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1 **Meta-modeling methods for estimating ammonia volatilization from**
2 **nitrogen fertilizer and manure applications**

3

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19

20 **Abstract**

21 Accurate estimations of ammonia (NH₃) emissions due to nitrogen (N) fertilization are
22 required to identify efficient mitigation techniques and improve agricultural practices. Process-based
23 models such as Volt'Air can be used for this purpose because they incorporate the effects of several
24 key factors influencing NH₃ volatilization at fine spatio-temporal resolutions. However, these
25 models require a large number of input variables and their implementation on a large scale requires
26 long computation times that may restrict their use by public environmental agencies. In this study,
27 we assess the capabilities of various types of meta-models to emulate the complex process-based
28 Volt'Air for estimating NH₃ emission rates from N fertilizer and manure applications. Meta-models
29 were developed for three types of fertilizer (N solution, cattle farmyard manure, and pig slurry) for
30 four major agricultural French regions (Bretagne, Champagne-Ardenne, Ile-de-France, and Rhône-
31 Alpes) and at the national (France) scale. The meta-models were developed from 106,092 NH₃
32 emissions simulated by Volt'Air in France. Their performances were evaluated by cross-validation,
33 and the meta-models providing the best approximation of the original model were selected. The
34 results showed that random forest and ordinary linear regression models were more accurate than
35 generalized additive models, partial least squares regressions, and least absolute shrinkage and
36 selection operator regressions. Better approximations of Volt'Air simulations were obtained for
37 cattle farmyard manure (3% < relative root mean square error of prediction (RRMSEP) < 8 %) than
38 for pig slurry (17% < RRMSEP < 19%) and N solution (21% < RRMSEP < 40%). The selected
39 meta-models included between 6 and 15 input variables related to weather conditions, soil properties
40 and cultural practices. Because of their simplicity and their short computation time, our meta-models
41 offer a promising alternative to process-based models for NH₃ emission inventories at both regional
42 and national scales. Our approach could be implemented to emulate other process-based models in
43 other countries.

44 **Keywords:**

45 Meta-modeling, Volt' Air, ammonia emission, fertilization, process-based model.

46

47 **1. INTRODUCTION**

48 In France, 98% of ammonia (NH₃) is emitted by the agricultural sector of which 35% is due
49 to the application of nitrogen (N) fertilization and 65% to manure management including manure
50 spreading (CITEPA, 2017a). A better estimation of the quantities of NH₃ emitted is necessary to
51 define effective strategies for reducing emissions at national and regional levels, and mitigating the
52 potential negative impact of NH₃ on human health (Moldanová et al., 2011) and ecosystems
53 (Bobbink et al., 1998; IPCC, 2006). Accurate NH₃ volatilization estimations are also required for
54 improved fertilization management and better adjustment of N fertilization to soil and climate
55 characteristics (Dupas et al., 2015; Parnaudeau et al., 2012).

56 Currently, the French national inventories of NH₃ emissions use the default Tier 2 emission
57 factors (EF) proposed by the EMEP Guidebook (CITEPA, 2017; EMEP/EEA, 2016a, b). Tier 2 EF
58 do not take into account national specificities such as agricultural practices that may differ between
59 countries. EF values were estimated from an international literature review of experimental
60 measurements of volatilization collected in different conditions (EMEP/EEA, 2016a, b) and with
61 various measurement methods (Sintermann et al., 2012). Process-based models represent an
62 interesting alternative by describing the effect of different factors on NH₃ volatilization. They can be
63 used to calculate EF for different regions with different pedo-climatic conditions and various types
64 of fertilizer, as shown by Ramanantenasoa et al. (2018). In France, Volt'Air was developed to
65 predict NH₃ volatilization after N fertilizer and manure applications. This model incorporates the
66 effect of the main agronomic, soil and meteorological factors influencing NH₃ volatilization in
67 agricultural fields (EMEP/EEA, 2016a, b; Garcia et al., 2012; Générumont and Cellier, 1997;

68 Huijsmans et al., 2014; Le Cadre, 2004; Smith et al., 2009; Sommer et al., 2003). However, Volt'Air
69 includes a rather large number of input variables and its implementation on a large scale requires
70 long computation times that may restrict its use by public environmental agencies.

71 In this study, we emulate the Volt'Air process-based model using simple statistical meta-
72 models. A meta-model is a model of a model. It may be seen as a simplified version of the original
73 model. A meta-model may be run as a substitute for the original complex model when that model
74 cannot be conveniently used, such as when the number of inputs in the original model is high, when
75 these inputs cannot be easily measured, or when the computation time of the original model is too
76 high for some applications. Meta-models are currently used in various research areas, for example,
77 in hydrology (Razavi et al., 2012), ecology (Conti and O'Hagan, 2010) and agronomy (Makowski et
78 al., 2015; Britz and Leip, 2009). However, not many publications describe meta-models for
79 estimating emissions of gas produced by agricultural practices, such as nitrous oxide (N₂O)
80 emissions (Giltrap and Ausseil, 2016; Villa-Vialaneix et al., 2012) and CH₄ emissions (Katayanagi
81 et al., 2016). Regarding NH₃ emissions, this study is the first attempt to develop meta-models
82 emulating an NH₃ volatilization process-based model as complex as Volt'Air.

83 Here we present an approach to producing meta-models used as surrogates of the Volt'Air
84 process-based model for estimating N emission rates after mineral fertilizer and manure applications
85 in France. We develop and compare a large number of meta-models based on different statistical
86 methods and on different sets of input variables for three types of fertilizer (N solution, pig slurry
87 and cattle farmyard manure (FYM)) with average agronomic properties, at regional and national
88 scales. All developed meta-models are fitted to a set of NH₃ emissions simulated by the Volt'Air
89 process-based model. The meta-models are evaluated by cross-validation for their ability to
90 approximate the emissions simulated by Volt'Air, and the results are used to select parsimonious
91 meta-models providing a good approximation of Volt'Air.

