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# Hydrogeology Journal

## Making kriging consistent with flow equations: application of Kriging with Numerical Covariances for estimating a contamination plume --Manuscript Draft--

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<b>Abstract:</b>	<p>When the data are few, kriging hydraulic head or concentration with usual variogram models can lead to physically inconsistent results, because the non-stationarity induced by the flow or transport equations is not taken into account properly. Several methods have been proposed to account for these equations in the geostatistical estimation. A recent and general approach consists in incorporating them through specific covariance models. A set of random fields sampling uncertain parameters (e.g. conductivity) is first used as input of a flow simulator. Empirical "numerical" spatial covariances are then calculated between pairs of points and for the variable of interest (e.g. hydraulic head, concentration) on the corresponding set of flow simulator outputs. These non-stationary "numerical" covariances are consistent with the specific spatial variability of hydraulic head or concentrations. They are used in the estimation. In this paper, flow-and-transport simulations are thus combined with kriging to estimate contaminant concentrations in groundwater. A non-stationary Gaussian anamorphosis is introduced for non-linear estimation so that the estimate of the concentration is positive.</p> <p>The method is first validated on synthetic data and then on real data from a two-dimensional cross-section of an aquifer downstream of a trench containing radioactive waste in the Chernobyl area. Kriging with the output of a simplified flow model as external drift and kriging with numerical covariances reproduce the spatial variability of the contaminant plume much better than usual (ordinary) kriging based on observations only. The comparison between the two best estimators is discussed</p>	
<b>Corresponding Author:</b>	Chantal de FOUQUET Mines Paris - PSL Fontainebleau, FRANCE	
<b>Corresponding Author Secondary Information:</b>		
<b>Corresponding Author's Institution:</b>	Mines Paris - PSL	
<b>Corresponding Author's Secondary Institution:</b>		
<b>First Author:</b>	Chantal de FOUQUET	
<b>First Author Secondary Information:</b>		
<b>Order of Authors:</b>	Chantal de FOUQUET	
	Mathieu Le Coz	
	Xavier Freulon	
	Léa Pannecoucke	
<b>Order of Authors Secondary Information:</b>		
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1 [Making kriging consistent with flow equations: application of kriging with numerical](#)  
2 [covariances for estimating a contamination plume](#)

3

4 Chantal de Fouquet<sup>1\*</sup>, Mathieu Le Coz<sup>2</sup>, Xavier Freulon<sup>1</sup>, Léa Pannecoucke<sup>1,2</sup>

5 1. Mines Paris, PSL University, Centre for geosciences and geoengineering, 77300  
6 Fontainebleau, France

7 2. Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-ENV/SEDRE/LELI, 92260  
8 Fontenay-aux-Roses, France

9 \* Corresponding author: [chantal.de\\_fouquet@minesparis.psl.eu](mailto:chantal.de_fouquet@minesparis.psl.eu)

10

11 **ABSTRACT**

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31

32

33 **Keywords:** Groundwater modelling, Geostatistics, Radionuclide migration, Contamination

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## 39 **1. Introduction**

40 Estimating the hydraulic head or concentrations of components in groundwater is necessary for  
41 drawing a map, locating "at best" new measurements by minimalizing redundancy (Júnez-Ferreira et  
42 al., 2013), or solving the inverse problem (Gomez-Hernandez et al., 1997; Angulo et al., 1998; de  
43 Marsily et al., 2000). However the head or concentration data are generally few and the kriging  
44 estimate of these variables with a usual variogram model is not necessarily consistent with the flow  
45 physics. Various methods have been proposed to constrain estimates by flow physics, a synthesis and  
46 non-exhaustive review of which is presented here.

47 As the head decreases in the flow direction, it is better to carry out the kriging using a non-stationary  
48 model (Kumar, 2007). An anisotropic variogram allows one to particularize the direction of flow at the  
49 macroscopic scale. Taking covariates into account allows for improvement of the modelling. Thus,  
50 Desbarats et al. (2002) introduce the topography as an external drift of the head, choosing the  
51 appropriate support of regularization of the digital elevation model. Varouchakis et al. (2012) make a  
52 non-linear estimation of the head (using a Gaussian anamorphosis or a modified Box-Cox  
53 transformation), with the necessary correction for the back transformation of the estimated values.  
54 On a real case, the cross-validation results are improved compared to the usual kriging.

55 Curvilinear coordinate transformation allows for bringing back the “natural coordinates” of flow, i.e.  
56 hydraulic head and stream functions (Roth, 1995; Rivest et al., 2012). In order to explicitly introduce  
57 flow physics, Brochu et al. (2003) use the formalism of dual kriging to impose to the estimate the form  
58 of the analytical solution of a linear or radial flow calculated for a homogeneous medium. In Rivest et  
59 al. (2008) the covariate used as external drift is a « smooth » output of a finite element flow model  
60 calculated on an uniform transmissivity field, for which boundary conditions must therefore be  
61 specified. Tests on synthetic cases show that the accuracy of the estimate and its realism are improved.  
62 The method was applied to the real case of a dam.

63 On a synthetic case in steady state, Yang et al. (2018, 2019) reconstruct a field of concentrations by  
64 «physics-informed kriging» or «physics-informed cokriging». The method is based on a regression  
65 between observations which are considered as “high fidelity” data, and the average of outputs of flow  
66 and transport simulations which are considered as “low fidelity” data. The difference between kriging  
67 and cokriging lies in the covariance of the discrepancy between low fidelity data and observations. To  
68 improve the modelling, Yang et al. (2018, 2019) suggest a non-stationary model for this discrepancy.

69 In another approach, covariance models are constructed so as to be approximately or accurately  
70 consistent with flow equations. In the case of steady-state linear macroscopic flow and in the absence  
71 of recharge, the linearization of the differential equation linking perturbations of head and  
72 transmissivity provides an approximated solution. Hoeksema and Kitanidis (1984) supposed the head  
73 to be stationary. Dong (1989, 1990) gave a general solution within the framework of the intrinsic

74 random function of order-k (IRF-k) theory and applied it to the inverse problem, by cokriging the  
75 transmissivity from transmissivity and head data. Hernandez et al. (2003) introduced a source term in  
76 the equations for performing geostatistical inversion based on the pilot points method (de Marsily,  
77 1978; de Marsily et al., 1984). Zhang and Neuman (1996) generalized the analytical form of the simple  
78 and cross-covariance to velocity by crossing two flows with different main directions, for comparing  
79 longitudinal and transverse dispersion.

80 However the hypothesis of a uniform head gradient is often too strict. Roth et al. (1998) construct a  
81 variogram  $\gamma(x, x')$  consistent with a steady state flow model with fixed aquifer geometry and  
82 boundary conditions. A set of geostatistical non conditional simulations of the conductivity are used  
83 as input of a flow simulation. The variogram  $\gamma(x, x')$  is calculated, up to the  $\frac{1}{2}$  factor, as the mean  
84 square deviation of the head at pairs of points  $x$  and  $x'$ , computed on a set of flow simulation outputs.  
85 The numerical version of simple and cross-variograms (or covariances) between head and  
86 transmissivity is then used for cokriging these two variables. These experimental non-stationary  
87 variograms are used by Schwelle and Cirpka (2010) to estimate the head and the concentration by  
88 conditional expectation, calculated by the median (rather than the mean) on a set of simulations  
89 constrained by data from different variables. However, the conditional simulations of the  
90 transmissivity (or permeability) they use are obtained by an iterative inverse approach.

91 Pannecoucke et al. (2020) generalize the approach to the vadose zone. Non conditional simulations of  
92 saturated hydraulic conductivity are used as input to flow-and-transport simulations of a non reactive  
93 contaminant. Boundary conditions can vary between the flow simulations, if these conditions are not  
94 precisely known. Applications to a wide variety of problems are presented in Pannecoucke (2020) in  
95 two dimensions (2D) or 3D, for spatial or spatio-temporal estimation: delimitation of a contaminated  
96 zone at fixed contamination threshold and identification of the associated “uncertainty zone”, back-  
97 tracing of the unknown location of a contaminant source, comparison of the estimation with the  
98 results of a (real) 3D tracing experiment, etc.

99 This paper presents original results of kriging with numerical covariance, following the approach of  
100 Pannecoucke (2020), in order to take into account the heterogeneity of a contamination plume.  
101 However an anamorphosis (variable transformation) step is introduced, so that the contamination  
102 estimates are positive. This non-stationary anamorphosis is calculated on the set of the outputs of the  
103 flow-and-transport simulations. The 2D application cases refer to the highly non-stationary context of  
104 a contamination plume from a radioactive waste deposit located near the Chernobyl power station in  
105 Ukraine. A synthetic case is first examined and the estimation using real-world data is then discussed  
106 for comparing three estimation methods: usual kriging, external drift kriging with a simplified flow  
107 simulation as covariate, and kriging with numerical covariance.

