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# Use of machine learning and deep learning to predict particulate $^{137}\text{Cs}$ concentrations in a nuclearized river

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## Abstract

Cesium-137, discharged by nuclear installations under normal operations and deposited in watersheds following atmospheric testing and accidents (i.e. Chernobyl, Fukushima...), has been studied for decades. Thus, modelling of  $^{137}\text{Cs}$  concentration in rivers have been developed based on geochemical approaches and equilibrium assumptions (solid/liquid ratio) as this radionuclide has moved into rivers and oceans due to soil erosion. Recently a new approach is possible to model these concentrations with the popularization of data-driven models based on data acquired in the environment by monitoring networks. In this study, the concentrations of particulate cesium-137 measured near the mouth of the Rhône River (France), a highly nuclearized river, are simulated using two data-driven models, a Hierarchical Attention-Based Recurrent Highway Networks (HRHN) and a Random Forest Regressor (RF). The data-driven predictions were done using only hydrological data (water discharge and suspended solid fluxes) and industrial input of  $^{137}\text{Cs}$ . Although the data-driven models provided a better prediction than a recent empirical model, the best prediction ( $R^2 = 0.71$ ) was obtained with HRHN, a model that considers the temporal aspect of the monitoring data. The most important predictors were the hydrological data at the monitoring station and of the tributary that generate the most sediment flux (Durance River). In fact, the concentration of  $^{137}\text{Cs}$  in the perimeter of this study was more related to hydrology than to nuclear release, as there were few events with high  $^{137}\text{Cs}$  concentrations (concomitant nuclear release and low water discharge). However, the HRHN approach, which is more complex to implement than RF, can predict the concentrations of such events correctly despite their low representation of these events. The results of this study demonstrate the usefulness of data-driven models to assist monitoring programs by filling in gaps or helping to understand observed concentrations.

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4 **Keywords:** radioactivity, suspended sediment, flux, neural network, modeling

## 5 1. Introduction

6 Monitoring the pollution in rivers has been an accepted necessity for many years as aquatic ecosystems  
7 are unfortunately the receptors of emissions from many sectors such as agriculture, industry or  
8 urbanization (Horowitz, 2009; Syvitski et al., 2005; WFD, 2000). For many reasons (logistical, economic,  
9 societal), it is sometimes difficult to set up a sustainable monitoring system (Dethier et al., 2020).  
10 Therefore it makes sense to model the concentrations and fluxes of pollutants (Yang and Wang, 2010)  
11 as modelling appear to be an alternative solution to improve water quality monitoring cost. Thus, many  
12 works have taken place during the last decades to set up empirical models or models based on  
13 geochemical processes to predict the concentration of pollutants such as heavy metals, mercury,  
14 nitrates or radionuclides (Braga et al., 2010; Hu et al., 2007; Zheleznyak et al., 2022; Zhu et al., 2018).  
15 However, these models are often based on restrictive assumptions and require parameters that are  
16 not always available (Cho et al., 2016; Ciffroy and Benedetti, 2018; Desai et al., 2013; Hilko et al., 2012;  
17 Lu et al., 2019). With the recent advances on computer calculations and artificial intelligence  
18 applications in many fields (Abiodun et al., 2018), the use of data-driven models bring a new tool to  
19 model the concentrations and the behavior of these pollutants (Lu et al., 2019; Yaseen, 2021; Ye et  
20 al., 2020). The number of published works on heavy metals simulation using machine learning models  
21 greatly increased from less than 5 publications by year before 2011 to 49 publications in 2020 (Yaseen,  
22 2021). Models such as Artificial Neural Network (ANN), classification and regression tree, linear  
23 regression or support vector machine models are now used to predict the concentration of pollutants  
24 in the environment.



25 However, their use to model radionuclides concentrations in water and suspended sediment (SS) of  
26 rivers remain sparse and new. In a recent review, Dragović (2022) highlights the potential use of ANN  
27 in environmental radioactivity but the studies regarding the hydrosphere are still sparse and mainly  
28 focus on water, not on sediment. For example, Korobitsyn et al. (2008) demonstrated that ANN could  
29 be used to understand the high concentrations of dissolved Strontium-90 observed in the Techa River  
30 (Russia). Kulahci et al. (2006) used an ANN construction to predict the total alpha and total beta  
31 radioactivity as a function of pH, total hardness, electrical conductivity and depth. Seasonal and spatial  
32 variability and classification of radionuclide concentrations might also be done using neural network  
33 such as self-organizing maps (Skwarzec et al., 2009; Tutu et al., 2005) and help improve monitoring  
34 programs. While other machine learning tools such as random forest (with predictive or causal  
35 techniques) had been recently used to analyze cesium-137 ( $^{137}\text{Cs}$ ) contamination of terrestrial plants  
36 (Shuryak, 2023, 2022), no similar work was conducted on liquid or particulate concentration in rivers.  
37 Due to its anthropogenic presence in many environmental compartments, this radionuclide and its  
38 dynamics has been studied for decades, especially in waterways (Antonelli et al., 2008; He and Walling,  
39 1996; Konoplev et al., 2020; Lepage et al., 2014). Thus, during the last decades, empirical modelling of  
40  $^{137}\text{Cs}$  concentration in rivers have been developed, based on assumption like the solid/liquid ratio or  
41 the size of the particles (Ciffroy et al., 2001; Konoplev et al., 2020; Tomczak et al., 2019). However, to  
42 our knowledge, the dynamic in rivers of this radionuclides was not yet subjected to a machine learning  
43 analysis. To pursue this effort of using artificial intelligence in radioecology, a machine learning and a  
44 deep learning approaches were used and compared to a semi-empirical model to estimate the  $^{137}\text{Cs}$   
45 particulate concentrations in a nuclearized river.

## 46 2. Material and method

### 47 2.1. Study area

48 This study was conducted near the outlet of the Rhône River basin ( $\approx 95\,000\text{km}^2$ ) within the Rhône  
49 Sediment Observatory (Lepage et al., 2021; Fabien Thollet et al., 2021) (Fig. 1). This river is the largest

50 source of freshwater to the Mediterranean sea and deliver 6.6 Mt yr<sup>-1</sup> of sediment each year (Poulier  
51 et al., 2019). The Rhône watershed is also characterized by a large climatic and geological  
52 heterogeneity (Olivier et al., 2022) that leads to strong variations of annual SS fluxes (from 1.4 Mt to  
53 18.0 Mt) (Delile et al., 2020). Moreover this river is one of the most nuclearized river over the world  
54 with many nuclear facilities (Eyrolle et al., 2020) such as Nuclear Power Plant (NPP), fuel cycle facilities  
55 and research centers (Fig. 1). The basin has also been affected by the fallouts from the atmospheric  
56 weapons tests and the accident of Chernobyl (Roussel-Debel et al., 2007). Total inventory of <sup>137</sup>Cs was  
57 estimated to be 9.4 10<sup>14</sup> Bq in 1986. This work focuses on the <sup>137</sup>Cs concentrations in SS measured at  
58 the SORA monitoring station from 2010 to 2019. Located at Arles (Eyrolle et al., 2010), this station is  
59 the reference station to evaluate SS and radionuclides concentrations and fluxes near the outlet of the  
60 Rhône River in the Mediterranean Sea (Fig. 1).

## 61 2.2. Suspended sediment sampling

62 The sampling method used to characterize the <sup>137</sup>Cs concentration in SS of the Rhône River is described  
63 in Eyrolle et al. (2020). Briefly, for baseflow period (water discharge lower than 3000 m<sup>3</sup> s<sup>-1</sup>), 13.5 L of  
64 water are automatically sampled and filtered at 0.5 µm every 80 min and integrated for a month. For  
65 flood period, the previous sampling strategy is stopped, and a specific flood event automatic sampling  
66 is settled using up to five acetate cartridges (0.5 µm) with 5 L collected every 60 min until the clogging  
67 of the cartridge.

## 68 2.3. Radionuclide analyses and concentrations (endogenous variable)

69 Measurement of <sup>137</sup>Cs is described in Eyrolle et al. (2020). The SS samples were slowly (approx. 2  
70 weeks) evaporated (80 °C) to dryness, ashed and put into tightly closed plastic boxes (17 mL or 60 mL)  
71 for gamma-ray spectrometry measurements (20–60 g) using low-background and high-resolution High  
72 Purity Germanium detectors. Measurements are regularly checked for background noise and drift  
73 using multi-gamma calibration sources in accordances with national standards. The detectors are also  
74 used in national and international intercomparisons. Results are expressed in Bq kg<sup>-1</sup> (d.w.) and each

75 sample was measured for 3 days to achieve detection limits of 0.5 Bq kg<sup>-1</sup> (d.w.). Measured activities  
76 are decay-corrected to the date of sampling. The activity uncertainty (k=2) was estimated as the  
77 combination of calibration uncertainties, counting statistics, and summing and self-absorption  
78 correction uncertainties.