92 **2. MATERIALS AND METHODS**

93 **2.1. The Volt'Air model**

94 Volt'Air is a process-based model predicting NH₃ emissions after N fertilization and manure
95 applications on bare soils. It explicitly describes the physical and chemical equilibria between the
96 various types of ammoniacal N (N as NH₄⁺ and NH₃) using Henry's law for air/water equilibrium,
97 the acid-base equilibrium for ionic dissociation and the Freundlich adsorption equation for soil
98 minerals and organic matter. This model takes into account the effect of agricultural, soil and
99 meteorological factors on NH₃ volatilization (Garcia et al., 2011; Garcia et al., 2012; Générumont and
100 Cellier, 1997; Voylokov et al., 2014; Le Cadre, 2004). It simulates NH₃ emissions following
101 applications of organic manures, such as typical slurries and farmyard manure, and mineral
102 fertilizers, such as ammonium nitrate and urea in granular form as well as N solutions. Volt'Air runs
103 at an hourly time step and at field scale for a period of several weeks, i.e., covering the duration of
104 the volatilization event after fertilizer application. A more detailed description of the model may be
105 found in Générumont and Cellier (1997) and Garcia et al. (2012).

106 Here we consider a single model output: the N emission rate over 30 days after fertilizer
107 application, i.e., the cumulative N emission over 30 days after fertilizer application divided by N
108 fertilizer dose. In most cases, volatilization simulated using Volt'Air is complete 30 days after the
109 date of fertilizer application, even for mineral fertilizers showing longer volatilization periods
110 compared to organic manure (Theobald et al., 2014). Volt'Air can be run for real or virtual fields,
111 with an average computation time of 32 s per simulation using a Dell precision T7910 workstation
112 with two Intel Xeon 2.4 GHz processors (20 cores) and 128 GB RAM.

113 **2.2. Simulated datasets used to develop the meta-models**

114 The dataset used to develop the meta-models includes 106,092 NH₃ emission simulations
115 performed within the CADASTRE_NH₃ framework using the Volt'Air model (Ramanantenasoa et

116 al., 2018). These simulations encompass three types of fertilizer, five geographical areas, and
117 weather conditions corresponding to three contrasted crop years.

118 Simulation units of CADASTRE_NH₃ are the Small Agricultural Areas (SAR): a SAR is
119 considered to be geographical entity with homogeneous characteristics of agricultural activities,
120 weather and soil conditions (<http://agreste.agriculture.gouv.fr>). SAR sizes range from 1,096 to
121 440,650 ha, and the number of SAR per region varies from 6 to 74. Within our dataset, each SAR is
122 characterized each year by the area of the cultivated crop species, the weather conditions, and soil
123 types. Fertilization management is characterized at the regional scale.

124 Areas of cultivated crop species were derived from the European Land Parcel Identification
125 System for France (Martin et al., 2014). Data on weather conditions were provided by the French
126 national meteorological service “Météo-France”. Data on the spatial distribution of soils were
127 derived from the European Soil DataBase (ESDB) (Panagos et al., 2012) and the soil properties
128 were derived from the Harmonized World Soil Database (FAO et al., 2012). Data on cultural
129 practices were derived from the survey conducted in 2005-06 by the *Service de la Statistique et de la*
130 *Prospective* of the French Ministry of Agriculture, for 10 main arable crops (soft wheat, durum
131 wheat, barley, grain maize, forage maize, oilseed rape, sunflower, peas, sugar beet and potato) and
132 two types of grasslands (grass leys and intensive permanent grasslands) in 21 French regions (Corse
133 region was excluded) (AGRESTE, 2006), following the methodology of Mignolet et al. (2007).
134 Fertilization was defined as a succession of fertilizations on a given crop species. Each fertilizer
135 application was characterized by the date of application, type of fertilizer (3 mineral fertilizers:
136 ammonium nitrate, N solution, urea; 5 organic manure: cattle farm yard manure (FYM), cattle
137 slurry, sheep FYM, pig slurry, and vinasse), and dose applied. For the simulations with Volt’Air,
138 fertilizers were assumed to be applied on the surface without abatement techniques, at a single time,
139 8:00 am UTC.

140 Each simulation covered the cumulative NH₃ emissions over 30 days after a specific
141 fertilizer application on a specific SAR*crop*fertilization*year combination. A more detailed
142 description of the dataset and of the simulation procedure may be found in Ramanantenasoa et al.
143 (2018). Part of the dataset including the simulations of Volt'Air was made freely available in the
144 related data paper (Génermont et al., 2018).

145

146 Meta-models were developed separately for each type of fertilizer. The three selected
147 fertilizers (N solution, cattle FYM, and pig slurry) were among the most applied fertilizers in France
148 in 2005-06; N solution accounted for 38% of N applications from simple mineral N fertilizers
149 (UNIFA, 2015); cattle FYM and pig slurry accounted for 82% and 7% of N applications from
150 organic fertilizers, respectively (AGRESTE, 2006, 2010).

151 Meta-models were developed separately for the national and the regional scales. Four French
152 regions characterized by contrasting agricultural, climatic and soil conditions were selected; the
153 North-Western Bretagne region is characterized by a high use of organic manure due to the
154 dominance of livestock farming, an oceanic temperate climate, and acidic soils; the North-Eastern
155 Champagne-Ardenne region is a major arable crop farming area, encountering a semi-continental
156 temperate climate, with a dominance of alkaline soils; Ile-de-France, located in the North Center of
157 France, is an arable crop farming area, with soil and weather conditions in-between these two former
158 regions; the South-Eastern Rhône-Alpes region is a more agriculturally diverse region, with
159 southern semi-continental climatic conditions.