## 108 **2. Methods**

### 109 **2.1. “Numerical” covariance**

110 In classical kriging approaches, the covariance is inferred from observations of the variable to be  
111 estimated. However kriging is more general and can be performed based on non-stationary  
112 covariances of type  $C(x, x')$ . Especially, these covariances can be computed between each pair of grid  
113 nodes of the domain from a set of  $N$  realizations of the variable (Pannecoucke et al., 2020), such as:

$$114 \quad C(x, x') = \frac{1}{N} \sum_{n=1}^N [Z_n(x) - \overline{Z(x)}][Z_n(x') - \overline{Z(x')}] \quad (1)$$

115 where  $C(x, x')$  is the “numerical covariance” between grid nodes  $x$  and  $x'$  and  $Z_n(x)$  (respectively  
 116  $Z_n(x')$ ) is the value of the variable  $Z$  at grid node  $x$  (respectively  $x'$ ) for the  $n$ -th realization. For each  
 117 grid node  $x$ , the “numerical” covariance at all 2D grid nodes  $x'$  draws a 2D map. Fig. 1 shows the maps  
 118 of numerical covariances related to two different points  $x$ , for a set of simulated concentration plumes  
 119 (see further section 3.2.). It can be shown that the numerical covariance  $C(x, x')$  (or the numerical  
 120 variogram  $\gamma(x, x')$ ) has the required mathematical properties of a (discrete) covariance or variogram  
 121 function (de Fouquet, 2019; Pannecoucke, 2020).

122 For the application (Section 3) the realizations of the variable of interest are generated by flow-and-  
 123 transport simulations of a contaminant plume. First, a set of geostatistical simulations of the hydraulic  
 124 conductivity is constructed and simulations of the source term of the contamination are taken from a  
 125 previous work (Nguyen, 2017). The flow-and-transport simulations are based on the synthesis of  
 126 several studies (Le Coz et al., 2023). Nevertheless, it should be noted that the influence of the range of  
 127 the hydrogeological parameters was reduced compared with the variability between realizations and  
 128 with the boundary conditions (Pannecoucke et al., 2019; Pannecoucke, 2020).

129 The kriging with numerical covariance can be seen as a generalization of kriging in natural flow  
 130 coordinates. Indeed, a stationary covariance  $K$  applied after a space transformation  $f$  can be written  
 131 as  $K(f(x) - f(x'))$  and therefore is of the form  $C(x, x')$  when the function  $f$  is not linear. Conversely,  
 132 in the case of a complex phenomenon, such as the development and movement of a plume, it is not  
 133 necessarily the case that a space transformation  $f$  and a stationary covariance  $K$  exist such that the  
 134 non-stationary covariance  $C(x, x')$  is of the form  $K(f(x) - f(x'))$ . It is thus pragmatic to calculate the  
 135 empirical non-stationary covariance (the “numerical covariance”) on a set of outputs of the numerical  
 136 flow-and-transport simulations.

## 137 **2.2. Non-stationary Gaussian transformation**

138 In practice, kriging with a numerical covariance does not ensure that the estimated concentration is  
 139 positive. The non-linear estimation in the well-known anamorphosed Gaussian model (Chilès and  
 140 Delfiner, 2012) allows avoiding negative estimates for positive quantities.

141 Due to the non-stationarity of the local distributions of the simulated contaminant concentrations  
 142 (especially, inside vs. outside the contaminant plume), a Gaussian transformation of these distributions  
 143 is performed at each grid node. The non-stationary anamorphosis  $\Phi_x$  is calculated from the  $N$  outputs  
 144 of the flow-and-transport simulation at the grid node  $x$ . Thus:

$$145 \quad Z_n(x) = \Phi_x[Y_n(x)] \quad (2)$$

$$146 \quad \text{and conversely } Y_n(x) = \Phi_x^{-1}[Z_n(x)]$$

147 where  $Y_n(x)$  is the Gaussian transform of the contaminant concentration  $Z_n(x)$  of the  $n$ -th flow-and-  
 148 transport simulation at grid node  $x$ . Here the anamorphosis is calculated using its Hermite polynomial  
 149 expansion (with 30 polynomials; Chilès and Delfiner, 2012).

150 The numerical covariance is then calculated on the set of the  $N$  fields of Gaussian transformed  
 151 variables.

152 The Gaussian data used for the estimation are deduced from the measurements (or from the reference  
153 data in the synthetic case) using the inverse of the anamorphosis ( $\phi_x^{-1}$ ). The kriging estimate and the  
154 kriging variance are calculated in the Gaussian world.

155 Finally the conditional expectation of the concentration, its quantiles or any related functions, are  
156 calculated through the known relationship with the spatial distribution of a Gaussian random function;  
157 for example (Matheron, 1976; Chilès and Delfiner, 2012):

$$158 \quad E(Z(x)|Z(x_i), i = 1, \dots, n) = \int \phi_x(Y^*(x) + \sigma(x)u) g(u) du \quad (3)$$

159 where  $E(Z(x)|Z(x_i), i = 1, \dots, n)$  is the conditional expectation of the concentration;  $Y^*(x)$  is the  
160 kriging with known mean (equal to 0),  $\sigma(x)$  the standard deviation of the kriging error,  $g$  the density  
161 of the reduced normal distribution and  $u$  is the variable to be integrated. This expression can be  
162 calculated using the formula of the Hermite-polynomial expansion of the Gaussian anamorphosis  
163 (Chilès and Delfiner, 2012). A known correction is applied on the standard deviation if the kriging with  
164 unknown mean (ordinary kriging) is used (Emery, 2006). Equation (3) ensures that the estimated  
165 concentration is positive, because the concentrations outputs of the flow-and-transport simulations  
166 used for calculating the anamorphosis are positive.

### 167 **2.3. Estimation workflow with numerical covariances**

168 In summary, the method involves the following steps:

- 169 • simulating a set of unconditional hydraulic conductivity fields (geostatistical unconditional  
170 simulations by any classical method, e.g. the turning bands as used here; Chilès and Delfiner, 2012);
- 171 • simulating initial source term of the contamination (geostatistical conditional simulations in the  
172 following example)
- 173 • simulating flow-and-transport on each hydraulic conductivity field to obtain a set of concentration  
174 fields;
- 175 • calculating the non-stationary anamorphosis of the concentrations at each grid node  $\Phi_x$  from the  
176 set of simulated concentration fields;
- 177 • calculating the Gaussian variable for each simulated concentration field using  $\Phi_x^{-1}$  ;
- 178 • calculating the numerical covariances from the set of Gaussian variables;
- 179 • calculating Gaussian data as the Gaussian transform of concentrations data using  $\Phi_x^{-1}$  at data  
180 points;
- 181 • kriging of Gaussian variable from Gaussian data and computing associated kriging variance, using  
182 the numerical covariances on Gaussian transforms;

- 183 • returning to the estimated concentration (or any function of the concentration, e.g. conditional  
184 quantiles or conditional distribution) thanks to Eq. (3)

### 185 3. Case study

#### 186 3.1. The trench T22 in Chernobyl

187 The Chernobyl accident of April 1986 released large amounts of radionuclides into the surrounding  
188 environment. In 1986 and 1987, emergency clean-up was carried out in the irradiated Red Forest, in  
189 the vicinity of the Chernobyl nuclear power plant. About  $500 \times 10^3 \text{ m}^3$  of contaminated topsoil,  
190 vegetation and other radioactive materials were buried in situ in hundreds of trenches dug within the  
191 permeable sandy superficial formation, directly above the water table and without engineered  
192 moisture-proof barriers (Kashparov et al., 2012).

193 In 1998, an experimental research facility was built around the trench T22 in order to investigate  
194 radionuclide migration processes. Then, for about 20 years, the trench T22 has been monitored  
195 extensively, allowing an advanced characterization of the site geology, the water flow regime in the  
196 unsaturated and saturated zones, the radionuclide distribution, and the speciation within and outside  
197 the trench (e.g., Bugai et al., 2012; Kashparov et al., 2012). Moreover, acquired data and knowledge  
198 were integrated into numerical models that simulate the migration of radionuclides, especially  $^{90}\text{Sr}$   
199 (Strontium-90), from the trench to the underlying aquifer (Bugai et al., 2012; Nguyen, 2017). Le Coz et  
200 al. (2023) present a detailed synthesis of previous works used for the flow-and-transport modeling  
201 near the trench.

#### 202 3.2. Flow-and-transport simulations

203 Between 2000 and 2015, groundwater was collected for  $^{90}\text{Sr}$  activity measurements generally once or  
204 twice a year at four sampling depths (layers L1 to L4, from top to bottom) in six wells (AP1 to AP6, from  
205 upstream to downstream) along a cross-section A-B, approximately perpendicular to the trench T22  
206 and assumed to be parallel to the groundwater flow lines (Fig. 2 a). A groundwater flow-and-transport  
207 model was used for simulating in 2D, i.e. along the vertical cross-section A-B (Fig. 2 b), the development  
208 of the  $^{90}\text{Sr}$  plume highlighted by this monitoring (Le Coz et al., 2023). Model parameters were constant  
209 for each geological unit: eolian deposits, alluvial deposits with floodplain or channel facies (Le Coz et  
210 al., 2023). These parameters, as well as the boundary conditions, were set such that the simulated  
211 time series of  $^{90}\text{Sr}$  activity were consistent with most of the measurements. This simulation with  
212 especially uniform hydraulic conductivity by geological unit is called the *calibrated simulation*  
213 hereafter.

214 For this study, 400 new flow-and-transport simulations are run by considering the uncertainties on the  
215 model parameters (e.g., the parameter that governs radionuclide retention on aquifer material) and  
216 boundary conditions (e.g., the percolation rate through the trench T22). In addition, for each run,



- 217       • the spatial variability of hydraulic conductivity is simulated through a random field for which  
218           the properties are determined based on textural analysis of aquifer material samples (eolian  
219           and alluvial sand). T
- 220       • the initial distribution of  $^{90}\text{Sr}$  activity within the trench T22 is a geostatistical conditional  
221           simulation (Fig. 2 b) constructed from both *in situ* emission profiles and laboratory analysis of  
222           waste samples (Nguyen, 2017).