79 For the studied period (2010-01-05 to 2019-06-04), 269 SS including 112 during baseflow and 157  
80 during flood events were measured (Fig. 2– supplementary information). From 2010 to 2019, the  
81 average particulate concentration was  $7.8 \pm 6.1$  Bq kg<sup>-1</sup> for <sup>137</sup>Cs. Higher mean concentration was  
82 observed during baseflow ( $11.4 \pm 7.4$  Bq kg<sup>-1</sup>) than during flood events ( $5.3 \pm 3.1$  Bq kg<sup>-1</sup>).

83 The annual flux of particulate <sup>137</sup>Cs is calculated by multiplying its particulate concentration by the flux  
84 of SS (water discharge multiplied by SS concentration) for the sampling periods and summed by year.  
85 In 2015, while water discharge and SS concentration were measured, most of the SS sampling was not  
86 conducted due to logistical issues. To estimate the flux during this period, the year has been cut  
87 according to the hydrology to distinguish floods and baseflows regarding the flood threshold (3000 m<sup>3</sup>  
88 s<sup>-1</sup>).

## 89 2.4. Machine learning methodology

### 90 2.4.1. Input variables (exogenous variable)

91 Two different types of exogenous variable were used: routine radioactive liquid releases and  
92 hydrological information. Note that with this approach, we do not apply any transit time between the  
93 monitored stations (Fig1) and Arles, and we do not consider physicochemical variables such as particle  
94 size or organic content as such variables were not available for the whole studied period. All the data  
95 used are available in the supplementary information (SI).

#### 96 2.4.1.1. Nuclear liquid effluents

97 As explained previously, nuclear facilities are allowed to release radioactive effluent directly into the  
98 Rhône River. Such release must respect concentration thresholds and must be carried out under

99 normal hydrological conditions (baseflow), excluding low-level water and flood. For  $^{137}\text{Cs}$ , the main  
100 source of liquid effluent in the Rhône River is the reprocessing center of Marcoule (Fig. 1) which  
101 represent most of the annual liquid emission of the  $^{137}\text{Cs}$ . In fact, the mean annual releases from 2010  
102 to 2016 was  $20.3 \text{ GBq y}^{-1}$  of  $^{137}\text{Cs}$  for the center of Marcoule while the sum of the mean annual releases  
103 of the four NPPs was  $0.1 \text{ GBq y}^{-1}$  of  $^{137}\text{Cs}$ . Quantity of  $^{137}\text{Cs}$  (MBq) released was estimated for each  
104 measurement at the monitoring station without applying a transit time. The duration of the releases  
105 (in hours) and the total flux of SS (t) that transited in the river during the release periods was also used  
106 as exogeneous data.

#### 107 2.4.1.2. Water discharge and suspended sediment concentration

108 In addition to the SORA station, water discharge ( $Q$  in  $\text{m}^3 \text{ s}^{-1}$ ) and SS concentration (SSC in  $\text{mg L}^{-1}$ ) were  
109 also acquired at Jons, which is the reference station to evaluate concentrations and fluxes from the  
110 Upper Rhône River, and at the outlet of the main tributaries: Saône, Isère, Ardèche and Durance rivers  
111 (Fig. 1). Acquisition of  $Q$  and SSC are described in (Lepage et al., 2021) and was conducted within the  
112 Rhône Sediment Observatory. Hourly water discharges ( $Q_H$ ) were calculated by conversion of water  
113 level measurements through stage-discharge rating curves, otherwise through numerical modelling (F.  
114 Thollet et al., 2021). During the studied period, the mean water discharge was  $1520 \pm 818 \text{ m}^3 \text{ s}^{-1}$ ,  $566$   
115  $\pm 317 \text{ m}^3 \text{ s}^{-1}$ ,  $65 \pm 131 \text{ m}^3 \text{ s}^{-1}$ ,  $93 \pm 125 \text{ m}^3 \text{ s}^{-1}$ ,  $312 \pm 150 \text{ m}^3 \text{ s}^{-1}$ ,  $399 \pm 420 \text{ m}^3 \text{ s}^{-1}$ , respectively at Arles,  
116 Jons, Ardèche, Durance, Isère and Saône stations (Fig. 3). The SSC at most stations are derived from in-  
117 situ turbidity measurements conducted every 10 minutes (Le Bescond et al., 2018). The SSC is then  
118 calculated through the site-specific turbidity-SS rating curve (Navratil et al., 2011), which is determined  
119 on each site for a wide range of concentrations. At Arles, the SSC is measured by filtration of water  
120 sample collected by an automatic water sampler by the MOOSE network (Mediterranean Ocean  
121 Observing System for the Environment) (Raimbault et al., 2014). During the studied period, the mean  
122 SSC was  $51 \pm 160 \text{ mg L}^{-1}$ ,  $19 \pm 46 \text{ mg L}^{-1}$ ,  $9 \pm 18 \text{ mg L}^{-1}$ ,  $114 \pm 606 \text{ mg L}^{-1}$ ,  $82 \pm 455 \text{ mg L}^{-1}$ ,  $14 \pm 17 \text{ mg L}^{-1}$ ,  
123 respectively for Arles, Jons, Ardèche, Durance, Isère and Saône rivers (Fig. 3). While  $Q_H$  was used as  
124 input variable (referred to as  $Q_{\text{Name-of-the-station}}$  in  $\text{m}^3 \text{ s}^{-1}$ ), SSC was not used in this form but after

125 calculating the flux of SS ( $t h^{-1}$ ) by multiplying SSC by Q. The SS flux is referred to as FSS\_Name-of-the-  
126 station.

## 127 2.4.2. Models

### 128 2.4.2.1. Random Forest - RF

129 The random forest (RF) is a supervised learning algorithm consisting of a set of decision trees (Breiman,  
130 2001). It uses the "bagging" method whose general idea is that a combination of learning models  
131 increases the overall result. The regressor algorithm is used to process the regressions. The RF  
132 algorithm randomly selects observations and features to construct multiple decision trees and then  
133 averages the results. Bootstrap samples are used when building trees. Several hyperparameters are of  
134 interest, including:

- 135 • The `n_estimators` hyperparameter which is the number of trees the algorithm built before  
136 taking the maximum vote or taking the averages of the predictions. In general, a higher  
137 number of trees increases performance and makes the predictions more stable, but it also  
138 slows down the computation. 10 values were chosen among a uniform distribution in [80,  
139 200].
- 140 • The `max_depth` hyperparameter is the maximum depth of the tree: 10 values were chosen  
141 regularly distributed in the interval [5, 30].
- 142 • The `max_features` hyperparameter which is the maximum number of features that the random  
143 forest considers splitting a node. It is chosen equal to the number of features which are in our  
144 case the columns of water discharges at the different stations.
- 145 • The `min_sample_leaf` hyperparameter which determines the minimum number of leaves  
146 needed to split an internal node. Ten values were chosen among a uniform distribution in [1,  
147 10].

- 148       • The `min_sample_split` hyperparameter which determines the minimum number of samples  
149       required to split an internal node. Ten values were chosen among a uniform distribution in [1,  
150       30].

151   A randomized search on hyper parameters has been performed, optimized by 5 cross-validated  
152   searches over parameter settings.