160 In order to capture the possible effect of weather variables between years, weather conditions
161 of two contrasting crop years were added: 2007-08 and 2010-11. Crop year 2005-06 was
162 characterized by a dry spring and a hot summer (AGRESTE, 2010); 2007-08 enjoyed favorable
163 conditions for crop growth, i.e., a humid spring with temperatures close to the seasonal average and
164 a hot summer with rainfall close to the seasonal average (JRC, 2008); 2010-11 was characterized by

165 a very dry, hot spring and a rainy summer (AGRESTE, 2014). The same sites and cultural practices
 166 were analyzed for the three years; only the weather conditions were changed.

167 The sizes of national and regional datasets per fertilizer type and per geographical area and
 168 the numbers of SAR per geographical area are given in Table 1. Note that no simulation was
 169 available for pig slurry in Champagne-Ardenne, Ile-de-France and Rhône-Alpes. This type of
 170 fertilizer is barely used in these three regions.

171 Table 1: Numbers of simulations of the Volt’Air model in the national and regional datasets and
 172 numbers of SARs per geographical area

Type of fertilizer		France	Bretagne	Champagne-Ardenne	Ile-de-France	Rhône-Alpes
N solution	Number of simulations	70,299	2,388	3,852	5,286	6,951
	Share of total (%)	100	3	5	8	10
Cattle FYM	Number of simulations	13,725	147	804	558	1,452
	Share of total (%)	100	1	6	4	11
Pig slurry	Number of simulations	405	225	0	0	0
	Share of total (%)	100	56	0	0	0
Total number of simulations		84,429	2,760	4,656	5,844	8,403
Total number of SARs		706	25	34	36	76

173 2.3. Inputs of the meta-models

174 A total of 16 original input variables were chosen from all the Volt’Air input variables for
 175 meta-modeling (Table 2) because they were expected to have an impact on NH₃ volatilization and
 176 showed a wide range of variation in the data sources. Some of these 16 inputs were not taken into
 177 account in a few fertilizer * region combinations considered. Thus, *Dose of application* was not used
 178 for cattle FYM and pig slurry because its range of variation for these two types of fertilizer was too
 179 narrow. *Bulk density* was not used for Bretagne, for the same reason. Note that variables describing
 180 fertilizer and manure properties were not considered here because specific meta-models were
 181 developed for each fertilizer separately.

182 The variables describing geographical location, soil and cultural practices were not time-
 183 dependent. In contrast, all variables describing weather conditions were defined for 23 different
 184 time-periods:

- 185 - 10 different daily periods; 1st (i.e., the day of fertilizer application), 2nd, 3rd, 4th, 5th, 6th, 7th,
 186 8th, 9th and 10th days following fertilizer application.
- 187 - 10 different cumulative periods; 2, 3, 4, 5, 6, 7, 8, 9, 20 and 30 days following fertilizer
 188 application.
- 189 - three ten-day periods; 1st, 2nd and 3rd, from the 1st to the 10th day, from the 11th to the 20th
 190 day, from the 21th to the 30th day respectively following fertilizer application,.

191 For each period, the values for *Mean air temperature*, *Relative air humidity*, *Incident solar*
 192 *radiation*, and *Wind speed* were averaged over the period considered. The values for *Minimum air*
 193 *temperature* and *Maximum air temperature* were set at the minimum and maximum air temperature
 194 over the period. The values for *Cumulative rainfall* were set at the sum of rainfall over the time-
 195 periods considered. A separate meta-model was constructed for each time-period.

196 Two additional non-weather related variables were considered to account for regional effects
 197 on volatilization; the categorical variable *Region* and the continuous variable *Longitude*. Note that
 198 the continuous variable *Latitude* is a Volt'Air variable. With these three variables, we were able to
 199 produce a unique meta-model per fertilizer type at the France scale while accounting for regional
 200 specificities.

201 Table 2: List of input variables of the meta-models

Category	Variable	Unit
Location of site	- Latitude	°
	- Longitude	°
	- Region ^(a)	Categorical variable
N fertilization management	- Date of N application	- Number of days from origin 01/01/2005
	- Dose of application ^(b)	- kg N.ha ⁻¹

Soil properties	- pH	
	- Bulk density	- kg.m ⁻³
	- Silt content	- g.kg ⁻¹ of dry soil
	- Clay content	- g.kg ⁻¹ of dry soil
	- Organic carbon content	- g.kg ⁻¹ of dry soil
	- Initial water content	- g.kg ⁻¹ of dry soil
Weather conditions	- Minimum air temperature	- K
	- Mean air temperature	- K
	- Maximum air temperature	- K
	- Specific air humidity	- kg.kg ⁻¹
	- Incident solar radiation	- W.m ⁻²
	- Wind speed	- m.s ⁻¹
	- Cumulative rainfall	- mm.s ⁻¹ / 3600

202 (a) The *Region* variable was added for the development of meta-models at the national scale only.

203 (b) *Dose of application* was not used for cattle FYM and pig slurry meta-models.

204 (c) *Bulk density* was not used for Bretagne.

205

206 Boxplots of the 17 quantitative explanatory variables chosen are presented in Figure S2 in
 207 the supplementary material.

208

209 Additional input variables were defined from the original variables listed in Table 2 for
 210 potential inclusion in the meta-models; the quadratic terms of several variables (see Tables S1 and
 211 S2 in the supplementary material) and interaction terms between pairs of variables (i.e.,
 212 multiplication between two variables). Interactions were chosen with respect to formalisms chosen
 213 to describe the processes of Volt' Air. Interaction between *Cumulative rainfall* and *Air temperatures*
 214 (minimum, mean, maximum) (i.e., three pairs), interaction between *Cumulative rainfall* and *Wind*
 215 *speed*, and interaction between *Cumulative rainfall* and *Soil texture (Silt, Clay)* were chosen because
 216 infiltration depends on both the quantity of water and the state of the soil surface, which in turn
 217 depends on evaporation potential, driven by both temperature and wind-speed, and on soil texture
 218 (Garcia et al., 2012). The interaction between *Cumulative rainfall* and *Air temperatures* reflects the
 219 effect of these factors on physicochemical equilibria, through dilution by rainfall and the high

220 dependence of the equilibrium constants on temperature. For N solution and pig slurry, interactions
221 between *Soil pH* and *Air temperatures* (minimum, mean, maximum) were also considered. As those
222 products are incorporated to the first soil layer, soil pH together with N solution pH or pig slurry pH
223 directly drive physicochemical equilibria, which are also highly dependent on temperature.