### 223       **3.3. Spatial estimation**

224       A set of 357 simulated fields (some simulations did not converge) of the  $^{90}\text{Sr}$  activity in the saturated  
225       zone along the cross-section A-B is extracted from the flow-and-transport simulations on 1<sup>st</sup> October  
226       2006 (01/10/2006). On this date, the  $^{90}\text{Sr}$  activity was measured in groundwater collected at all  
227       sampling depths in wells AP1 to AP6 (Fig. 2 b). On the *calibrated* simulation, the  $^{90}\text{Sr}$  plume extends up  
228       to 40 m from the trench with a downward component (Fig 3a). The geometry of the plume in the  
229       simulation set is similar, although its extent varies significantly (e.g., Fig 3b). The distribution of the  $^{90}\text{Sr}$   
230       activities computed from the simulation set at the sampling wells shows significant ranges, especially  
231       in the core of the plume (highest values) (Fig. 4), yet these ranges generally include the activity of the  
232       *calibrated simulation* and of the measurements.

233       Three estimation methods of the  $^{90}\text{Sr}$  activity are compared in a cross-validation approach: (i) ordinary  
234       kriging (OK); (ii) kriging with an external drift (KED), the drift being the  $^{90}\text{Sr}$  activity field extracted from  
235       the *calibrated simulation* (Fig. 3a); and (iii) kriging with numerical covariances (KNC) using a non-  
236       stationary Gaussian transformation, with covariances computed on the Gaussian transforms of the  
237       simulated fields of  $^{90}\text{Sr}$  activity (simulations 2 to 357).

238       First, the simulated field 1 (Fig. 3b) is used as a synthetic case in order to compare the results with a  
239       known “reality” (this field is not considered for the covariance computation). Estimations are  
240       conditioned (i) at the wells AP1 to AP6 and then (ii) only at the most downstream well (AP6), the  $^{90}\text{Sr}$   
241       activity at the wells AP1 to AP5 being used for the assessment.

242       Then the actual measurements are considered. As for the synthetic case (ii) the estimations are  
243       performed only with the data belonging to the most downstream well (AP6), the  $^{90}\text{Sr}$  activity at the  
244       wells AP1 to AP5 being used for the assessment.

## 245       **4. Results**

### 246       **4.1. Synthetic case**

247       The  $^{90}\text{Sr}$  activities extracted from the simulated field 1 (i.e. the synthetic data) at the sampling locations  
248       are relatively high with respect to the activities extracted from the other simulations, included the

249 *calibrated* one (Fig. 4). At the wells, the relationship between activity "data" and covariate (*calibrated*  
250 simulation) is approximately linear (Fig. 4).

251 Pragmatically, OK and KED kriging are done using an isotropic linear variogram, for avoiding the  
252 variogram fitting phase under unfavorable conditions (few data and high variability). When all data at  
253 the wells AP1 to AP6 are used in the estimation, the OK estimation (Fig 5a) is consistent with the  
254 simulated field 1 (Fig. 3b) only close to these wells, whereas the KED and the KNC estimations (Fig. 5 b  
255 and c) reproduce the geometry (extent and downward component) of the <sup>90</sup>Sr plume on the whole  
256 modeling domain. In addition, OK and KED result in negative <sup>90</sup>Sr activity estimated values in the  
257 bottom part of the domain, below the <sup>90</sup>Sr plume. KED and KNC are similar according to the root mean  
258 square error (RMSE) criterion (Tab. 1).

259 When only synthetic data at the well AP6 are used, the geometry of the <sup>90</sup>Sr plume is still well  
260 reproduced by KED and KNC but not at all by the OK (Fig. 6). KED results in negative concentrations  
261 outside the <sup>90</sup>Sr plume. The RMSE criterion calculated on all nodes (Tab. 1), or on the five validation  
262 wells only, favors the KNC, and it is nearly the same for the other statistical criteria (Fig. 7 and Tab 2).

263 A scatter diagram between the estimate and "reality" (Fig. 8) confirms the good accuracy of KED and  
264 KNC.

265 A test with the average of the 356 others simulations (instead of the *calibrated* simulation) as external  
266 drift leads to very similar results, only slightly less precise.

#### 267 **4.2. Actual measurements**

268 The actual measured <sup>90</sup>Sr activities at some sampling locations differ significantly from the activities  
269 extracted from the *calibrated simulation*, but are generally (except in one location) included in the  
270 range of activities extracted from the 357 additional simulations (Fig. 4). The relationship between the  
271 activity at the wells and the covariate is no longer linear.

272 As for the synthetic case, when the estimations are conditioned by the actual measurements at the  
273 well AP6, OK does not reproduce the lateral variability of the <sup>90</sup>Sr activity with depth (Fig. 9a). KED  
274 maintains the geometry of the <sup>90</sup>Sr plume of the *calibrated simulation*, although the simulated activity  
275 is about a half order of magnitude lower (Fig. 9b). The geometry of the <sup>90</sup>Sr plume estimated by KNC  
276 differs significantly: its extent is reduced but the <sup>90</sup>Sr activity in its core is higher (Fig. 9c). At the five  
277 validation wells, all estimates are positive. The relative performances of KED and KNC are here more  
278 difficult to decide, because the "best" method varies according to the criterion (Fig. 10 and Tab. 3): the  
279 mean error is smaller for KNC, the median error and the RMSE are similar for the two methods,  
280 whereas the mean absolute error favors the KED. The scattered points between the estimates and  
281 observations are in this case further away from the first bisector (Fig. 11). This confirms the greater  
282 inaccuracy than for the synthetic case.

#### 283 **5. Discussion and conclusion**

284 The main processes that govern the development of a contaminant plume in groundwater can be  
285 simulated using a flow-and-transport model. However, due to uncertainties in input data or  
286 misrepresentation of some sub-processes, model output cannot be considered as an accurate

287 representation of the spatial distribution of the contaminant in groundwater at a given date. Usual  
288 kriging, e.g., ordinary kriging (OK), offers an alternative, but a high density of observations is necessary  
289 for providing realistic estimations. When observations are scarce, geostatistical approaches that take  
290 account of physically-based information, e.g., kriging with external drift (KED) or kriging with numerical  
291 covariance (KNC), are more relevant.

292 In KED, physically-based information consists of one or several drifts, which governs the geometry of  
293 the estimated contaminant plume. Following the literature, the output of a simplified flow model,  
294 namely with constant conductivity, is chosen here as external drift. KED requires, therefore, only one  
295 flow-and-transport simulation. In the case of actual observations, this drift is not necessarily consistent  
296 with those data; then KED could modify the variability of the estimated concentrations (location and  
297 level) of the plume.

298 With KNC, a set of several hundred flow-and-transport simulations are used as physically-based  
299 information for computing the numerical covariances (and optionally the non-stationary  
300 anamorphosis). On synthetic cases, when the observations are consistent with the simulation set (i.e.,  
301 when observations are extracted from one reference simulation), the geometry of the contaminant  
302 plume of the reference simulation is fairly well reproduced. In the case of actual observations, the  
303 geometry of the contaminant plume could significantly differ from that of the simulated plumes. If the  
304 numerical covariance does not reflect this discrepancy, then the KNC will lead to inaccurate results.  
305 For the actual measurements of the trench T22, the estimated concentrations seem more contrasted  
306 with KNC than with KED (Fig. 9), with larger maximum values. In addition, in all cases, concentration  
307 fields estimated by KNC associated with a Gaussian anamorphosis are strictly positive.

308 These results corroborate the conclusions of the previous work of Pannecoucke et al. (2020), which  
309 focused on a synthetic case that represents the development of a contaminant plume in the  
310 unsaturated zone. Indeed, this study, based on a real dataset from the saturated zone, confirms that  
311 the non-stationary numerical covariances allow integrating phenomenological knowledge in the  
312 kriging estimation. Moreover, it also shows that kriging with numerical covariance could be more  
313 accurate than external drift kriging, which does not consider the uncertainties related to this covariate,  
314 especially when the geometry of the contaminant plume is complex. The quality of the estimation  
315 depends on the ability of the physically-based realizations used for computing the numerical  
316 covariances to be consistent with the actual flow-and-transport processes and the corresponding  
317 uncertainties.

318 However, the (unique) simplified flow-and-transport simulation used as drift for the KED requires a  
319 much lower calculation time than the set of simulations needed for computing the numerical  
320 covariances. One reviewer points out that “the workflow needed for the KNC also requires a consistent  
321 number of additional steps as compared with the other alternatives.”

322 Rather than a new "better method", kriging with numerical covariance appears to be an alternative to  
323 external drift kriging, which enriches the estimation methods useful to the practitioner. This general  
324 method can be applied to diverse issues in hydrogeology, in multivariate or spatio-temporal modeling,  
325 when estimating or conditioning simulations of hydraulic head or fluxes, backtracking of a plume to  
326 locate the source of a pollution (Pannecoucke, 2020), and in hydro-geochemical modelling. The  
327 method obviously applies to other contexts (e.g. air quality), and provides a very general class of  
328 covariances, e.g. on graphs or other non-Euclidean geometry.

329

### 330 **Acknowledgments**

331 The authors cordially thank the reviewers and editor whose comments greatly helped to improve the  
332 paper. Following the remarks of one reviewer, the “calibrated” simulation was used as external drift,  
333 in preference to the average of the set of simulations.

334

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338 The authors declare there is no conflict of interest in connection with the submitted material

339

### 340 **References**

341 Angulo J. M., Ruiz-Medina M. D. (1998) Series Expansion Approach to the Inverse Problem Journal of  
342 Applied Probability, 35(2)371- 382.