#### 153           2.4.2.2. Hierarchical Attention-Based Recurrent Highway Networks - HRHN

154   The proposed deep learning method aims at predicting the future  $T$  of the timeseries of  $^{137}\text{Cs}$   
155   (endogenous variable) from a set of past information  $t$  at  $T-1$  of the presented exogenous variable and  
156   the associated  $^{137}\text{Cs}$  evolution. The architecture chosen here is the HRHN (Hierarchical Attention-Based  
157   Recurrent Highway Networks) an encoder-decoder neural network (Tao et al., 2018). The principle of  
158   this architecture is the use of two neural networks. The first one encodes the input data (exogenous  
159   variable at time  $[t, T-1]$ ) into a latent representation. A hierarchical attention layer weights the  
160   importance of the different elements of this representation. Then, the second network must decode  
161   this information to predict the future  $T$  of the time series of  $^{137}\text{Cs}$  concentration in relation to the  
162   history of concentration  $^{137}\text{Cs}$  at time  $t$  to  $T-1$ . The decoder and encoder may have different layers.  
163   Here the encoder is composed of convolutional layers (Lecun et al., 2015) and Recurrent Highway  
164   Network (RHN) (Zilly et al., 2017) layers. The decoder is composed of RHN layers. The purpose of the  
165   recurrent layers is to capture the temporal dynamics of the series. The convolutional layer aims to  
166   detect patterns in the series (seasonality, peak). Max pooling (Aggarwal, 2018) is also performed  
167   between successive convolutional layers, which can reduce the size of feature maps to avoid  
168   overfitting and improve efficiency. For more information on the nature of the layers we refer the  
169   reader to (Goodfellow et al., 2016).

170   This neural network architecture is associated with several hyperparameters linked to the nature of  
171   these layers. The optimization is carried out with the Hyperband method (Li et al., 2018). The aim of  
172   the optimization algorithm is to minimize the error on both the training and the test base to propose

173 the best performing model with the most interesting parameters and hyper-parameters. The detail of  
174 the grid of possible combinations for hyperparameters is presented. We have decided to keep the  
175 number of convolution layers given in (Tao et al., 2018) (i.e. 3), however their size and the associated  
176 max-pooling will be determined in the following interval for each:

- 177 • CNN window size (`nbr_filter_cnn`) in [3,5,7]. The convolution window size determines the size  
178 of the region over which convolution is applied at each time step. It is important to choose an  
179 appropriate window size to capture the relevant temporal patterns in the data.
- 180 • Number of filters (`dim_filter_cnn`) in [8,16,32,64,128,256]. The number of filters determines  
181 how many different patterns the network can learn. The higher the number of filters, the more  
182 complex the network can be, but this can also make training more difficult.
- 183 • Max pooling size (`dim_max_pooling`) in [2,3,4,5]. The pooling window size determines the  
184 region of the input that will be aggregated into a single output element. In general, a larger  
185 pooling window size reduces the spatial resolution of the output but can also improve the  
186 robustness of the network to minor variations in the input. In contrast, a smaller pooling  
187 window size retains more detail of the input but may also make the network more sensitive to  
188 noise or minor variations.

189 As in Tao et al. (2018), we assume the RHN has same structure in the encoder and the decoder:

- 190 • Hidden layers (`layer_RHN`) in [1,2,3,4,5]. The hidden layers allow the neural network to model  
191 non-linear relationships between inputs and outputs. Each hidden layer in a deep neural  
192 network computes a non-linear transformation of the previous layer's outputs, allowing the  
193 network to learn increasingly abstract and complex features as information is propagated  
194 through the network.
- 195 • Dimension of hidden state (`dim_RHN`) in [8,16,32,64,128,256]. The dimension of the hidden  
196 state determines the size of the hidden state vectors that are calculated at each time step of  
197 the model. A higher dimension of the hidden state can allow the model to capture more

198 complex and subtle information in the data, but it can also make the model slower to train and  
199 require more training data.

200 The intervals chosen for these different hyperparameters are based on the following references  
201 (Chollet, 2018; Goodfellow et al., 2016) and documentation available on Tensorflow. The algorithm  
202 Hyperband has been customized to include optimization of the number of time steps. It implies that  
203 for each combination of selected hyperparameters an update of the data size is performed. This  
204 hyperparameter linked to the data history allows us to determine the quantity of past information  
205 most relevant to predict the future evolution of the endogenous variable. The sequence length (here  
206 monthly) is selected between 9 and 20 past data.

### 207 2.4.3. Splitting methods and cross-validation

208 Two processes were used to split the dataset:

- 209 • The dataset was chronologically separated as a time series as it is required for the HRHN  
210 approach. The train dataset ranges between 2010 to end of 2014 with 198 observations and  
211 the test dataset ranges between end of 2015 to 2019 with 72 observations (Table 1). For RF,  
212 this modelling will be referred as RF\_TS. All the mean with their range of the data used by the  
213 models are displayed in the Table 1.
- 214 • For RF, an additional five cross-validation procedure was performed on the whole data. The  
215 split was performed randomly on the 269 observations. This modelling will be referred as  
216 RF\_CV. With this method, several subsets of training and test data were used, thus considering  
217 the full range of exogenous and endogenous variables.

218 *Table 1: mean (min - max) values of the data used by the data-driven models after chronological splitting.*

<b>Mean (min - max)</b>	<b>Train dataset</b>	<b>Test dataset</b>
Number of data	198	71
water discharge at Arles ( $\text{m}^3 \text{s}^{-1}$ )	2646 (595 - 5420)	2586 (579 - 6857)
water discharge of Ardèche River ( $\text{m}^3 \text{s}^{-1}$ )	202 (5 - 2167)	209 (6 - 2502)



water discharge of Durance River ( $\text{m}^3 \text{s}^{-1}$ )	201 (8 - 1010)	270 (10 - 1715)
water discharge of Isère River ( $\text{m}^3 \text{s}^{-1}$ )	415 (97 - 1130)	375 (118 - 1083)
water discharge at Jons ( $\text{m}^3 \text{s}^{-1}$ )	880 (187 - 2143)	891 (214 - 2932)
water discharge of Saône River ( $\text{m}^3 \text{s}^{-1}$ )	792 (56 - 1713)	853 (38 - 2124)
suspended sediment flux at Arles ( $\text{t h}^{-1}$ )	3119 (16 - 48947)	8547 (8 - 105011)
suspended sediment flux of Ardèche River ( $\text{t h}^{-1}$ )	53 (0,05 - 1640)	68 (0,04 - 2066)
suspended sediment flux of Durance River ( $\text{t h}^{-1}$ )	655 (0,1 - 19807)	7874 (0,1 - 113703)
suspended sediment flux of Isère River ( $\text{t h}^{-1}$ )	389 (3 - 6715)	1351 (4 - 20508)
suspended sediment flux at Jons ( $\text{t h}^{-1}$ )	293 (1 - 4324)	434 (1 - 4714)
suspended sediment flux of Saône River ( $\text{t h}^{-1}$ )	96 (0,5 - 622)	195 (1 - 1593)
$^{137}\text{Cs}$ release by nuclear industries (MBq)	479 (0 - 7586)	671 (0 - 3428)
Duration of the nuclear release (h)	18 (0 - 211)	16 (0 - 81)
Total suspended sediment flux during nuclear release (t)	4320 (0 - 103254)	4397 (0 - 32921)
$^{137}\text{Cs}$ concentration ( $\text{Bq kg}^{-1}$ )	7,8 (2,1 - 24,7)	7,9 (2,2 - 43,9)

219

#### 220 2.4.4. Prediction indicators

221 The proposed models were evaluated using the Root Mean Squared Error (RMSE) following Eq1 and  
 222 the coefficient of determination ( $R^2$ ) following Eq2 as it is commonly used in such studies:

$$223 \quad RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N ([^{137}\text{Cs}] i_{measured} - [^{137}\text{Cs}] i_{modeled})^2} \quad \text{Eq.1}$$

$$224 \quad R^2 = 1 - \frac{(\sum_{i=1}^N ([^{137}\text{Cs}] i_{measured} - [^{137}\text{Cs}] i_{modeled})^2)}{(\sum_{i=1}^N ([^{137}\text{Cs}] i_{measured} - [^{137}\text{Cs}]_{mean})^2)}$$

$$225 \quad \text{Eq. 2}$$

226 With:

- 227 • N is the number of data
- 228 •  $[^{137}\text{Cs}] i_{measured}$  and  $[^{137}\text{Cs}] i_{modeled}$  respectively the  $i$ th occurrence of the measured and  
 229 modeled concentrations of particulate  $^{137}\text{Cs}$
- 230 •  $[^{137}\text{Cs}]_{mean}$  the mean of the  $[^{137}\text{Cs}]_{measured}$ .

231 These performance indicators are classically used in model evaluation. The  $R^2$  measures the degree of  
232 variability of the dependent variable that can be explained by the model. The  $R^2$  is a good measure of  
233 how well the model fits the dependent variables.