224 **2.4. Types of meta-model**

225 For a given type of fertilizer, meta-models were fitted to each regional dataset of simulated
226 NH₃ emissions and then to the whole dataset at the national scale. Five different types of meta-
227 model were developed with each dataset. The first three types of model (ordinary linear regression,
228 Least Absolute Shrinkage and Selection Operator, and Partial Least Squares Regression) assumed a
229 linear relationship between N emission rate over 30 days (or its transformed-value) and input
230 variables. The last two types of model (Generalized additive model, Random Forest) dealt with
231 nonlinear relationships between volatilization rates and input variables. The inputs of generalized
232 additive model and random forest were selected using a regression method (ordinary linear
233 regression, Least Absolute Shrinkage and Selection Operator, or Partial Least Squares Regression,
234 see section 2.5 for details on the selection procedure). Below, we briefly present the main
235 characteristics of each type of model. Details on the meta-model selection procedure are given in
236 Section 0. The equations of the meta-models are described in Table 3 together with the R functions
237 used for their implementation. The R functions mentioned in Table 3 are freely available from
238 <https://cran.r-project.org>. They can be easily used to develop meta-models from the dataset
239 described in 2.2. The present study was performed with the 3.2.3 version of R.

240 Ordinary linear regression (LM, Linear Model) was used to relate emission rates (noted y in
241 Table 3) to quantitative and categorical input variables (noted X_1, \dots, X_p in Table 3) through a linear
242 equation. Here the flexibility of linear models was increased through the inclusion of quadratic
243 terms and interactions between inputs. Several variants of ordinary linear regression models were

244 developed. In some variants, model inputs were selected using a stepwise or a forward selection
245 procedure based on the Akaike Information Criterion (AIC) (Figure 1). All linear models were fitted
246 using Ordinary Least Squares (OLS) with the R function `lm()`.

247 Least Absolute Shrinkage and Selection Operator (LASSO) is a well-known regularization
248 technique that can be applied to reduce variance of the parameter estimators in linear regression
249 models (Tibshirani, 1996). LASSO is based on the same model equation as LM, i.e., a linear
250 equation relating the emissions rates to the input variables. However, instead of using OLS for
251 obtaining the parameter estimates, LASSO is subjected to constraints on parameters defined through
252 a penalty term based on the absolute values of the parameters. With this approach, some parameters
253 become equal to zero by tuning the penalty term for minimizing prediction errors, leading to the
254 selection of a reduced subset of input variables. LASSO was implemented here using the R function
255 `cv.glmnet()` of the package `glmnet`.

256 Partial Least Squares Regression (PLSR) can improve the accuracy of linear regression
257 models when the input variables are strongly correlated (Abdi, 2010). PLSR may be seen as an
258 improved version of principal component regression (PCR). In PCR, principal components are
259 expressed as linear combinations of original variables, such that the variability in the data may be
260 described by a small number of these new variables. In PLSR, the original input variables are also
261 replaced by linear combinations of inputs, but these combinations are selected to describe most of
262 the input data variability while showing a strong correlation to the response (Table 3). PLSR was
263 implemented here using the R function `plsr()` of the `pls` package.

264 Generalized Additive Models (GAM) assume that the response variable Y (emission rate)
265 can be expressed as a linear combination of several unknown smooth functions of the inputs (Wood,
266 2017). The equation used by GAM is more general than the linear equations used by LM, LASSO,
267 and PLSR and is expressed as $\mu + s(X_1) + \dots + s(X_P)$, where $s(\cdot)$ are several smoothing functions

268 (splines) of the inputs. Here, the volatilization rate is expressed as a linear combination of the
269 smoothing functions. We used smoothing splines as smoothing functions because these types of
270 function are very flexible. This model does not assume that the volatilization rates are linearly
271 related to the inputs and can describe complex response curves. As the model is expressed as a sum
272 of independent functions, the effect of each input on the output can be easily interpreted. GAMs
273 were fitted here using the R function `gam()` of the `mgcv` package.

274 Random Forest (RF) is a machine learning technique that builds an ensemble of
275 classification (or regression) trees (Breiman and Cutler, 2003). RF creates a set of binary decision
276 rules based on the input variables. The first step of the method is the random selection, with
277 replacement, of a sample of data (called the “in-bag”) of equal size to the total number of data. The
278 remaining part of the data is referred to as “out-of-bag” (OOB) data. The second step involves the
279 random selection of explanatory variables. The third step involves the building of a tree based on the
280 in-bag data and a subset of variables selected. The tree is built by recursively partitioning the initial
281 data into smaller groups, called nodes, through binary splits based on a single input variable.
282 Examples of regression trees are given in Figure S3 in the supplementary material. The whole
283 process is repeated several thousand times, to generate many trees. At each iteration, the OOB data
284 are used to evaluate the accuracy of the fitted trees. The output of RF is calculated by averaging the
285 outputs of the individual trees (Table 3). Here RF models were fitted with the R function
286 `randomForest()` of the `randomForest` package and the R function `ranger()` of the `ranger` package.
287 Ranger is a fast implementation of random forests (Breiman 2001) and was used here to fit the RF
288 meta-model for N solution in France to reduce computation time.