343 Brochu Y., Marcotte D. (2003) A Simple Approach to Account for Radial Flow and Boundary  
344 Conditions When Kriging Hydraulic Head Fields for Confined Aquifers. Mathematical Geology 35 (2)  
345 111–139.

346 Bugai D., Skalsky A., Dzhepo S., Kubko Y., Kashparov V., Van Meir N., Stammose D., Simonucci C.,  
347 Martin-Garin A. (2012) Radionuclide migration at experimental polygon at Red Forest waste site in  
348 Chernobyl zone. Part 2: Hydrogeological characterization and groundwater transport modeling. J.  
349 Appl. Geochem. 27(7), 1359-1374.

350 Chilès J.-P., Delfiner P. (2012) Geostatistics: Modeling Spatial Uncertainty. Wiley.

351 Desbarats A.J. , Logan C.E., Hinton M.J., Sharpe D.R. (2002) On the kriging of water table elevations  
352 using collateral information from a digital elevation model. Journal of Hydrology, 255( 1–4) 25-38.  
353 DOI :10.1016/S0022-1694(01)00504-2.

354 Dong A. (1989). Kriging variables that satisfy the partial differential equation  $\Delta Z=Y$ . *In* Geostatistics.  
355 Vol 1. 3rd Int. Geostat. Cong. M. Armstrong, ed. Kluwer academic publishers.

356 Dong A. (1990). Estimation géostatistique des phénomènes régis par des équations aux dérivées  
357 partielles (Geostatistical estimation of phenomena governed by partial differential equations). Thèse  
358 de Doctorat en Géostatistique. Ecole Nationale Supérieure des mines de Paris ; 262p.

359 Emery X. (2006) Ordinary multigaussian kriging for mapping conditional probabilities of soil  
360 properties. Geoderma 132(1), 75-88.

361 de Fouquet C. 2019. Exercices corrigés de géostatistique .(Geostatistical exercises with correction).  
362 Presses des mines. Paris. 320p.

363 Gómez-Hernández J. J., Sahuquillo J. E., Capilla A. (1997) Stochastic simulation of transmissivity fields  
364 conditional to both transmissivity and piezometric data.1. Theory. J. Hydrol 203(1–4):163–74.

365 Hernandez A. F., Neuman S. P., Guadagnini A., Carrera J. (2003). Conditioning mean steady state  
366 flow on hydraulic head and conductivity through geostatistical inversion. Stochastic Environmental  
367 Research and Risk Assessment 17, 329-338.

368 Hoeksema R.J., Kitanidis P.K. (1984) An application of the geostatistical approach to the inverse  
369 problem in two-dimensional groundwater modelling. Water Resour. Res. 20, 1003-1020.

370 Institut de Radioprotection et de Sûreté Nucléaire (IRSN) (2009). MELODIE Modèle d’Evaluation à  
371 L’ong terme des Déchets Irradiants Enterrés. Notice théorique du code de calcul MELO version 5.0.  
372 (MELODIE Model of Evaluation at L’ong term of Buried Irradiant Waste. Theoretical notice of the  
373 MELO calculation code version 5.0.)

374 Júnez-Ferreira H.E., Herrera G S. (2013) A geostatistical methodology for the optimal design of  
375 space-time hydraulic head monitoring networks and its application to the Valle de Querétaro aquifer.  
376 Environ. Monit. Assess. 185(4):3527-49. doi: 10.1007/s10661-012-2808-5.

377 Kashparov V., Yoschenko V., Levchuk S., Van Meir N. Simonucci C., Martin Garin A. (2012)  
378 Radionuclide migration in the experimental polygon of the Red Forest waste site in the Chernobyl  
379 zone - Part 1: Characterization of the waste trench, fuel particle transformation processes in soils,  
380 biogenic fluxes and effects on biota. J. Appl. Geochem. 27, 1348-1358.

381 Kumar V. (2007) Optimal contour mapping of groundwater levels using universal kriging—a case  
382 study, Hydrological Sciences Journal, 52:5, 1038-1050, DOI: 10.1623/hysj.52.5.1038

383 Le Coz M., Pannecoucke L., Saintenoy A., de Fouquet C., Freulon X., Cazala C. (2023) Do transient  
384 hydrological processes explain the variability of strontium-90 activity in groundwater downstream of  
385 a radioactive trench near Chernobyl? Journal of Environmental Radioactivity 259–260, 107101.  
386 <https://doi.org/10.1016/j.jenvrad.2022.107101>

387 de Marsily G. (1978). De l’identification des systèmes hydrogéologiques. (Identification of  
388 hydrogeological systems) PhD thesis, Univ. Pierre & Marie Curie, Paris.

389 de Marsily G., Lavedan G., Boucher M., Fasanino G. (1984). Interpretation of interference tests in a  
390 well field using geostatistical techniques to fit the permeability distribution in a reservoir model. In  
391 Verly G., David M., Journel A. G., Maréchal A. (eds). Geostatistics for Natural Resources  
392 Characterization, 2nd NATO Advanced Study Institute, South Lake Tahoe, CA, 1983, 831–849. D.  
393 Reidel, Hingham, MA. Pt. 2

394 de Marsily G., Delhomme J.-P., Coudrain-Ribstein A., Lavenue A. M. (2000) Four decades of inverse  
395 problems in hydrogeology. In Zhang D. and Winter C. L., eds. Modeling and Field Investigation in  
396 hydrogeology : a special volume in honor of Shlomo P. Neuman’s 60th Birthday. Geological Society of  
397 America. Special Paper 348, 1-17

398 Matheron G. (1976) A Simple Substitute for Conditional Expectation: The Disjunctive Kriging. In:  
399 Guarascio, M., David, M., Huijbregts, C. (eds) Advanced Geostatistics in the Mining Industry. NATO  
400 Advanced Study Institutes Series, vol 24. Springer, Dordrecht.

401 Nguyen H.L. (2017) Contribution à la quantification des incertitudes portées par la variabilité spatiale  
402 des déchets radioactifs enterrés à Tchernobyl. (Contribution to the quantification of uncertainties  
403 caused by the spatial variability of radioactive waste buried in Chernobyl.) Thèse de doctorat. Mines  
404 ParisTech, PSL Research University.

405 Pannecoucke L., Le Coz M., Houzé C., Saintenoy A., Cazala C., de Fouquet C. (2019). Impact of spatial  
406 variability in hydraulic parameters on plume migration within unsaturated surficial formations.  
407 Journal of Hydrology. 574, 160-168. <https://doi.org/10.1016/j.jhydrol.2019.04.016>

408 Pannecoucke L. (2020) Combinaison de la géostatistique et des simulations à base physique –  
409 application à la caractérisation de panaches de contaminants. (Combining geostatistics and physical-  
410 based simulations – application to contaminant plume characterization) Thèse de doctorat. Mines  
411 ParisTech, PSL Research University.

412 Pannecoucke L., Le Coz M., Freulon X., de Fouquet C. (2020) Combining geostatistics and simulations  
413 of flow and transport to characterize contamination within the unsaturated zone. Sci. Total Environ.  
414 699, 134216.

415 Rivest M., Marcotte D., Pasquier P. (2008) Hydraulic head field estimation using kriging with an  
416 external drift: A way to consider conceptual model information. J. Hydrol. 361 (3–4), 349-361.

417 Rivest M., Marcotte D., Pasquier P. (2012) Sparse data integration for the interpolation of  
418 concentration measurements using kriging in natural coordinates. J. Hydrol., 416-417,72-82

419 Roth C. (1995). Contribution de la géostatistique à la résolution du problème inverse en  
420 hydrogéologie. (Contribution of geostatistics for solving the inverse problem in hydrogeology)  
421 Doctorat en Géostatistique, Ecole nationale supérieure des mines de Paris.

422 Roth C., Chilès J.-P., de Fouquet C. (1998) Combining geostatistics and flow simulators to identify  
423 transmissivity. Adv. Water Resour. 21(7), 555-565.

424 Schwede R. L., Cirpka O. A. (2010). Interpolation of Steady-State Concentration Data by Inverse  
425 Modeling. Groundwater 48(4), 569-579.

426 Varouchakis E.A., Hristopoulos D.T., Karatzas G.P. (2012) Improving Kriging of Groundwater Level  
427 Data Using Non-Linear Normalizing Transformations – A field application, Hydrological Sciences,  
428 57(7) 1404-1419. DOI: 10.1080/02626667.2012.717174, 2012.

429 Yang X., Tartakovsky G., Tartakovsky A. M.(2018). Physics-Informed Kriging: A Physics-Informed  
430 Gaussian Process Regression Method for Data-Model Convergence. arXiv:1809.03461v2 [stat.ML],1–  
431 24

432 Yang X., Barajas-Solano D., Tartakovsky G., Tartakovsky A.M. (2019) Physics-Informed Cokriging: A  
433 Gaussian-Process-Regression-Based Multifidelity Method for Data-Model Convergence. Journal of  
434 Computational Physics 395, 410-431. <https://doi.org/10.1016/j.jcp.2019.06.041>

435 Zhang D., Neuman S. P. (1996) Head and velocity covariance under quasi-steady-state flow and their  
436 effects on advective transport. *Water resources research*, 32(1) 77-83

437

438

439 **Figure captions**

440 Figure 1a,b. Examples of numerical covariance maps, for two different points  $x$  (red circles).

441 Figure 2. Views of the 3-D model of the  $^{90}\text{Sr}$  inventory in 1987 in the trench T22 (computed from the  
442 mean inventory estimated by Nguyen, 2017) and locations of the wells monitored for groundwater  
443 activity. (a) Horizontal view with cumulative  $^{90}\text{Sr}$  activity in depth; (b) vertical cross-section A-B .