#### 234 2.4.5. Sensitivity of the input variables

235 The importance of a feature is computed as the (normalized) total reduction of the criterion brought  
236 by that feature. It is also known as the Gini importance (Breiman, 2001). It calculates each feature  
237 importance as the sum over the number of splits (across all trees) that include the feature,  
238 proportionally to the number of samples it splits.

239 Regarding the HRHN approach, sensitivity analysis in neural networks with a complex architecture is  
240 still a subject in full development notably because of the large number of hyperparameters and  
241 parameters of these models. It (Doshi-Velez and Kim, 2017). We propose here the Permutation feature  
242 importance, a simplistic approach to try to bring elements of answer on the importance of the various  
243 exogenous variables. Initially used for random forests, it is applicable to any model (Wei et al., 2015).  
244 This method has the advantage of not requiring a re-training phase for the model or long simulations,  
245 which can be costly in terms of computing time for HRHN. The concept is straightforward: we measure  
246 the importance of a feature by calculating the increase in the model's prediction error after permuting  
247 the feature. A feature is "important" if shuffling its values increases the model error, because in this  
248 case the model relied on the feature for the prediction. A feature is "unimportant" if shuffling its values  
249 leaves the model error unchanged, because in this case the model ignored the feature for the  
250 prediction. This method is applied to the test set rather than the training set to assess the importance  
251 of the variables in the ability of the model to generalize to the unknown data. This is because the model  
252 is trained on the training set and its coefficients are adjusted to minimize the error on the training  
253 data. If we use the training set to assess the importance of variables, this may lead to an overestimation  
254 of the importance of some variables, as the model has been optimized to minimize the error on these  
255 specific data. In contrast, the validation set is used to assess the ability of the model to generalize to

256 new data. By calculating the importance of the variables on the validation set, we can assess the  
 257 importance of the variables in the ability of the model to generalize and predict new data. The results  
 258 obtained correspond to the average of 100 simulations of Permutation feature importance for each  
 259 variable.

## 260 2.5. Empirical modelling of $^{137}\text{Cs}$

261 In the Rhône River, recent work was conducted to model the solid/liquid fractionation of  $^{137}\text{Cs}$  as a  
 262 function of the hydro-sedimentary conditions (water discharge, suspended sediment concentration  
 263 and particle size), the radioactive liquid discharges and the mean  $^{137}\text{Cs}$  concentrations of the superficial  
 264 soils of the watershed (Tomczak et al., 2021). With this approach, hourly  $^{137}\text{Cs}$  concentrations in  
 265 suspended sediments were modeled in the Rhône River at the SORA station using the following  
 266 equations:

$$267 \quad [^{137}\text{Cs}]_{SS} = \left( Kd_{\delta} \cdot [^{137}\text{Cs}]_w \cdot [SS]_R + [^{137}\text{Cs}]_{SOIL} \cdot [SS]_{NR} \right) / [SS] \quad \text{Eq. 3}$$

268 With:

- 269 •  $Kd_{\delta}$  ( $\text{l}\cdot\text{kg}^{-1}$ ), the equilibrium solid/liquid ratio of the superficial reactive layer of particles of  
 270 thickness  $\delta$  ( $\mu\text{m}$ ).
- 271 •  $[^{137}\text{Cs}]_{SOIL}$  ( $\text{Bq}\cdot\text{kg}^{-1}$ ), the mean  $^{137}\text{Cs}$  concentration of the superficial soils of the catchment.
- 272 •  $[SS]_R$  ( $\text{mg}\cdot\text{l}^{-1}$ ), the reactive fraction of suspended sediments given as a function of  $d_{50}$  ( $\mu\text{m}$ ), the  
 273 median of their granulometric distribution:

$$274 \quad [SS]_R = [SS] \cdot (1 - (1 - 2 \cdot \delta / d_{50})^3) \text{ if } d_{50} > 2 \cdot \delta \text{ and } [SS]_R = [SS] \text{ if } d_{50} \leq 2 \cdot \delta \quad \text{Eq. 4}$$

- 275 •  $[SS]_{NR}$  ( $\text{mg}\cdot\text{l}^{-1}$ ), the non-reactive fraction of suspended sediments:  $[SS]_{NR} = [SS] - [SS]_R$

- 276 •  $[^{137}\text{Cs}]_w$  ( $\text{Bq}\cdot\text{l}^{-1}$ ), the dissolved concentration of  $^{137}\text{Cs}$ :

$$277 \quad [^{137}\text{Cs}]_w = [^{137}\text{Cs}]_E / (1 + Kd_{\delta} \cdot ([SS]_c + [SS]_R)) \quad \text{Eq. 5}$$

- 278 •  $[SS]_c$  ( $\text{mg}\cdot\text{l}^{-1}$ ), the colloidal fraction of suspended sediments.

279 •  $[^{137}\text{Cs}]_E = \frac{q_{\text{release}}}{Q} + [^{137}\text{Cs}]_{\text{SOIL}} \cdot [\text{SS}]_R$  (Bq.l<sup>-1</sup>), the total exchangeable fraction of the <sup>137</sup>Cs,  
 280 where  $Q$  (m<sup>3</sup>.s<sup>-1</sup>) is the waterflow of the river and  $q_{\text{release}}$  (Bq.s<sup>-1</sup>) is the flux of <sup>137</sup>Cs releases by the  
 281 routine discharges.

282 To compare this model approach with the measures and the other approaches, the calculated hourly  
 283 series is integrated over the sampling period of average activity was calculated for each SS sampling  
 284 period.

### 285 3. Results

#### 286 3.1. Parameters

287 The optimized parameters used for the Random Forest and the HRHN approaches are detailed in the  
 288 Table 2. Before making a prediction, the HRHN model receives as input a batch of data corresponding  
 289 to a small history. According to the optimization, the hyperparameter of the table indicates a sequence  
 290 length of data history equal to 16: the first 16 values from the input time series and the target time  
 291 series (<sup>137</sup>Cs concentration) are therefore used to predict the future <sup>137</sup>Cs concentration (i.e. the 17<sup>th</sup>  
 292 value of the target series). This 17<sup>th</sup> value will then be included in the next history of the target variable  
 293 in the next prediction. The process is repeated until the last time in the series is reached.

294 *Table 2: Values of the hyperparameters of random forest and HRHN*

Random Forest		HRHN	
Parameter	Value	Parameter	Value
n_estimators	146	dim_RHN	8
max_depth	13	layer_RHN	2
min_sample_leaf	1	dim_max_pooling	[3,4,2]
min_sample_split	3	dim_filter_cnn	[7,5,5]
		nbr_filter_cnn	[8,32,16]

		sequence_length of data history	16
--	--	------------------------------------	----

295

### 296 3.2. Modeling

297 The modeling of particulate <sup>137</sup>Cs in the SS in the Rhône River at Arles is presented in the Figure 4. For  
 298 the different approaches, the modelled concentrations fall within the range of the measurements with  
 299 the exception of the empirical approach that overestimates the concentration in the early dataset (Fig.  
 300 4A).The approaches correctly follow the trends with lower concentrations during floods and higher  
 301 concentrations during baseflow and low-level water but the RF and empirical approaches tend to  
 302 underestimate the concentration during low-level water (Fig. 4B) while the HRHN correctly estimate  
 303 the concentration for the whole range of hydrological data. The difference between measured and  
 304 modelled values is lower for the HRHN and RF\_CV approaches with values of RMSE lower than 4 Bq  
 305 kg<sup>-1</sup> (Table 3) and R2 higher than 0.5.

306 *Table 3: Prediction indicators for the different approaches.*

	Empirical	RF_TS	RF_CV	HRHN
RMSE Train (Bq kg <sup>-1</sup> )	5.74	0.96	3.54 ± 2.7	0.69
RMSE Test (Bq kg <sup>-1</sup> )		6.52		3.59
R2 Train	0.10	0.96	0.59 ± 0.31	0.98
R2 Test		0.41		0.71

### 307 3.3. Sensitivity

308 The sensitivity analysis on the input variables shows contrasted results between the HRHN approach  
 309 and the two others (Fig. 5). For HRHN, SS fluxes of the Durance River and at Arles had a sensitivity  
 310 higher than 25% while their sensitivity was lower than 5% for RF\_TS. For RF\_CV, FSS Durance only was

311 negligible. For both RF approaches, the water discharge at Arles (Q\_Arles) and from the Saône River  
312 (Q\_Saone) had a sensitivity higher than 20%. For RF\_TS, the water discharge from the upper Rhône  
313 River at Jons (Q\_Jons) had a sensitivity higher than 10%. Finally, nuclear release had a sensitivity lower  
314 than 5% for the different approaches.

