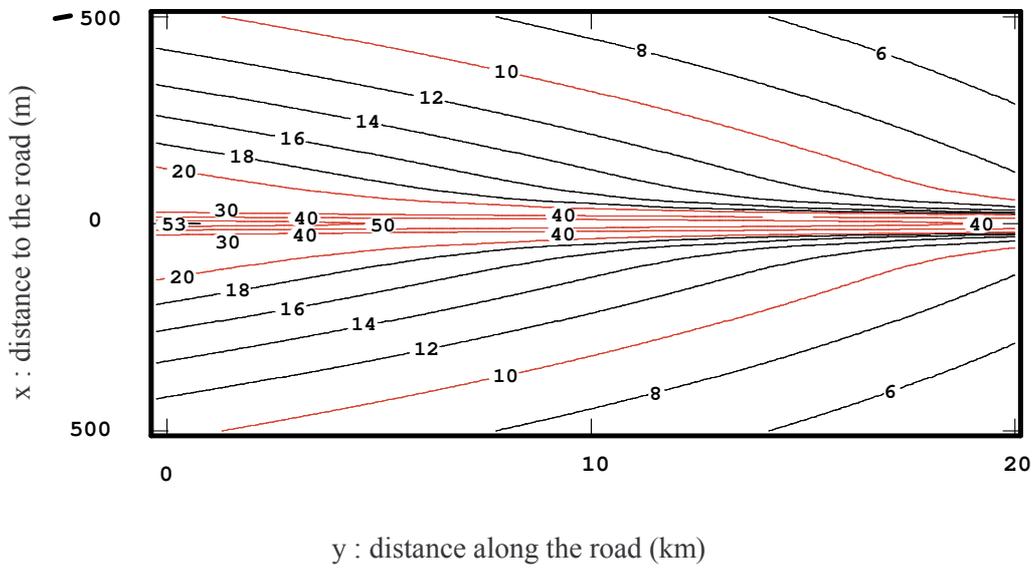
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409 Fig. 2

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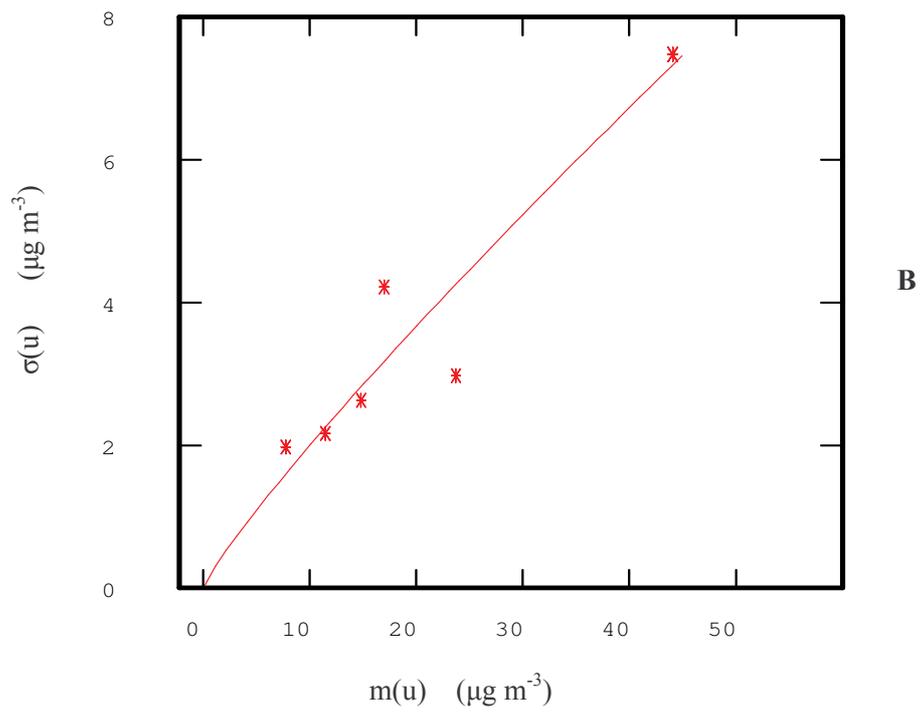