444 Figure 3. Activity of  $^{90}\text{Sr}$  in the saturated zone downstream of the cross-section A-B of the trench T22  
445 simulated with the flow-and-transport model on 01/10/2006: (a) *calibrated* simulation used as  
446 external drift; and (b) simulation 1 taken as reference for the synthetic case study

447 Figure 4.  $^{90}\text{Sr}$  activity at the sampling locations. Scatter diagram between (a) the synthetic case in  
448 abscissa (horizontal axis) and the *calibrated* simulation in ordinate (vertical axis) and (b) the actual  
449 measurements (horizontal axis) and the *calibrated* simulation in ordinate, . The grey bar represents  
450 the range of the set of 357 simulations. The black line corresponds to the first bisector.

451 Figure 5. Synthetic case (i.e., flow-and-transport simulation 1). Activity of  $^{90}\text{Sr}$  in the saturated zone  
452 downstream of the cross-section A-B of the trench T22 (Fig. 2) estimated on 01/10/2006 conditioned  
453 at wells AP1 to AP6: (a) ordinary kriging (OK); (b) kriging with external drift (KED); and (c) kriging with  
454 numerical covariances using a non-stationary Gaussian transformation (KNC). The non-filled areas (in  
455 a and b) correspond to negative estimated activities.

456 Figure 6. Synthetic case. Activity of  $^{90}\text{Sr}$  in the saturated zone downstream of the cross-section A-B of  
457 the trench T22 (Fig. 2) estimated on 01/10/2006 conditioned at well AP6 only: (a) ordinary kriging  
458 (OK); (b) kriging with external drift (KED); and (c) kriging with numerical covariances using a non-  
459 stationary Gaussian transformation (KNC). The non-filled area (in b) corresponds to negative estimated  
460 activities.

461 Figure 7. Synthetic case. Profiles of estimated activity by OK, EDK or KNC at wells AP1 to AP5,  
462 conditioned by “data” at well AP6 and “reality”. Logarithmic scale.

463 Figure\_8. Synthetic case. Scatter diagram between estimate (abscissa) and reference value (ordinate)  
464 for the three estimation methods (OK, KED, KNC) at the five validation wells for the estimation from  
465 AP6.

466 Figure 9. Activity of  $^{90}\text{Sr}$  in the saturated zone downstream of the cross-section A-B of the trench T22  
467 (Fig. 2) estimated on 01/10/2006 conditioned with measurements at well AP6: (a) ordinary kriging  
468 (OK); (b) kriging with external drift (KED); and (c) kriging with numerical covariances using a non-  
469 stationary Gaussian transformation (KNC). The non-filled area (in b) corresponds to negative estimated  
470 activities.

471 Figure 10. Profiles of estimated activity by OK, EDK or KNC at wells AP1 to AP5, conditioned by actual  
472 measurements at well AP6, and validation measurements. Logarithmic scale.

473 Figure 11. Actual measurements. Scatter diagram between estimate (abscissa) and exact value  
474 (ordinate) for the three estimation methods (OK, KED, KNC) at the five validation wells for the  
475 estimation from AP6.



476 **Tables**

477 Table 1. RMSE at all 1785 grid nodes, for the two conditioning sets of wells. The very weakly negative  
 478 values of KED are retained. The numbers have been rounded

Well set	OK	KED	KNC
6 conditioning wells	363.	102.	106.
1 conditioning well (AP6)	658.	135.	94.

479

480

481 Table 2. Cross-validation statistics at the five validation wells (AP1 to AP5, 20 values) estimated from  
 482 the data of the AP6 well, for the synthetic case. Negative estimates are zeroed for the statistics of the  
 483 error, but their number is reported. The error is calculated as *estimate – exact data*.

Statistic	OK	KED(0)	KNC
Mean error	-127.	-103.	3.
Median error	306.	0.	10.
Mean absolute error	692.	134.	87.
RMSE	908.	227.	135.
Number of negative estimates	0	9	0