### 315 3.4. Fluxes

316 For the periods with measurement of  $^{137}\text{Cs}$  in SS, fluxes of  $^{137}\text{Cs}$  were calculated for the three modelling  
317 approaches and compared to the measured flux (Fig. 6A). For HRHN, the data history (the sixteen first  
318 values of train and test sub datasets – Table 1) represented around 35% of the annual flux of 2010 and  
319 2016 so the estimated fluxes are at two third recalculated by this approach in 2010 and 2016. Overall,  
320 the different approaches had annual fluxes close to the measured, with slight overestimation in 2010,  
321 2013 2016 and 2017. The average absolute differences for the test sub-datasets are respectively  $36.0$   
322  $\pm 25.0\%$ ,  $46.7 \pm 22.4\%$ ,  $21.7 \pm 19.3\%$  and  $30.6 \pm 24.7\%$  for HRHN (excluding 2016 due to the data history  
323 used), RF\_TS, RF\_CV and empirical approaches. Finally, flux of 2015 was reconstructed by the three  
324 approaches and the result is similar with a flux of  $^{137}\text{Cs}$  estimated between 20.0 to 24.5 GBq (Fig. 6B).

## 325 4. Discussion

326 The two data-driven approaches (HRHN and RF) achieved good prediction scores using only  
327 hydrological data (water discharge et SPM fluxes) and industrial input of  $^{137}\text{Cs}$ . These results show that  
328 the use of the main Rhône River tributaries hydrology alone is sufficient to obtain good results, without  
329 any knowledge of the actual input of  $^{137}\text{Cs}$  through erosion from the catchment (i.e. soil affected by  
330 the global fallout and the Chernobyl accident). Furthermore, all these input variables do not seem to  
331 be necessary as shown in the sensitivity analysis. Indeed, industrial releases represent only 5% of the  
332 sensitivity analysis (Fig. 5). These results show that near the mouth of the Rhône River, it is rather the  
333 hydrology (and the  $^{137}\text{Cs}$  concentration in eroded soils) that governs the concentrations of particulate  
334  $^{137}\text{Cs}$  than the industrial releases. However, a bias is possible as there are very few events with high  
335 concentrations of  $^{137}\text{Cs}$  (only 4 on 269 values over  $25 \text{ Bq kg}^{-1}$  – Fig. 2), events related to the combination

336 of nuclear release with low water discharge ( $Q_{\text{events}} = 717 \pm 98 \text{ m}^3 \text{ s}^{-1}$  vs  $Q_{\text{mean}} = 2586 \text{ m}^3 \text{ s}^{-1}$ ) resulting in  
337 a low dilution of the concentration released. So, the weight of the releases could be higher if the  
338 representation of these events was larger in the dataset and the score improved for all the models as  
339 they underestimate the concentration during low-level water (Fig. 4B). This confirms the interest of  
340 regularly updating the parameterization of the models with larger datasets. Also, the sensitivity  
341 analysis shows that some tributaries have more weight than others: the Durance tributary for HRHN  
342 and the Saône tributary for RF. The weight of the Durance River is logical as this tributary is  
343 characterized by intense floods which generate very important sediment flux (Bodereau et al., 2022;  
344 Delile et al., 2020; Poulhier et al., 2019) for a relatively low water discharge compared to the other  
345 tributaries (Fig. 7A). This result confirms the necessity to not only use the water discharge but also the  
346 flux of SS as the relation between the two parameters is not linear (Fig. 7B). For the RF approaches,  
347 the negligibility of the Durance tributary and the weight of the Saône tributary illustrate the difficulty  
348 in the choice of input variables when they are correlated (Gregorutti et al., 2017) which is the case of  
349 the hydrological data of this study. For these approaches, the weight of the hydrology at the  
350 monitoring station (Arles) is high and might also explain the low weight of the Durance tributary due  
351 to the proximity of this tributary (Fig. 1). Therefore, the models could be improved by removing the  
352 insignificant variables that indicate redundancy of information in the algorithm.

353 Regarding the score between the different models, the data-driven approaches have a better  
354 prediction of  $^{137}\text{Cs}$  than the empirical approach and among the data-driven approaches, the most  
355 complex one (HRHN) displayed better prediction. However, these machine learning and deep learning  
356 approaches proposed in this article are very different. First, Random Forest (RF) is a machine learning  
357 method that will make a prediction based on a set of regression trees, each having learned its tree  
358 structure from a sample of the training base. Therefore, its operation will be sensitive to the training  
359 frame and to a small number of extreme values (peaks in the data). This sensitivity explains the better  
360 scores obtained after the cross validation splitting method (RF\_CV) than with temporal splitting  
361 (RF\_TS) as the CV splitting considered a larger number of extreme events. In fact, the FSS\_Arles and

362 FSS\_Durance variables used for RF\_TS training are not representative of the full dataset (Table 1),  
363 which explains the low sensitivity scores for these variables with this approach. Thus, a number of  
364 nodes and leaves will not be present, making it difficult to predict particular cases. However, RF is  
365 distinguished by the very intuitive understanding of its algorithm and will find the average trends.  
366 Furthermore, its low number of hyperparameters makes the algorithm quickly applicable, so it can be  
367 considered to increase its learning base in order to increase its search area. Score of the random forest  
368 could also be improved by considering a causal techniques as the algorithm causal forest (Shuryak,  
369 2023). Second, the HRHN approach aims to understand the dynamics of the target variable:  
370 understanding past events in order to predict future behavior. This understanding is achieved through  
371 the different layers of HRHN, by detecting patterns in the time series (seasonality, peak...) through  
372 convolutional layers and by trying to capture the temporal dynamics through recurrent layers. During  
373 training, the network will parameterize itself (weighting) to stimulate the most important information  
374 in the network to estimate the future prediction. The model will therefore a priori have a greater  
375 robustness in the most extreme cases, which explain why the results are better than RF\_TS despite the  
376 non-representativeness of the FSS\_Arles and FSS\_Durance variables in the training set (Table 1).  
377 However, its architecture is associated with a large number of hyperparameters and parameters which  
378 makes its optimization phase expensive in terms of computation time. Therefore, the interpretability  
379 of the model is made complex. Although basic sensitivity analysis methods can be implemented to  
380 provide a measure of the relative importance of input characteristics, it will be necessary to develop  
381 more suitable algorithms to capture the full richness of the model. It should be noted here that the  
382 impact of the history of the target variable has not been studied. Even more, prediction with HRHN  
383 requires knowledge of a time history which limits its application to a framework where past data is  
384 available. Finally, regarding the particulate  $^{137}\text{Cs}$  annual fluxes, the different approaches can be used  
385 to predict it as the differences are generally low, especially in 2015 (Fig. 7). At the outlet of the Rhône  
386 River, the flux of  $^{137}\text{Cs}$  is mostly related to the flux of SS than the concentration of  $^{137}\text{Cs}$ . In fact, the



387  $^{137}\text{Cs}$  concentration varies by one order of magnitude while the FSS\_Arles varies by five orders of  
388 magnitude (Table 1).

389 To conclude, while the data-driven models can estimate concentrations in the perimeter of this study  
390 (i.e. normal condition of nuclear releases) without the need of the  $^{137}\text{Cs}$  released by nuclear industries  
391 (as lower than 5% of the sensitivity analysis), the empirical model needs this information which is  
392 difficult to obtain. This limitation shows that an AI approach seems to be easier to apply on this  
393 catchment to predict particulate  $^{137}\text{Cs}$  in normal condition and that this could be a useful approach for  
394 interpreting and supplementing routine monitoring datasets. In case of accidental releases (e.g.  
395 concentration several orders of magnitude above those in this study), the AI models will struggle to  
396 predict the right concentrations if their training dataset does not incorporate such situations and  
397 empirical approaches would then be more useful. However, in this example, the AI approach would  
398 reveal an anomaly between the prediction and the measurement, and therefore allow further work to  
399 be undertaken to understand the anomaly. Moreover, the AI approach might be used to estimate the  
400 quantity of  $^{137}\text{Cs}$  released using the  $^{137}\text{Cs}$  concentration in SS as input data. Going further, whatever  
401 the sediment-bound contaminant of interest, AI seems to be a good tool for predicting concentrations  
402 or sources as long as any of this information is available.