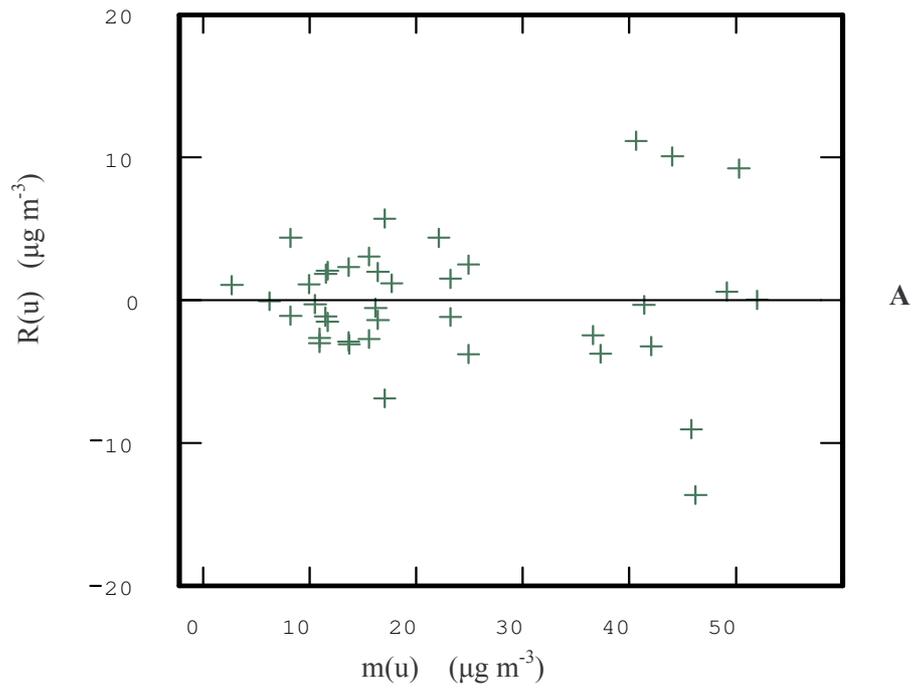
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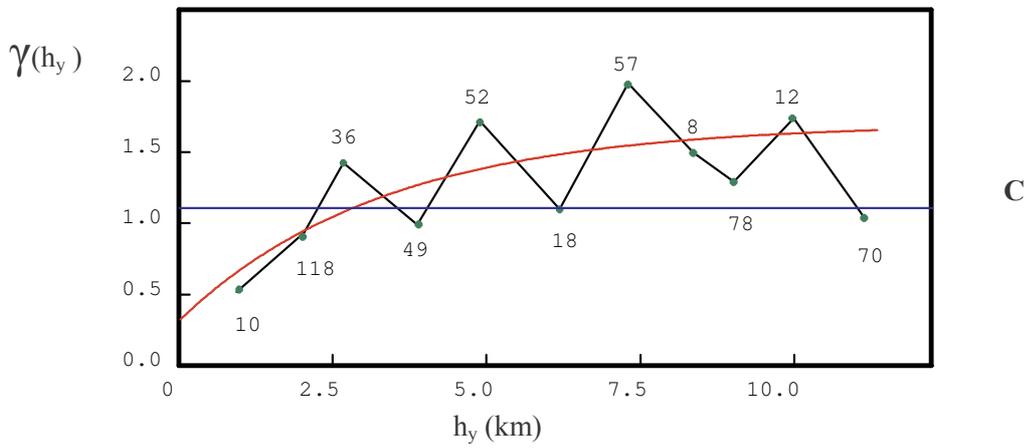
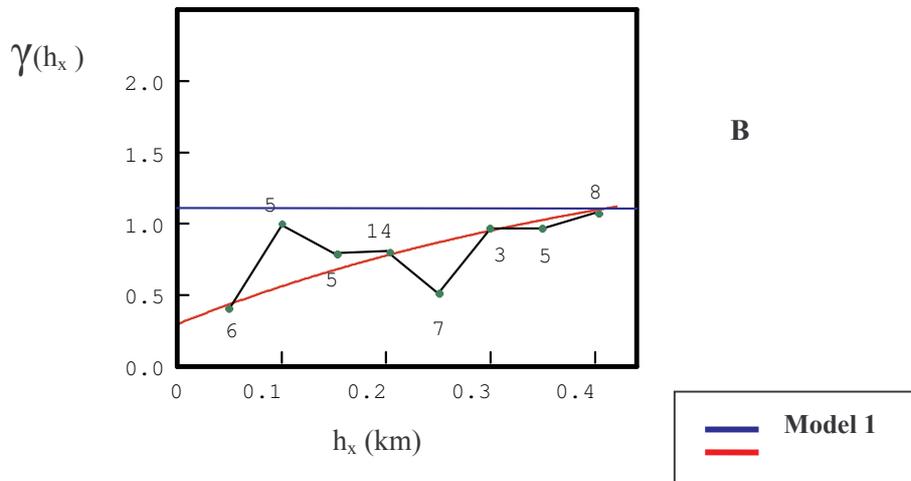
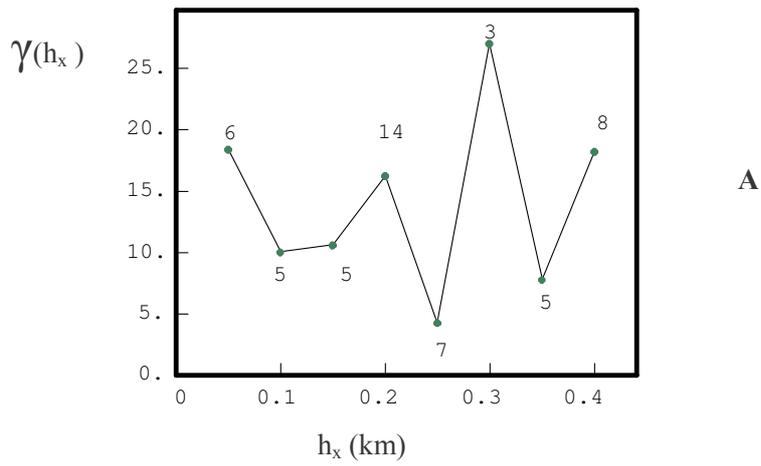