484

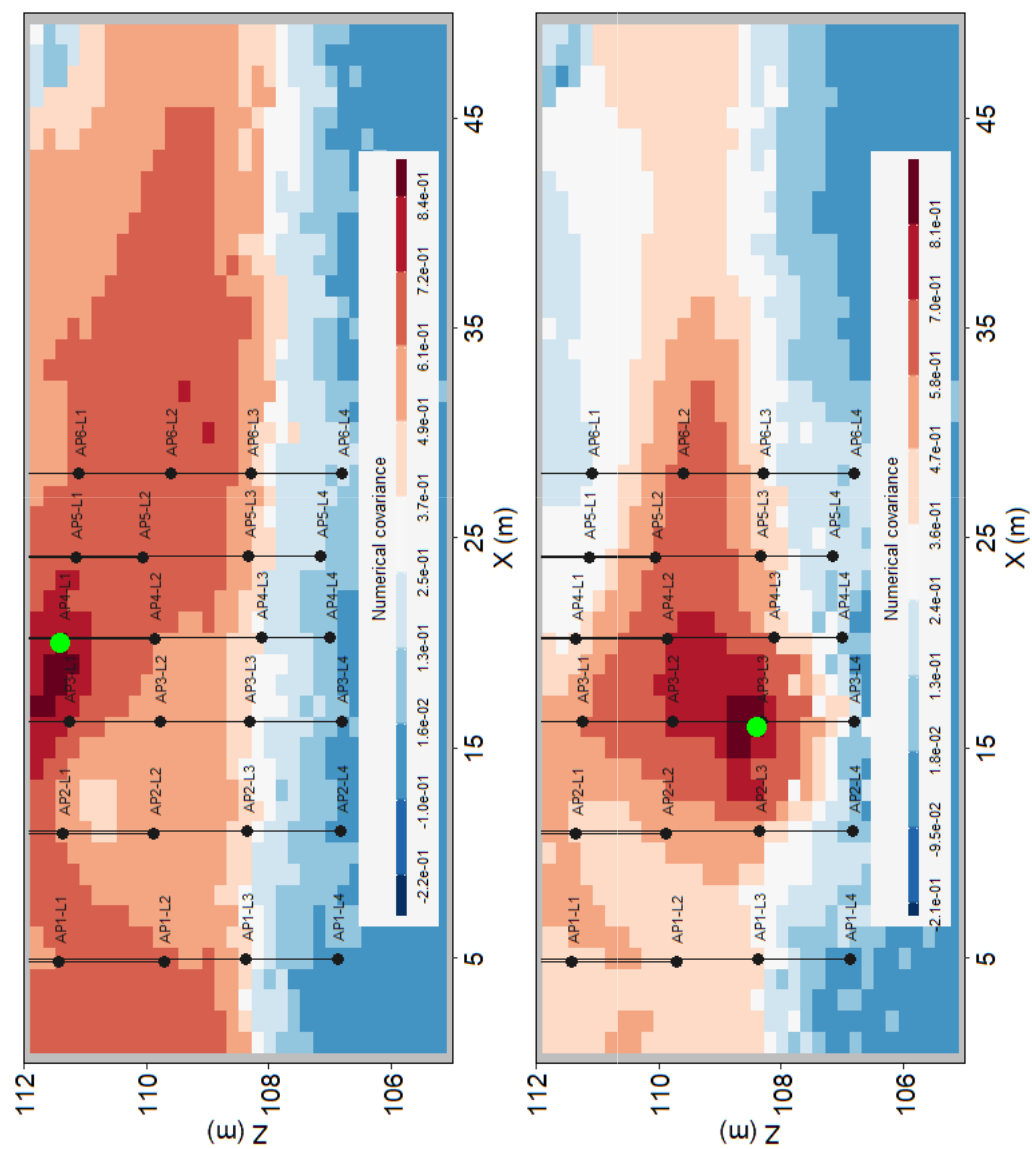
485

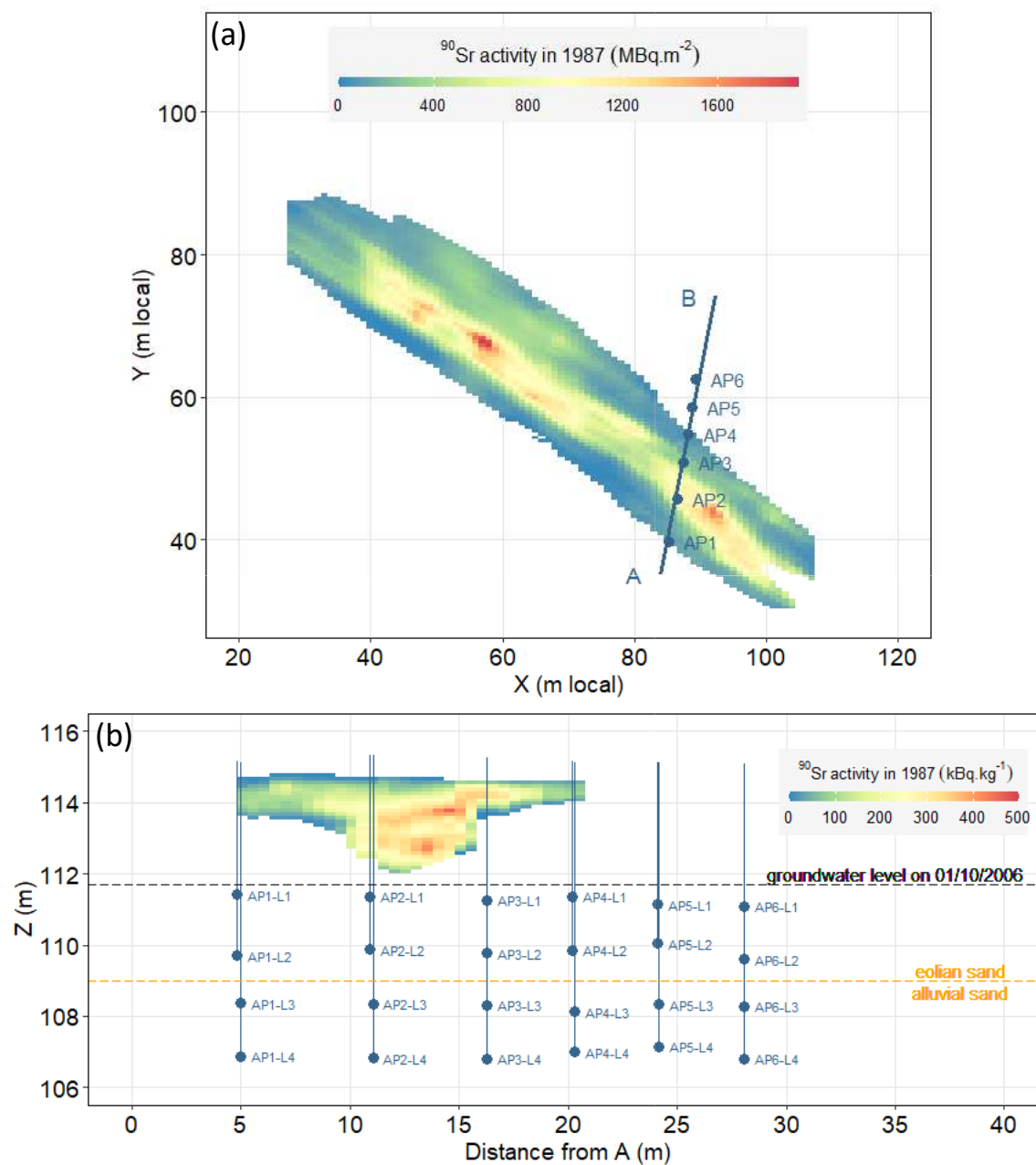
486 Table 3. Cross-validation statistics at the five validation wells (AP1 to AP5, 20 values) estimated from  
 487 the data of the AP6 well, for the actual measurements.

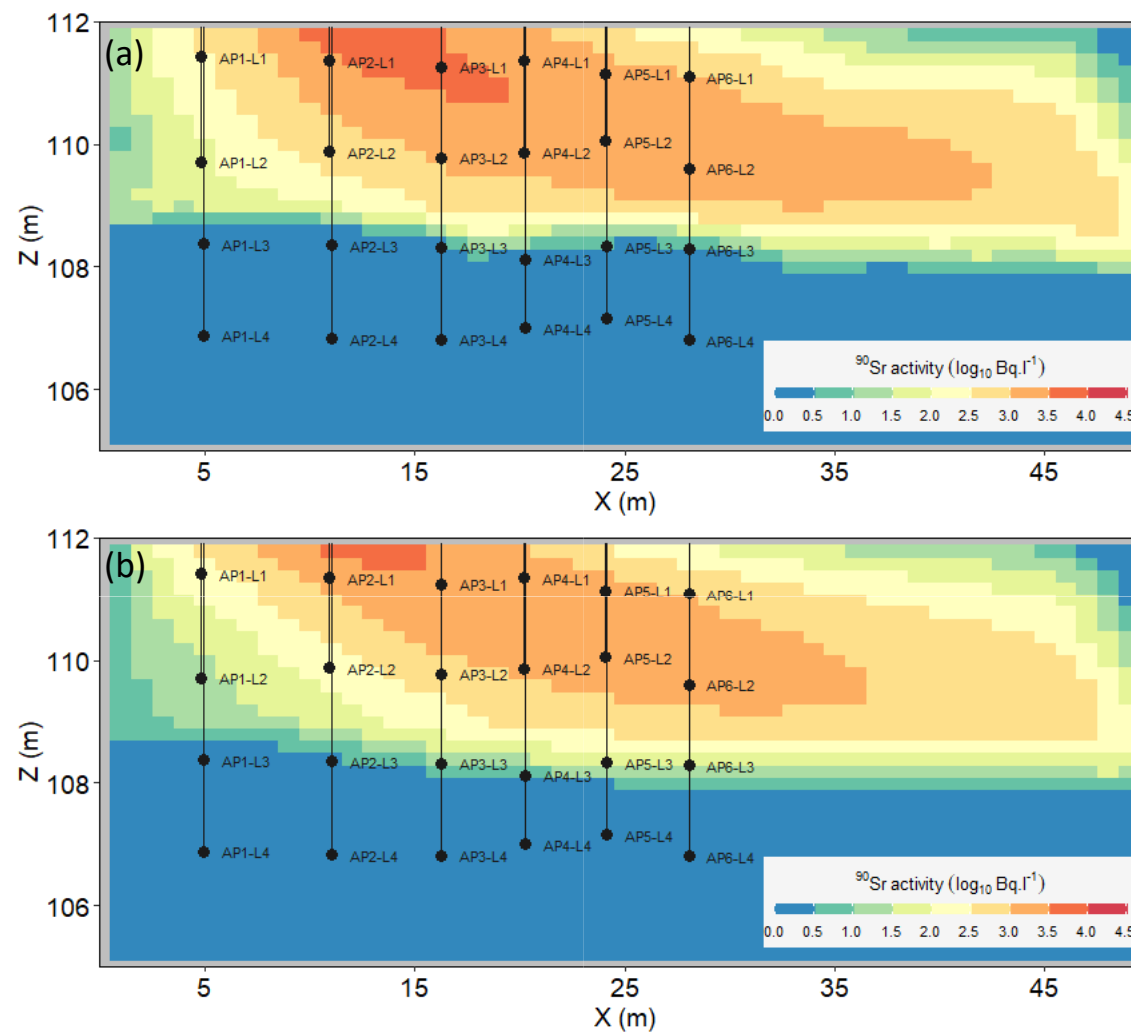
Statistic	OK	KED	KNC
Mean error	-199.	-195.	-80.
Median error	37.	0.	-3.
Mean absolute error	312.	227.	267.
RMSE	589.	472.	477.
Number of negative estimates	0	0	0