## 403 5. Conclusion

404 Concentrations of  $^{137}\text{Cs}$  in suspended sediment near the mouth of the Rhône River were estimated  
405 using three data-driven models and an empirical approach. Thanks to the long-term monitoring of this  
406 radionuclide, the data-driven models were able to accurately predict its concentration with better  
407 scores than the empirical approach. These results demonstrate that the use of a basic data-driven  
408 model (random forest) allow to obtain a better prediction of this radionuclide with a few diversities of  
409 input variables: water discharge and suspended sediment flux, and  $^{137}\text{Cs}$  release from nuclear facilities.  
410 In order to improve the prediction, a more complex model was used (HRHN) with an architecture that  
411 permit to understand the temporality of the  $^{137}\text{Cs}$  concentration. In summary, these methods offer

412 different means of prediction with greater or lesser advantages. It will then be up to the user to choose  
413 the most suitable model according to the objectives and the data available.

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## 424 Author contributions.

425 HL conceptualized the study; HL curated the data; VNG, KP and PB performed the modelling; HL  
426 acquired the funds; all authors developed the methodology; HL performed the visualization; all authors  
427 contributed to the original draft of the paper.

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585

Figure 1: Location of the monitoring stations and the nuclear facilities within the French part of the Rhône River downstream Lake Geneva and inventory of  $^{137}\text{Cs}$  estimated at the date of may 1986 (Roussel-Debel et al., 2007).

Figure 2: Concentration of particulate  $^{137}\text{Cs}$  measured at Arles during the studied period.

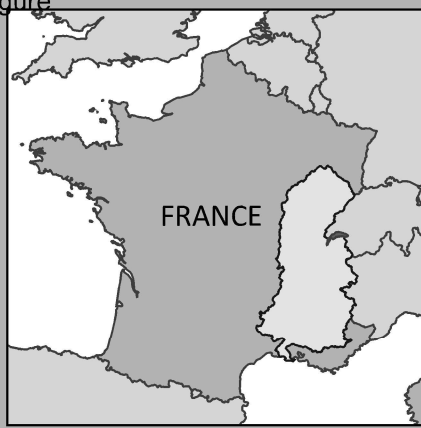
Figure 3: (A) Mean hourly water discharge and (B) suspended sediment concentration at the monitored stations during the studied period.

Figure 4: (A) measured (black) and modeled (grey) concentration of particulate  $^{137}\text{Cs}$  and (B) relation between measure and modelling on the test sub-datasets. For the HRHN approach, the 16th firsts values of the train (2010 – 2014) and the test (2015 – 2019) datasets were used as historic in the input dataset and not reported on the B graphic.

Figure 5: Sensitivity (%) of the input variables for the different approaches.

Figure 6: (A) measured and modelled annual flux of  $^{137}\text{Cs}$  including only periods with suspended sediment sampling and (B) estimation of the total flux in 2015 with completion of the periods without sampling by the four models.

Figure 7: (A) mean annual flux of water and suspended sediment for the 5 stations and (B) their relation between the water discharge and the suspended sediment flux.