289 Table 3 : Description of the statistical and machine learning methods used to develop the meta-models of Volt'Air. The methods are
 290 based on different types of models whose parameters are estimated using an optimization algorithm implemented with R functions of specific R
 291 packages. Each meta-model computes NH3 volatilization rates y (or log/box-cox transformed volatilization rates) from a series of inputs X_1 ,
 292 X_2, \dots, X_p .

Acronym	Method	Equation	Optimization algorithm	R package	R function used for parameter estimation
LM	Ordinary linear regression model with stepwise selection	$y = \mu + \alpha_1 X_1 + \alpha_2 X_2 + \dots + \alpha_p X_p$ where X_1, X_2, \dots, X_p are p input variables, and $\mu, \alpha_1, \dots, \alpha_p$ are parameters estimated from the dataset of volatilization rates.	Least squares. Input variables are selected using a stepwise procedure based on the AIC criterion.	stats	lm(Volat_rate ~ X1+X2+...+Xp)
LASSO	Least Absolute Shrinkage and Selection Operator	$y = \mu + \alpha_1 X_1 + \alpha_2 X_2 + \dots + \alpha_p X_p$ where X_1, X_2, \dots, X_p are p input variables, and $\mu, \alpha_1, \dots, \alpha_p$ are parameters estimated from the dataset of volatilization rates.	Penalized least squares. A penalty term is used to select input variables. The optimal value of the penalty term is chosen by cross validation in order to minimize prediction errors.	glmnet	cv.glmnet(X, Volat_rate) where X is a matrix including the values of the inputs X_1, X_2, \dots, X_p .
PLSR	Partial Least Squares Regression	$Y=UQ'$ $X=TP'$ where X is an $n \times p$ matrix including the values of the p inputs for the n volatilization rate data. Y is a vector including the n volatilization rate data, T and U are projections of X and of Y, respectively. P	Partial least squares. The number of inputs (components) are selected by cross-validation.	pls	pls(Volat_rate ~ X, validation="CV") where X is a matrix including the values of the inputs X_1, X_2, \dots, X_p .

		and Q are orthogonal loading matrices. The decomposition of X and Y are made so as to maximize the covariance between U and T.			
GAM	Generalized additive model	$y = \mu + s(X_1) + s(X_2) + \dots + s(X_p)$ where the functions $s(\cdot)$ are smooth functions of the input variables (regression splines). Each smooth function defines the response of y to a given input.	Penalized maximum likelihood. Penalty terms are used to specify the smooth functions.	mgcv	gam(Volat_rate ~ s(X1)+ s(X2) + ...+s(Xp))
RF	Random Forest	$y = \frac{1}{Q} \sum_{q=1}^Q h(Xs_q)$ with $h(Xs_1) \dots h(Xs_Q)$ a collection of outputs of Q decision trees. Each tree consists in a series of partitions based on a subset of inputs Xs_q . Each subset of inputs includes variables chosen at random among X_1, X_2, \dots, X_p .	Mean squared error calculated using “out-of-bag” data (i.e., data not used for fitting the regression trees).	randomForest	randomForest(Volat_rate ~ X1+X2 + ...+Xp)

293

294

295 2.5. Selection of meta-models

296 For each type of fertilizer and each geographical area, 602 meta-models were developed and
297 assessed according to the four-step selection procedure presented in Figure 1 and detailed below.
298 Each model was defined by a specific statistical method (among LM, LASSO, PLSR, RF, GAM),
299 the type of response variable (untransformed or log/box-cox-transformed emission rate), the time-
300 period used to define the weather inputs, the inclusion or not of quadratic terms and interactions, and
301 the input selection method (none, stepwise, forward). The box-cox transformation has the form $w_i =$
302 y_i^λ , if $\lambda \neq 0$ and $w_i = \log(y_i)$, if $\lambda = 0$ with w_i the transformed variable, y_i the initial response
303 variable, and λ the parameter for the transformation (estimated from the data).

304 In Step 1, three types of linear model were fitted and compared by cross-validation: ordinary
305 linear models (LM), LASSO, and PLSR. Two sets of inputs were considered successively: the
306 inputs listed in Table 2, and an expanded set of inputs including quadratic terms and interactions.
307 With LM, input variables were selected using either a stepwise or a forward selection procedure
308 based on the AIC criterion (Akaike, 1974). The total number of models tested in Step 1 was 598.

309 In Step 2, the two best linear meta-models were selected using cross-validation. Cross-
310 validation is a standard method for evaluating the accuracy of the model predictions (Efron, 1983).
311 It allows one to estimate how the model is expected to perform when used to make predictions on
312 data independent from those used for estimating the model parameters. The general principle is to
313 split the dataset into K groups. The first group is removed from the dataset. The remaining part of
314 the dataset is used to fit the model. The fitted model is then used to predict the discarded group of
315 data. The whole procedure is repeated over all groups, and the accuracy of the prediction is
316 evaluated by computing the root mean square error of prediction (RMSEP) over the K groups. Here,
317 two procedures were used successively to define the groups of data, leading to two different types of
318 cross validation. The first type of cross-validation was implemented SAR by SAR in order to

319 estimate the root mean square error of prediction of each meta-model (RMSEP_SBS). RMSEP_SBS
320 measures the ability of each meta-model to approximate the emission rates simulated by Volt'Air in
321 a new SAR, i.e., a SAR not originally included in the dataset used to develop the meta-model
322 considered. The second type of cross-validation consisted of dividing the dataset by years. This is
323 also referred to as year-by-year cross-validation and the associated RMSEP is denoted
324 RMSEP_YBY. This value of RMSEP measures the ability of each meta-model to approximate the
325 emission rates simulated by Volt'Air for a new year, not originally included in the dataset used to
326 develop the meta-model.