488

489







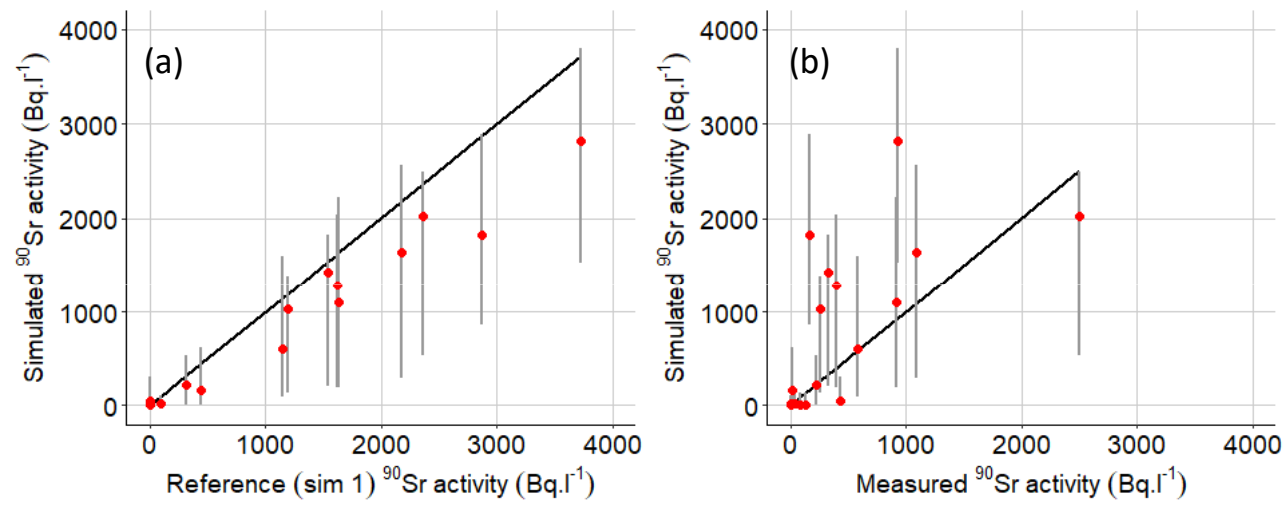


Figure 5

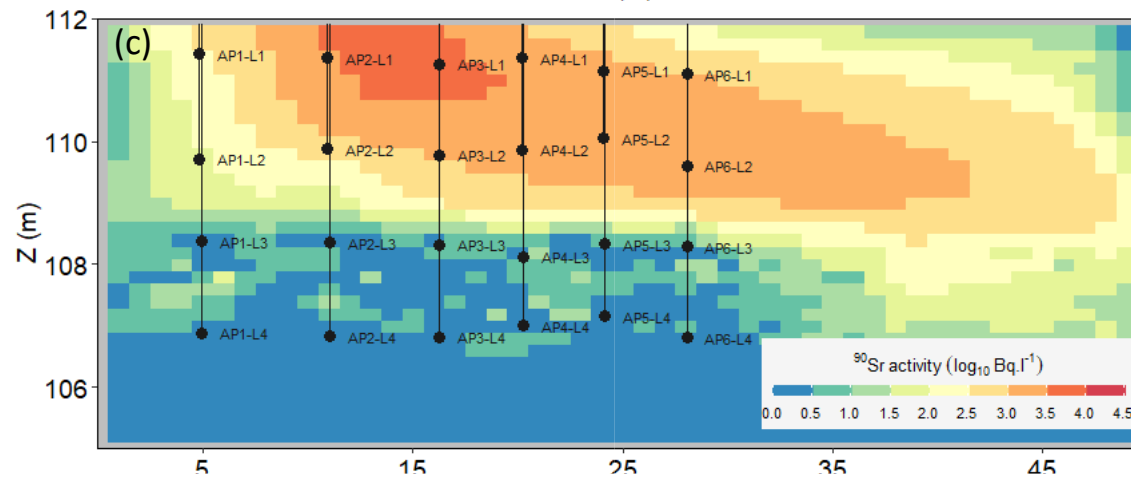
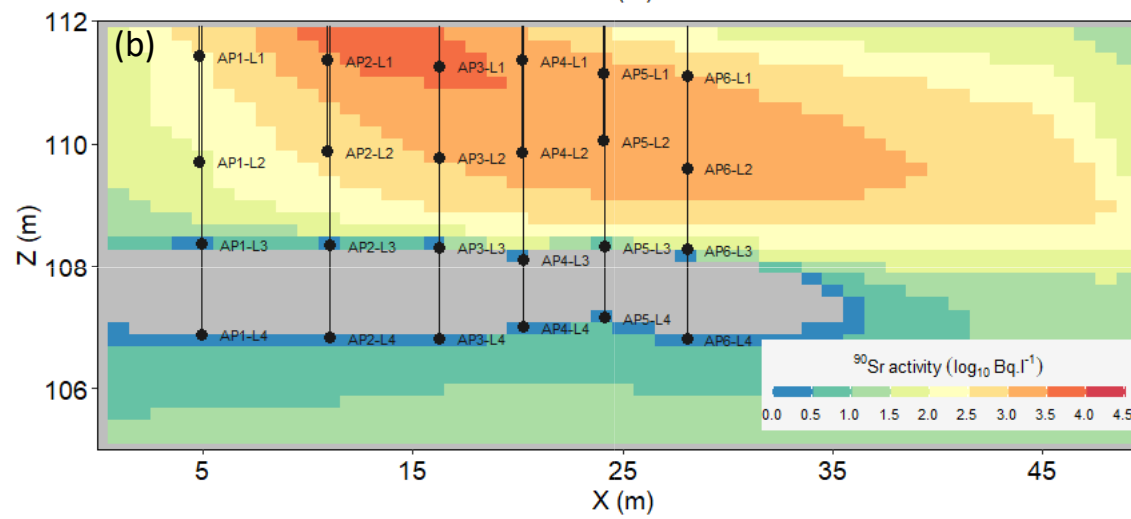
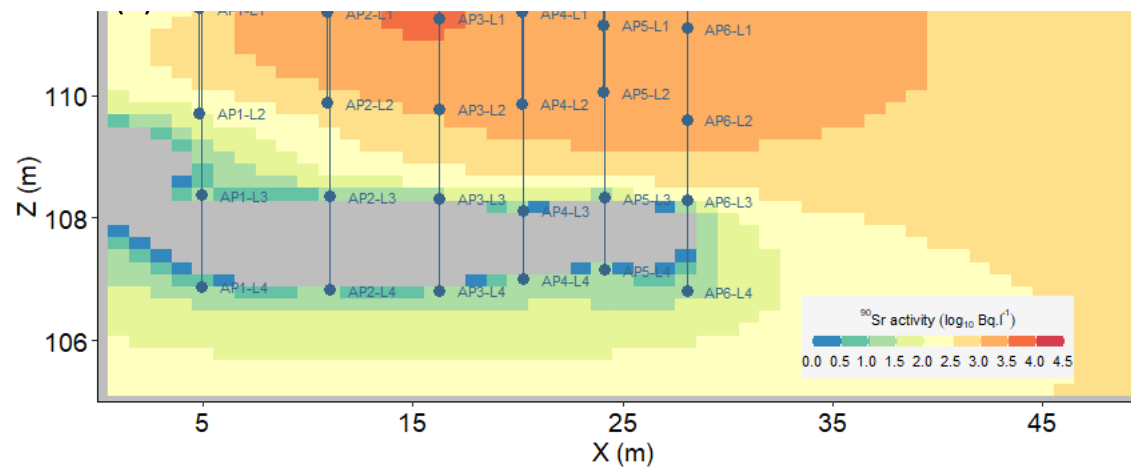
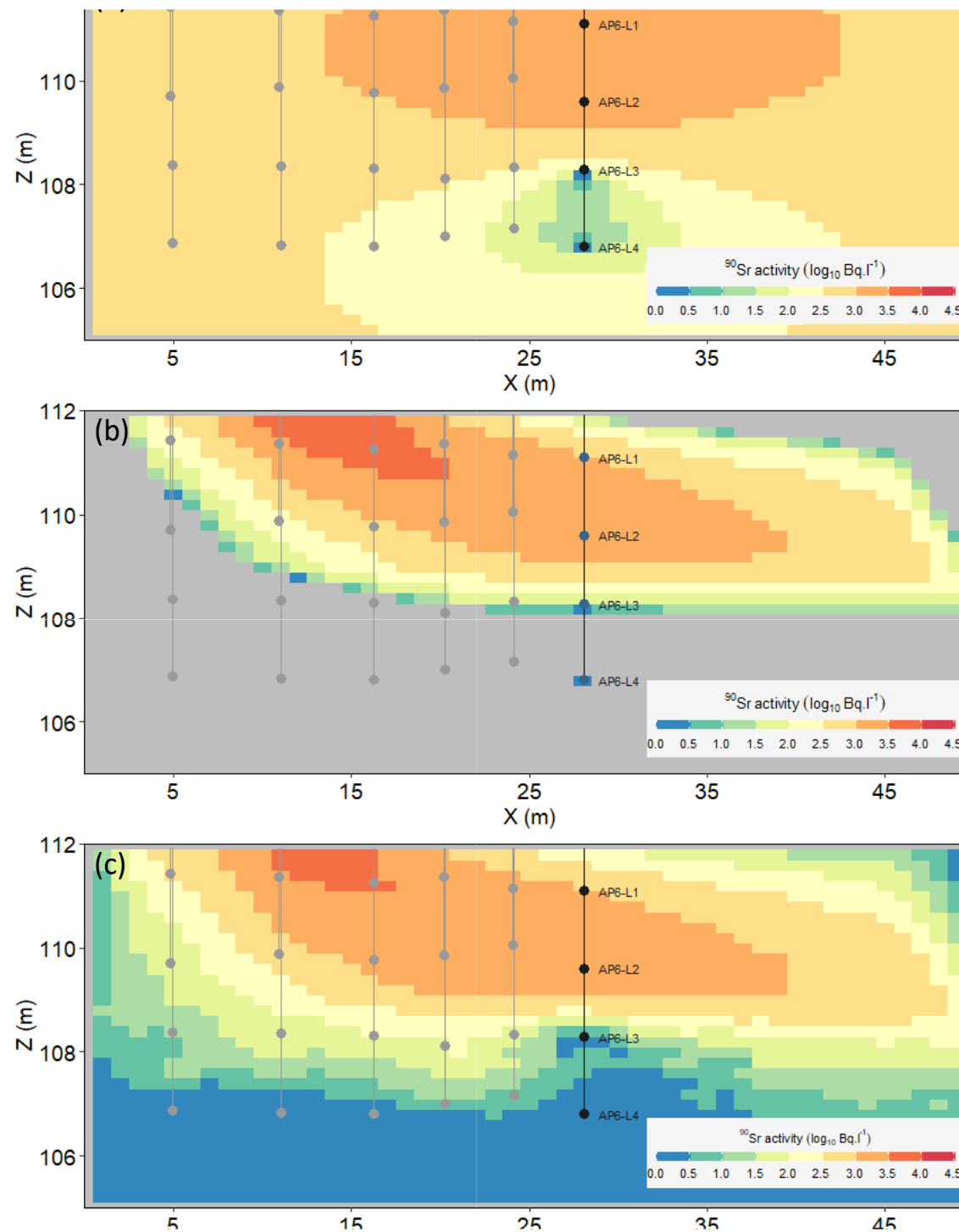
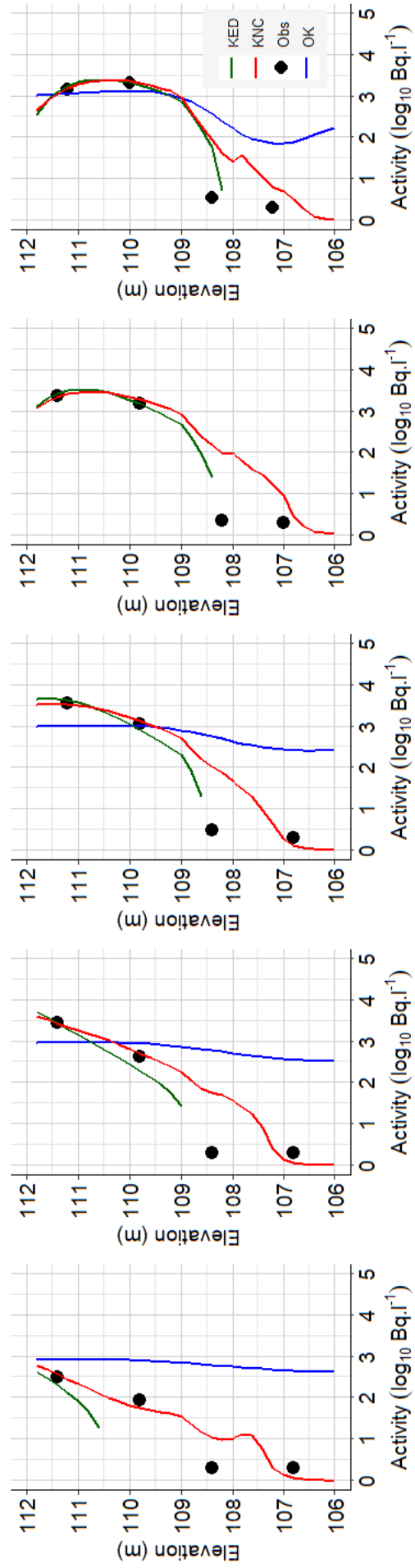