## Rhône River bassin

- Rhône River
- Main tributary
- Secondary tributary

## Monitoring stations

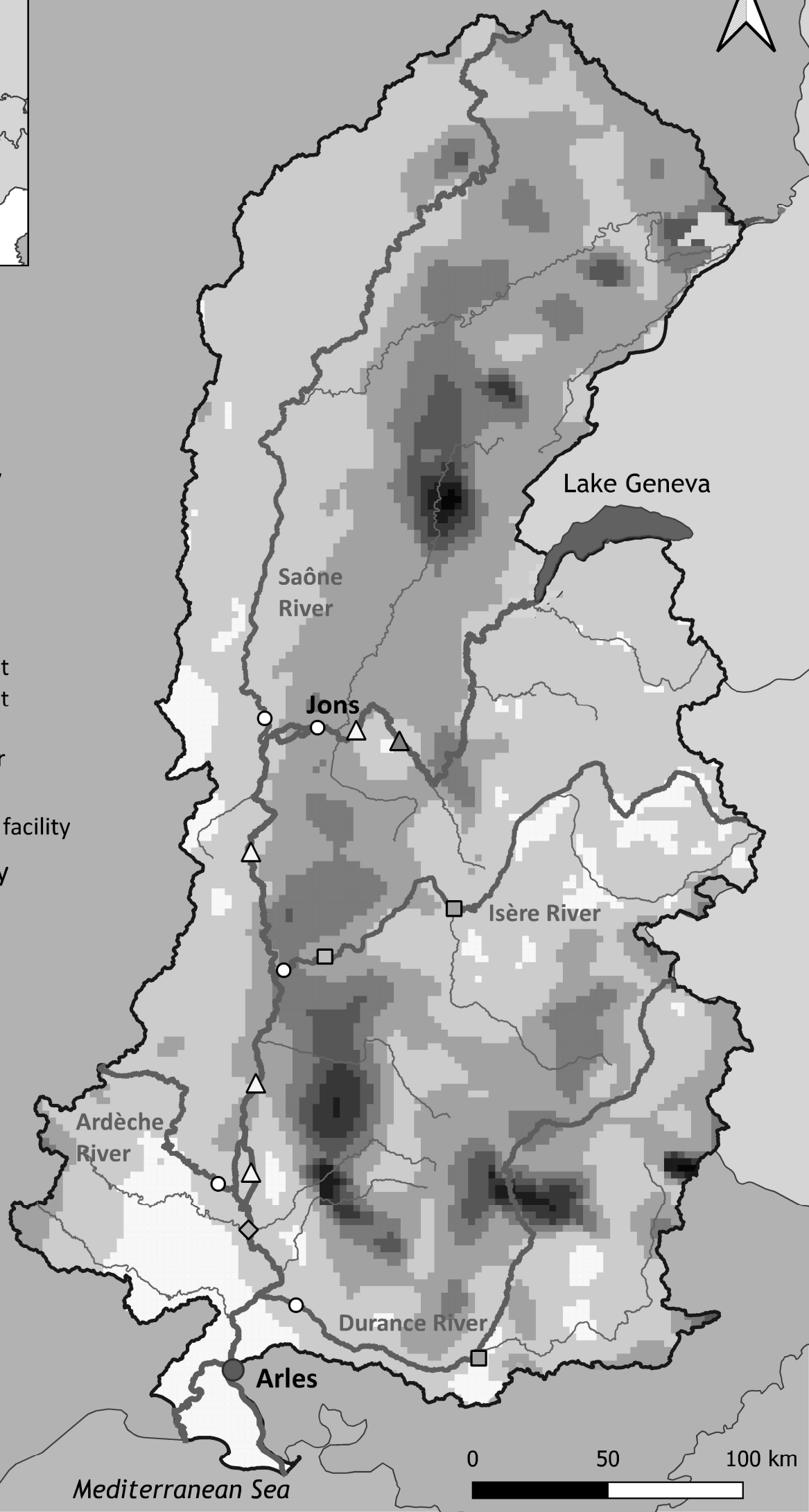
- Main
- Other

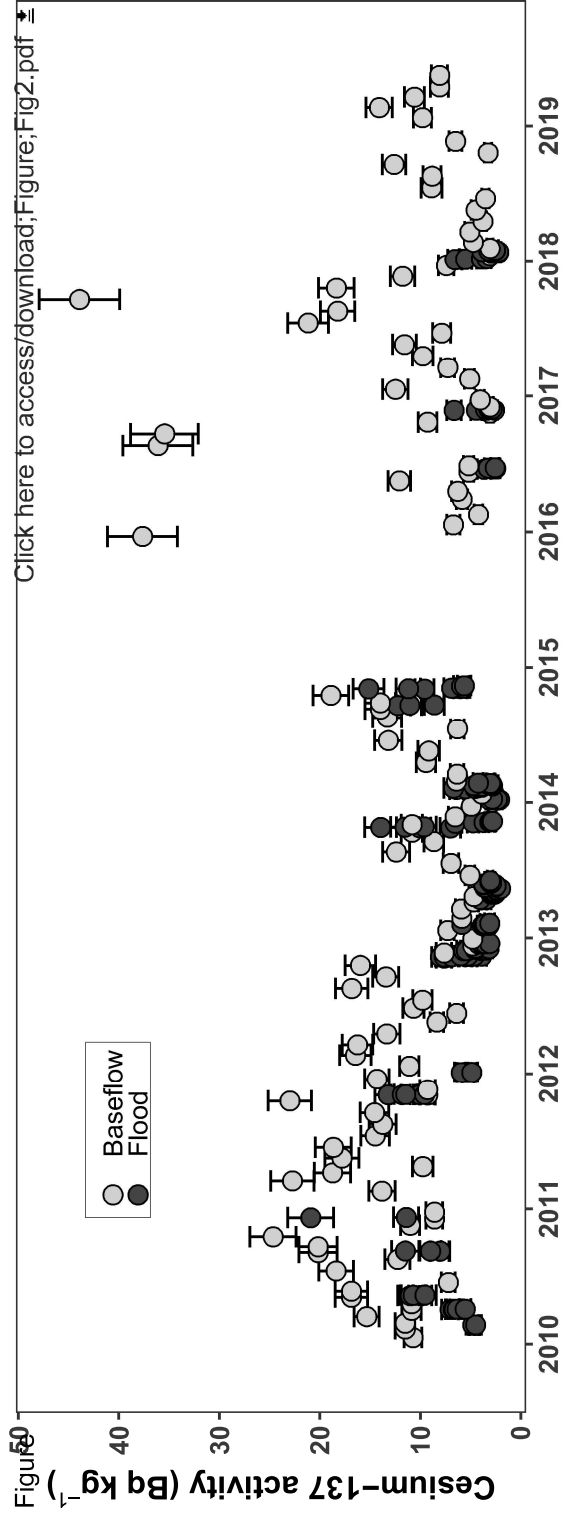
## Nuclear facilities

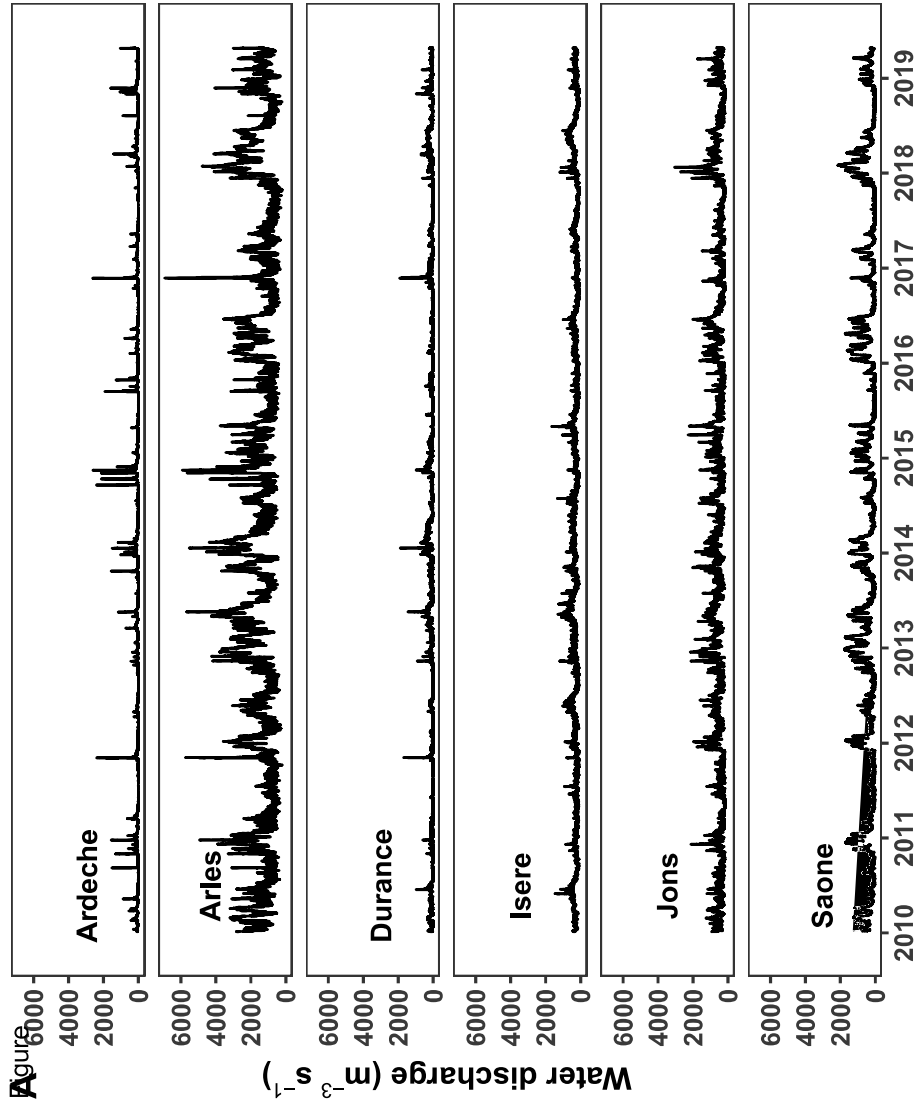
- △ Nuclear power plant
- ▲ Nuclear power plant in dismantelment
- ◇ Reprocessing center
- Research center
- Other nuclear cycle facility

Cesium-137 inventory in 1986 ( $\text{Bq m}^2$ )

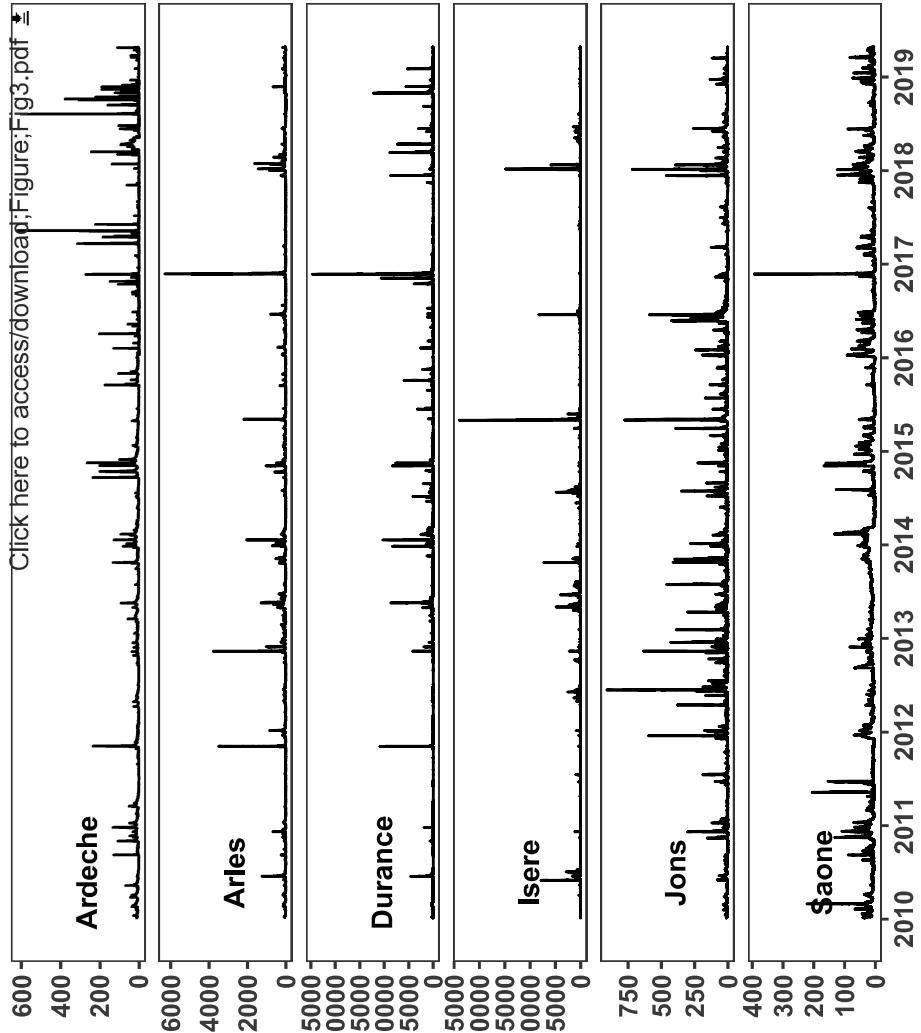
- <5k
- 5-10k
- 10-15k
- 15-20k
- 20-25k
- 25-30k
- 30-35k
- >35k