327 In Step 3, two RF and two GAM meta-models were developed. They included the input
328 variables of the two best linear models selected in Step 2, i.e., the linear models characterized by the
329 lowest values of RMSEP_SBS and RMSEP_YBY (Linear MM_SAR and MM_YEAR). These
330 models were thus developed from the input variables selected in Step 2. The output and input
331 variables were used in their original form, i.e., without transformation or interaction.

332 In Step 4, cross-validations (SAR-by-SAR and year-by-year) were carried out again to select
333 the two best meta-models from the two best linear models, RF, and GAM. The two meta-models
334 selected at this step were denoted Final MM_SAR and MM_YEAR.

335 The whole procedure was applied for each fertilizer type, for each region and for France.
336 When two or more meta-models had similar RMSEPs (to two decimal places), we favored the meta-
337 model including the smallest number of inputs (inputs related to the geographical location of the
338 site, i.e., latitude, longitude and region, were not counted because they are always available at zero
339 cost). When two meta-models had similar RMSEPs and the same number of inputs, we chose the
340 meta-model with weather condition variables calculated over the longest time-period because these
341 variables were smoother than variables calculated over shorter time-periods, and were thus less
342 sensitive to extreme, short-term meteorological events.

STEP 1	LM1			LM2			LM1 stepwise selection			LM2 stepwise selection			LM1 forward selection			LM2 forward selection			LASSO1		LASSO2		PLSR1		PLSR2		Method
	none	log	box-cox	none	log	box-cox	none	log	box-cox	none	log	box-cox	none	log	box-cox	none	log	box-cox	none	none	none	log	box-cox	none	none	Output transformation	
	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	23	Number of tested time-periods ⁽¹⁾	
STEP 2	SELECTION OF THE TWO BEST LINEAR METAMODELS  Linear MM_SAR with the lowest RMSEP_SBS Linear MM_YEAR with the lowest RMSEP_YBY																										
STEP 3	RF			RF			GAM			GAM			Method														
	Inputs are those selected in the linear model with the lowest RMSEP_SBS			Inputs are those selected in the linear model with the lowest RMSEP_YBY			Inputs are those selected in the linear model with the lowest RMSEP_SBS			Inputs are those selected in the linear model with the lowest RMSEP_YBY			Inputs														
STEP 4	SELECTION OF THE FINAL TWO BEST METAMODELS AMONG THE TWO BEST LINEAR MODELS, THE TWO GAMs, AND THE TWO RFs  Final MM_SAR with the lowest RMSEP_SBS Final MM_YEAR with the lowest RMSEP_YBY																										

343 ⁽¹⁾ For each type of linear model, 23 variants were developed; each variant corresponds to a specific time-period.

344 Figure 1: Meta-modeling selection procedure. This four-step approach is applied for each type of fertilizer for each region and at the national scale. The
 345 selected models are those minimizing the root mean square error of prediction (RMSEP) computed by cross-validation. Two types of cross-validation are
 346 implemented, i.e., by removing each small agricultural region (SAR) in turn, or each year in turn. The two cross-validations lead to two different RMSEPs
 347 (RMSEP_SBS and RMSEP_YBY, respectively). Two sets of inputs were considered: Set 1 with the original variables, and Set 2 including in addition
 348 quadratic terms and interactions. LM1-LM2: ordinary linear models (with/without quadratic and interaction terms). LASSO1-LASSO2: LASSO regression
 349 models (with/without quadratic and interaction terms). PLSR1-PLSR2: PLSR regression models (with/without quadratic and interaction terms). RF: random
 350 forest. GAM: generalized additive models.

2.6. Comparison of meta-models at the regional scale

After the selection described in Figure 1, two types of meta-model were available for estimating emission rates in a given region: the regional meta-models derived from the set of simulated NH_3 emissions obtained for that region specifically, and the France meta-models derived from the whole set of simulated emissions at the national level (i.e., over the 21 French regions).

The meta-models selected at the national level can be used to estimate emission rates in a specific region because they include the categorical input *Region* and/or the quantitative inputs *Latitude* and *Longitude* accounting for a possible regional effect. So we compared regional and France meta-models for their ability to estimate emission rates for each selected type of fertilizer and each region. To do this, the best France meta-models were run on site*crop*fertilization*year of the regional datasets, and the resulting RMSEP values were compared with those calculated with the best regional meta-models. For each selected region and fertilizer type, the RMSEP of the regional and France meta-models were compared and the model producing the lowest value was finally selected.