Figure 6







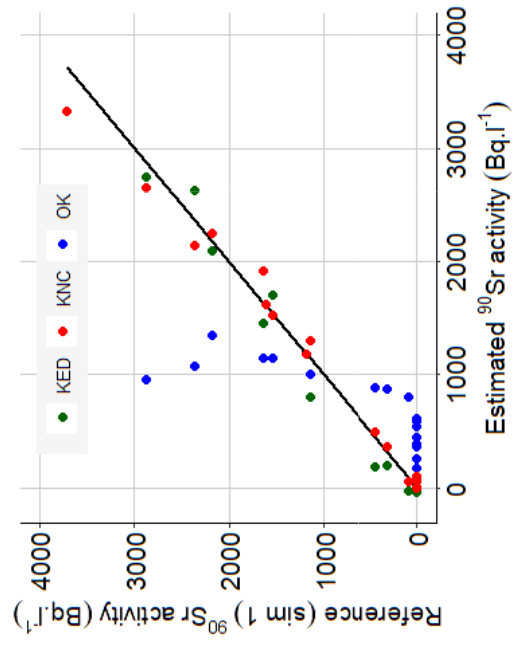
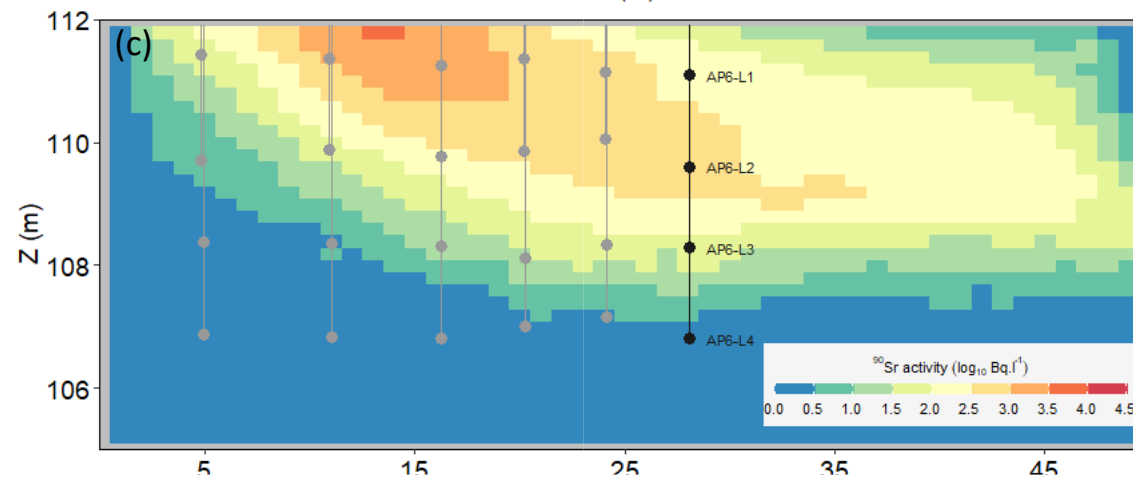
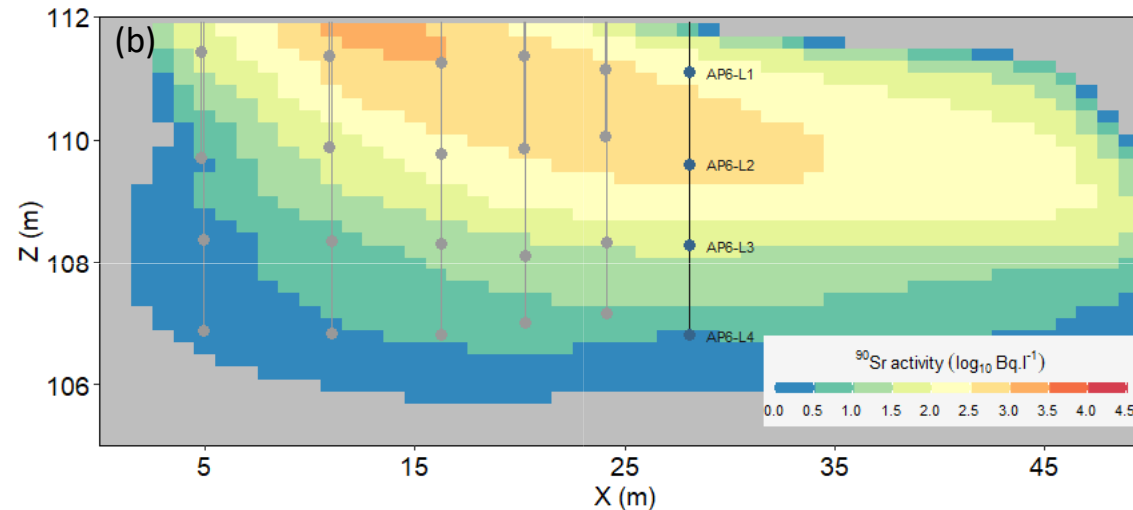
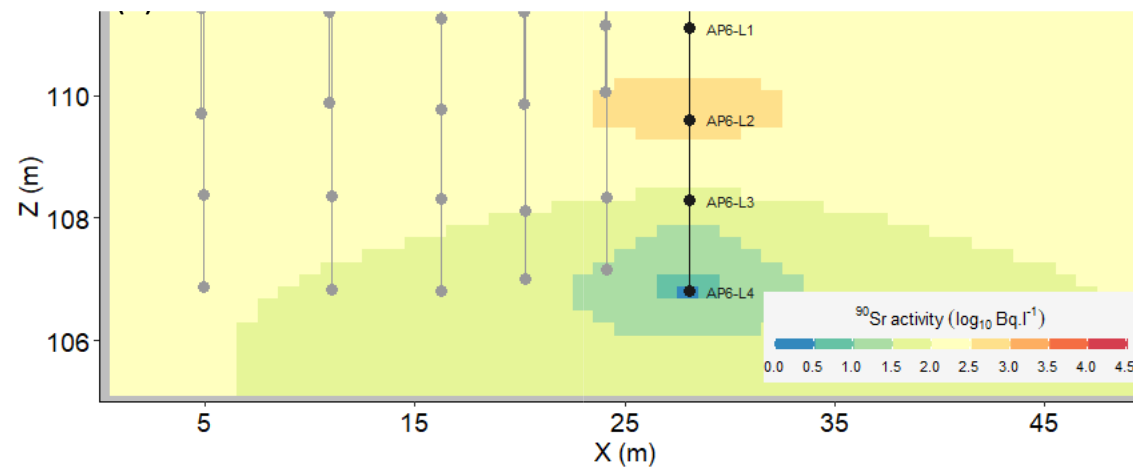
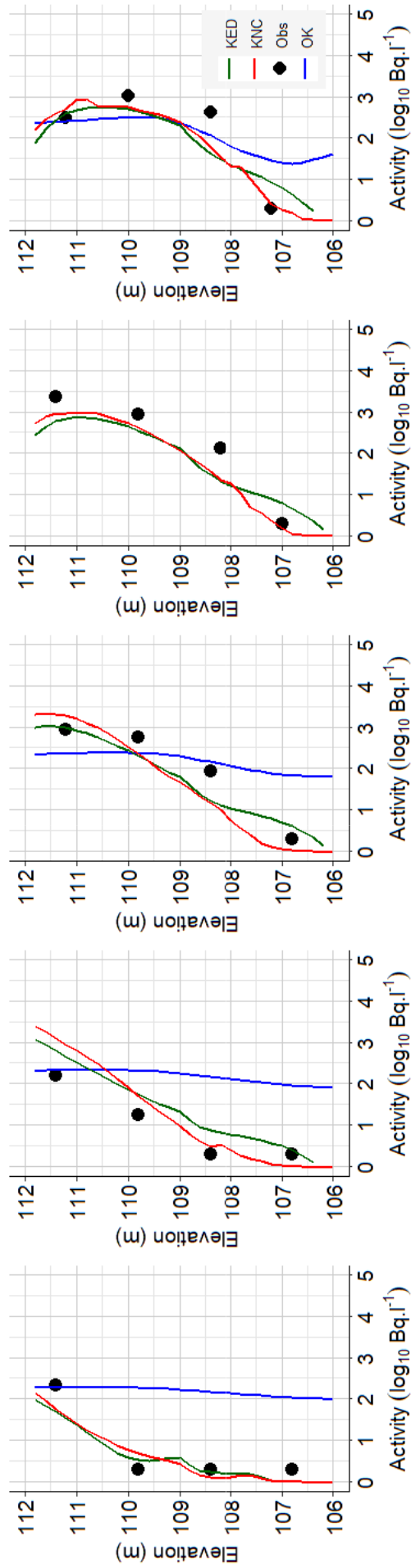


Figure 9







There are no comments