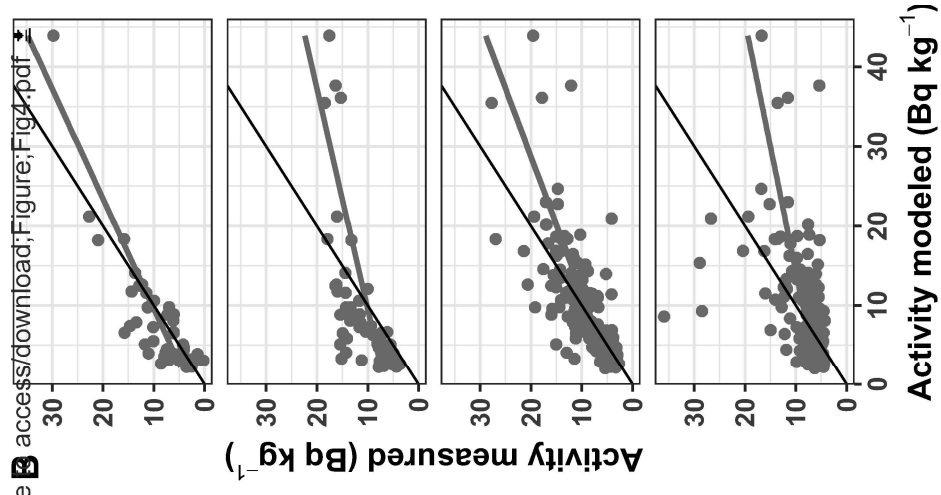
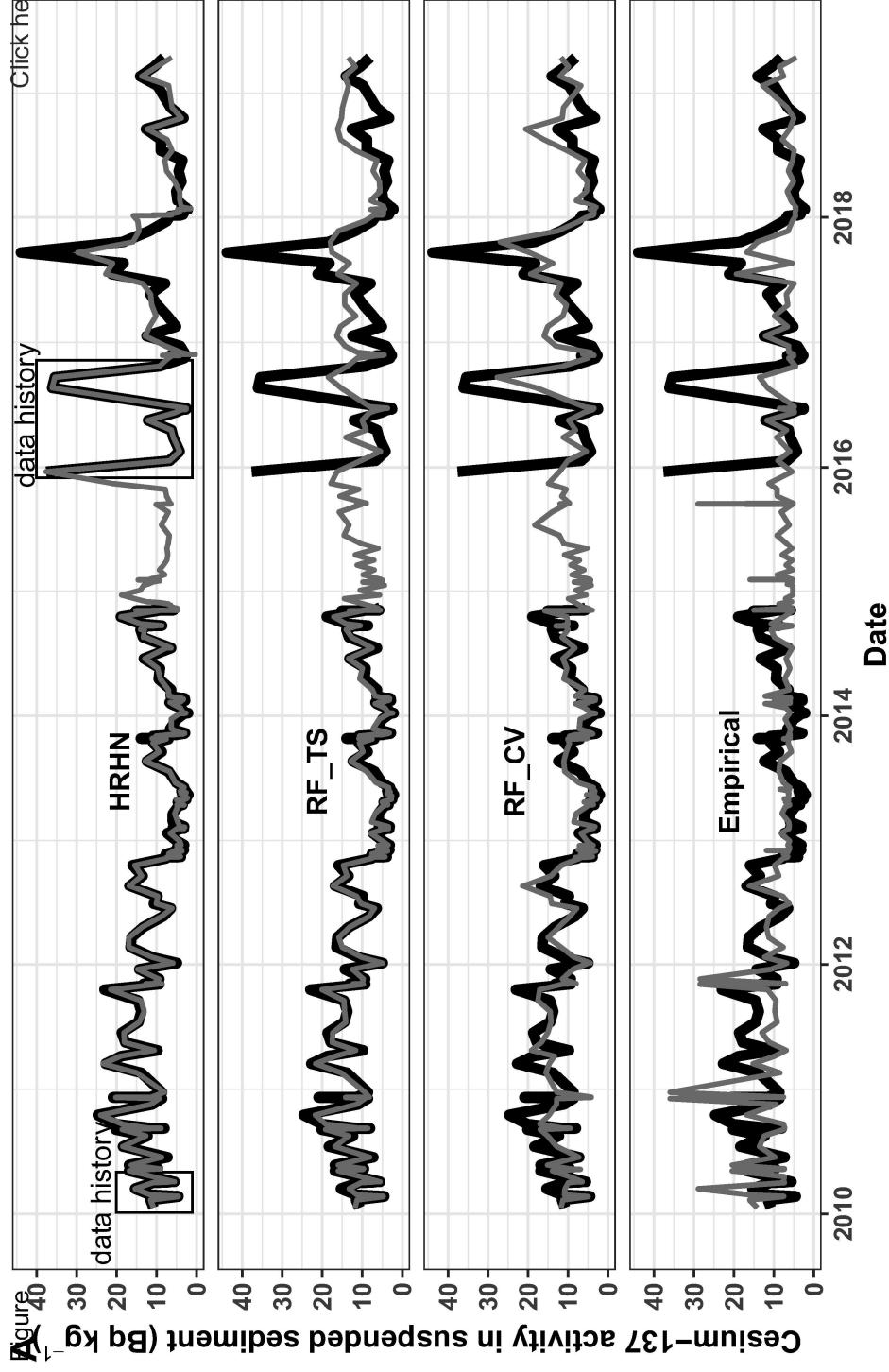




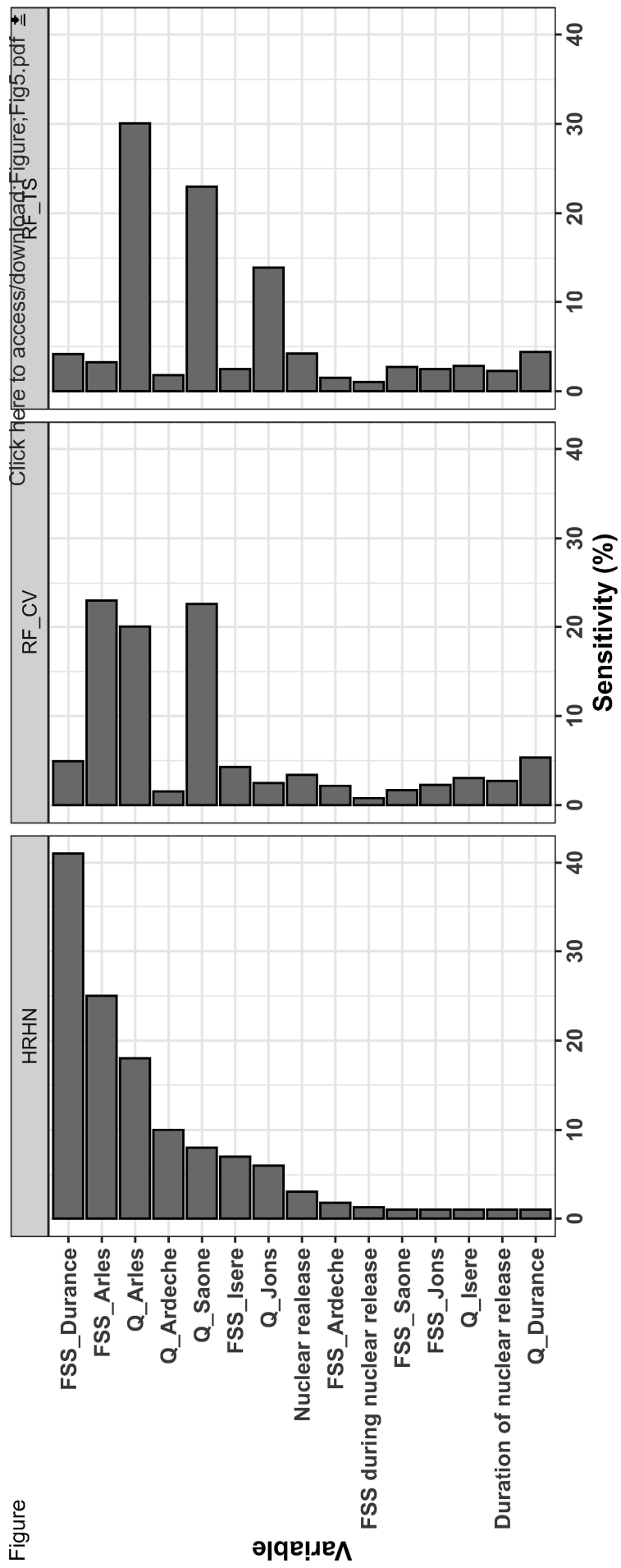
**B** Suspended sediment concentration ( $\text{mg L}^{-1}$ )



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Figure



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