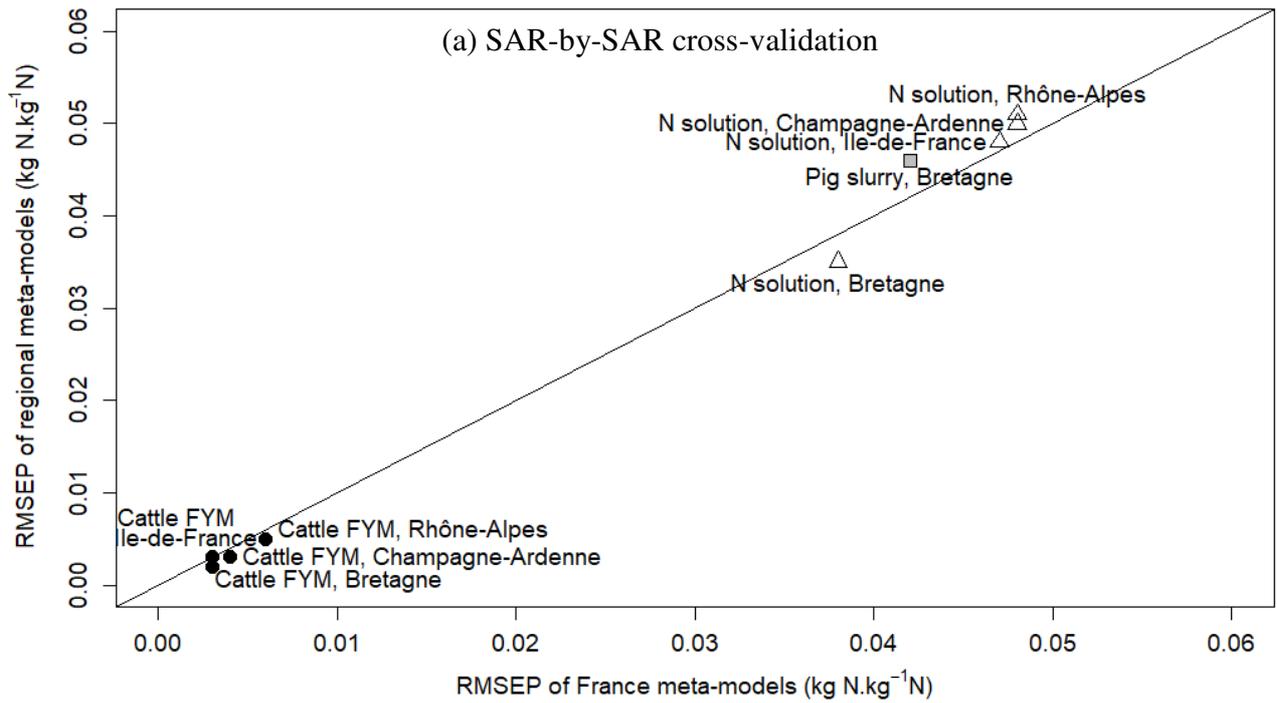
3. RESULTS

3.1. Comparison of Volt'Air and meta-model outputs

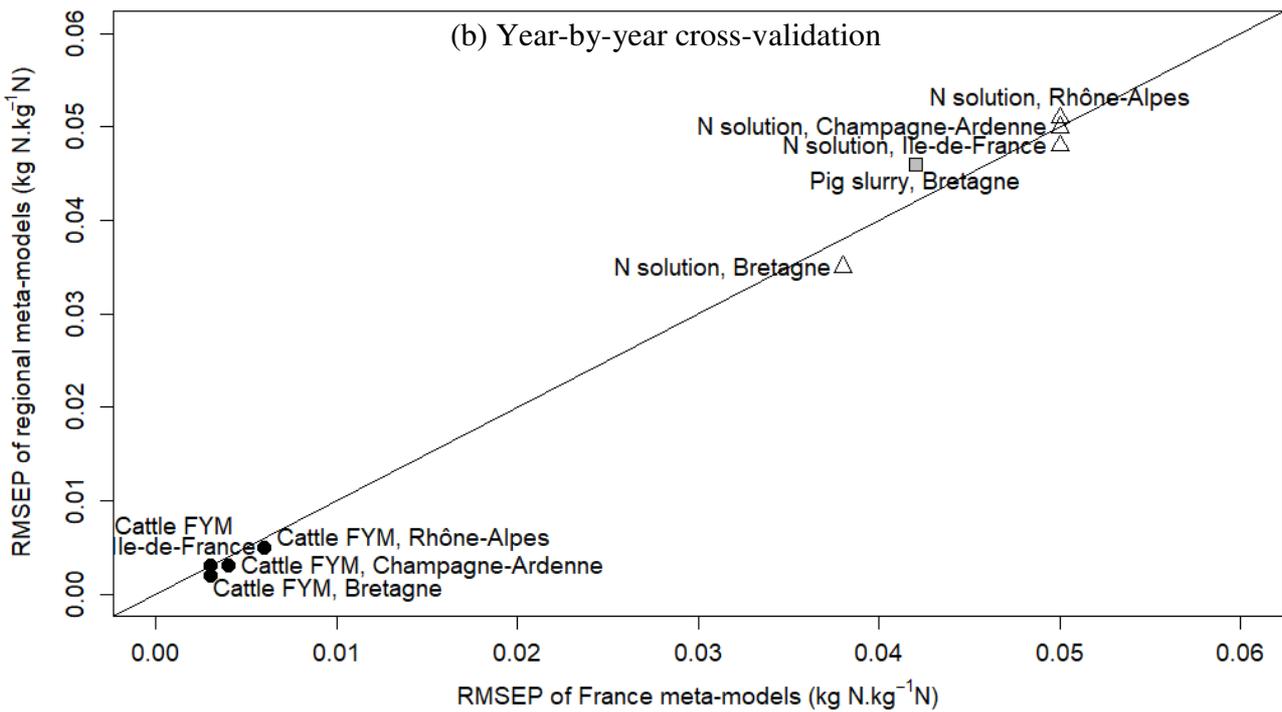
Over all meta-models (before selection), values of RMSEP range from 0.002 to 0.729 kg N.kg⁻¹ N and values of RRMSEP range from 3% to 755% of the mean emission rates. For N solution, values of RMSEP range from 0.033 to 0.729 kg N.kg⁻¹ N and values of RRMSEP range from 24 to 755 % of the mean emission rates. For cattle FYM and pig slurry, values of RMSEP range from 0.002 to 0.074 kg N.kg⁻¹ N and 0.045 to 0.598 kg N.kg⁻¹ N, respectively, and values of RRMSEP range from 3 to 105 % and 17 to 260 % of the mean emission rates, respectively.

373 For a given fertilizer type, the RMSEP of the best meta-models fitted at the regional and national
374 scales are close, and the regional meta-models do not systematically outperform the national meta-
375 models (Figure 2). For N solution, the France meta-model produces a smaller RMSEP, thus providing a
376 more accurate approximation of Volt'Air than regional meta-models in all considered regions except in
377 Bretagne for the meta-model selected using a year-by-year cross-validation. For pig slurry in Bretagne,
378 the France meta-model performs better than the regional meta-model. For cattle FYM, regional and
379 France meta-models show very similar performances.