411 Fig. 3

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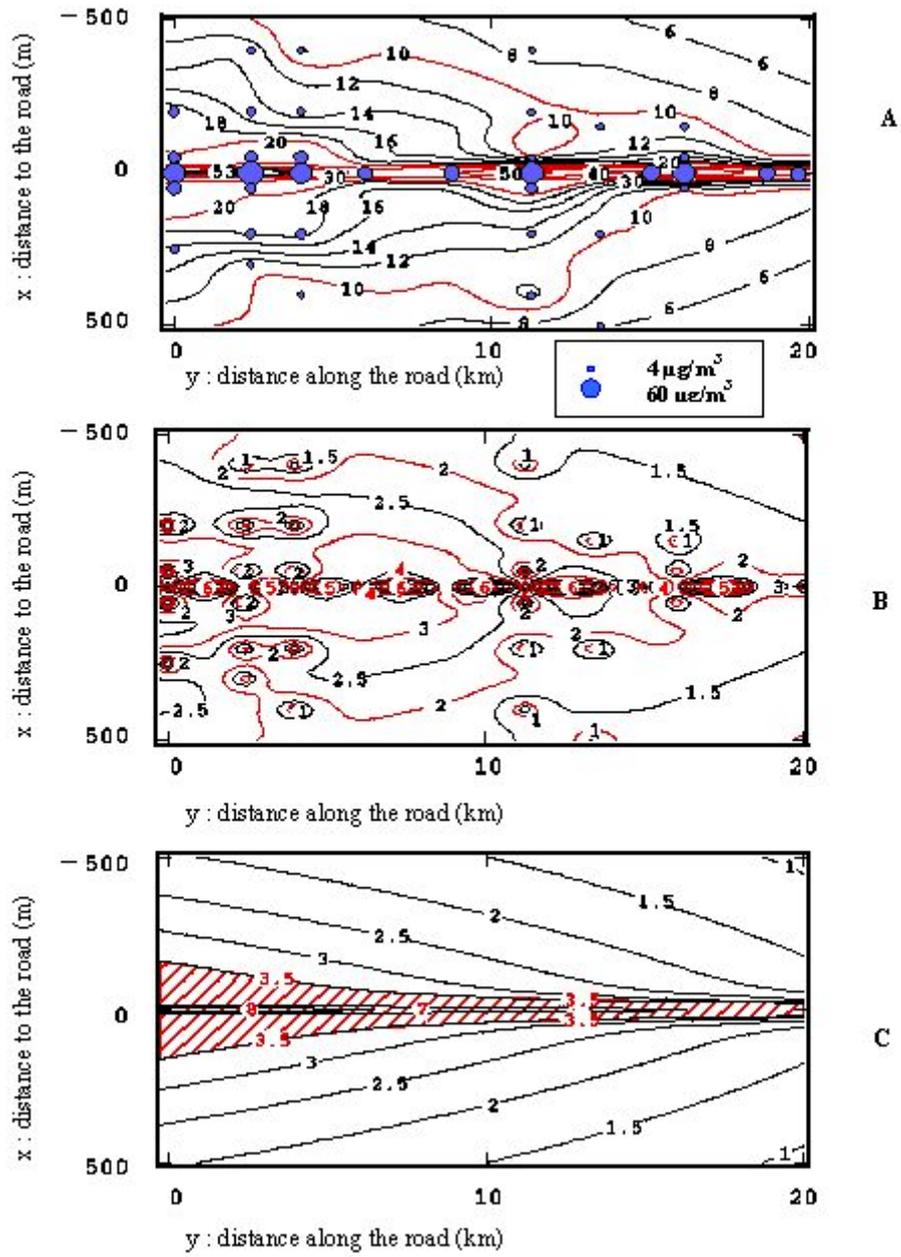
413 Fig. 4

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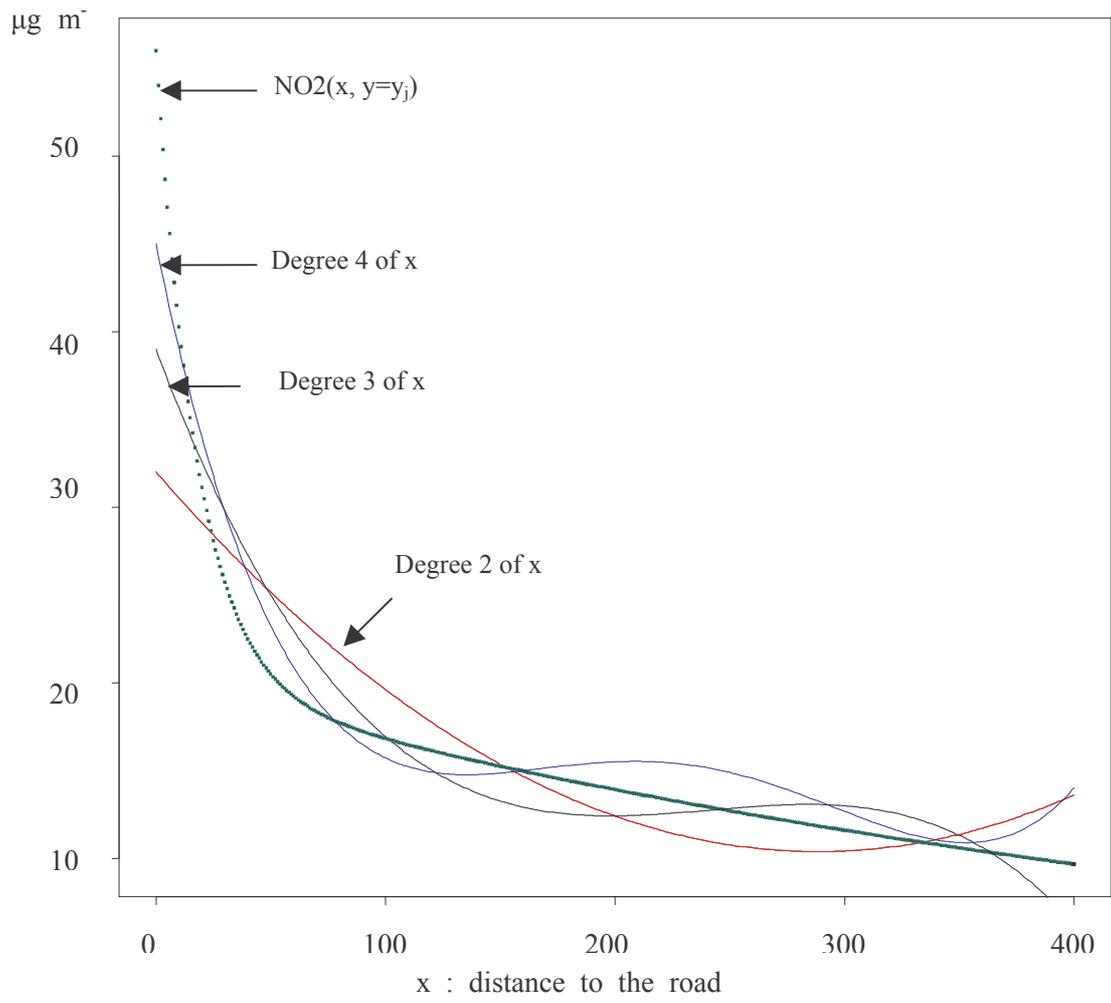
415 Fig. 5

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417 Fig. 6

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419 Fig. 7

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