380 In the following, only the meta-models showing the lowest RMSEP are considered. Regional
381 emission rates are thus further estimated using either France or regional meta-models, depending on
382 their RMSEP values. Similar results are observed for SAR-by-SAR and year-by-year cross-validation
383 methods.



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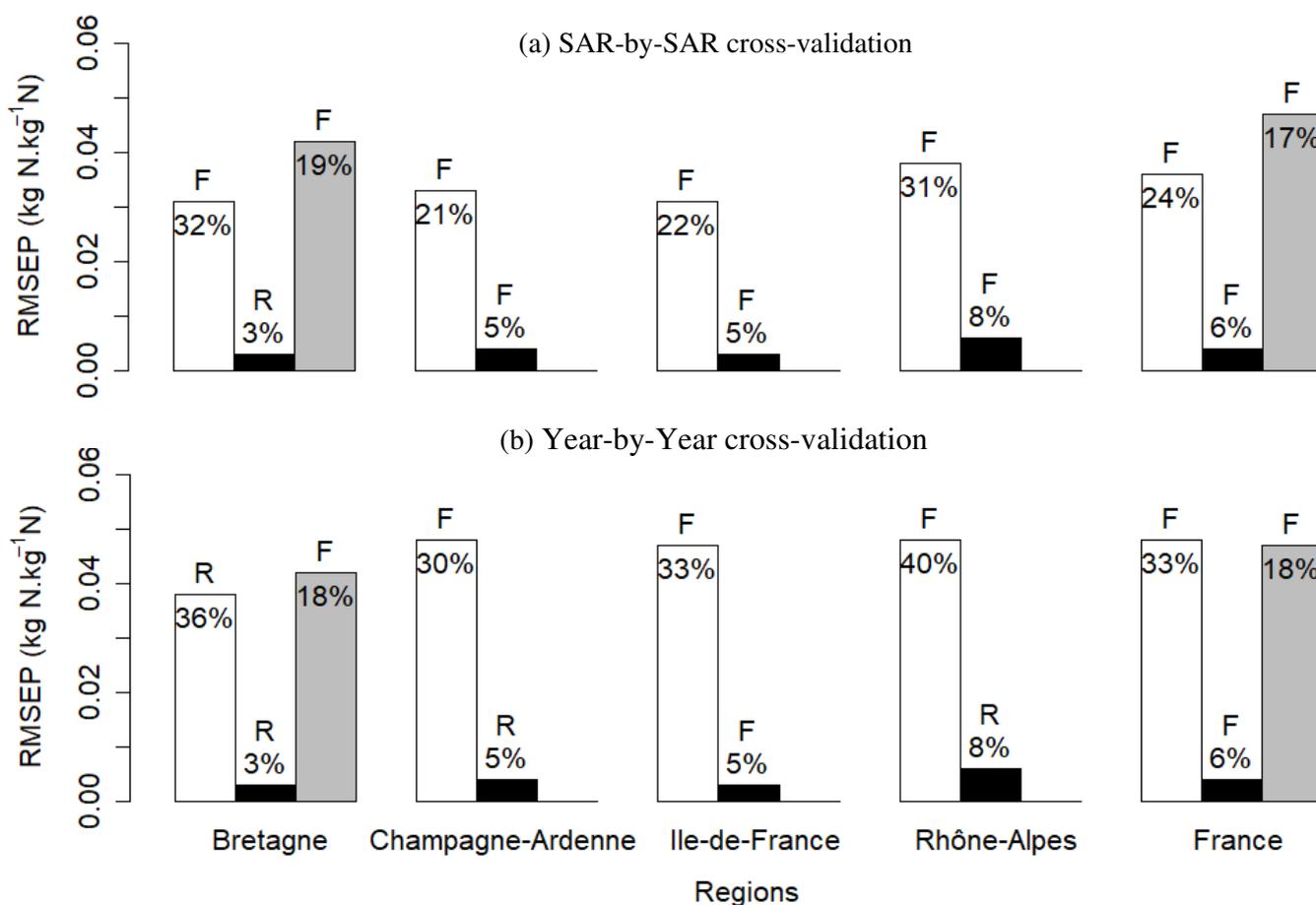
386 **Figure 2:** Comparison of RMSEP by (a) SAR-by-SAR cross-validation and (b) year-by-year cross-
 387 validation, obtained for each region and each type of fertilizer with meta-models fitted at the regional
 388 and France scales.

389 The RMSEP values from the best meta-models are shown in Figure 3, as absolute values and as
 390 relative values compared to the mean emission rates (relative root mean square error of prediction

391 (RRMSEP) as %). For cattle FYM, the RMSEP values were very low ($<0.005 \text{ kg N.kg}^{-1} \text{ N}$) and the
392 RRMSEP did not exceed 8% of the mean emission rates. Values of RMSEP were higher for pig slurry
393 and the corresponding RRSMEP ranged from 17 to 19% for this type of fertilizer. For N solution, the
394 RMSEP values were of the same order of magnitude as those obtained for pig slurry; they never
395 exceeded $0.06 \text{ kg N.kg}^{-1}$. But as the mean emission rates were low for N solution, the corresponding
396 RRMSEP values were systematically higher than 21% (Figure 3). These results show that the meta-
397 models provided a better approximation of Volt'Air for cattle FYM than for pig slurry and N solution.
398 This result is confirmed in Figure 4, where the relationships between the meta-model and the Volt'Air
399 outputs are stronger for cattle FYM (RRMSEP $<8\%$) than for the other two types of fertilizer
400 (RRMSEP $>17\%$).

401

□ N solution ■ Cattle FYM ■ Pig slurry



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Figure 3: Root Mean Square Error of Prediction (RMSEP) of the best meta-models selected per type of fertilizer per region. The letters R and F refer to the regional and France meta-model, respectively, for each fertilizer type. The percentages indicate the relative root mean square error of prediction (RRMSEP) of each meta-model (i.e., RMSEP/mean of emission rates).

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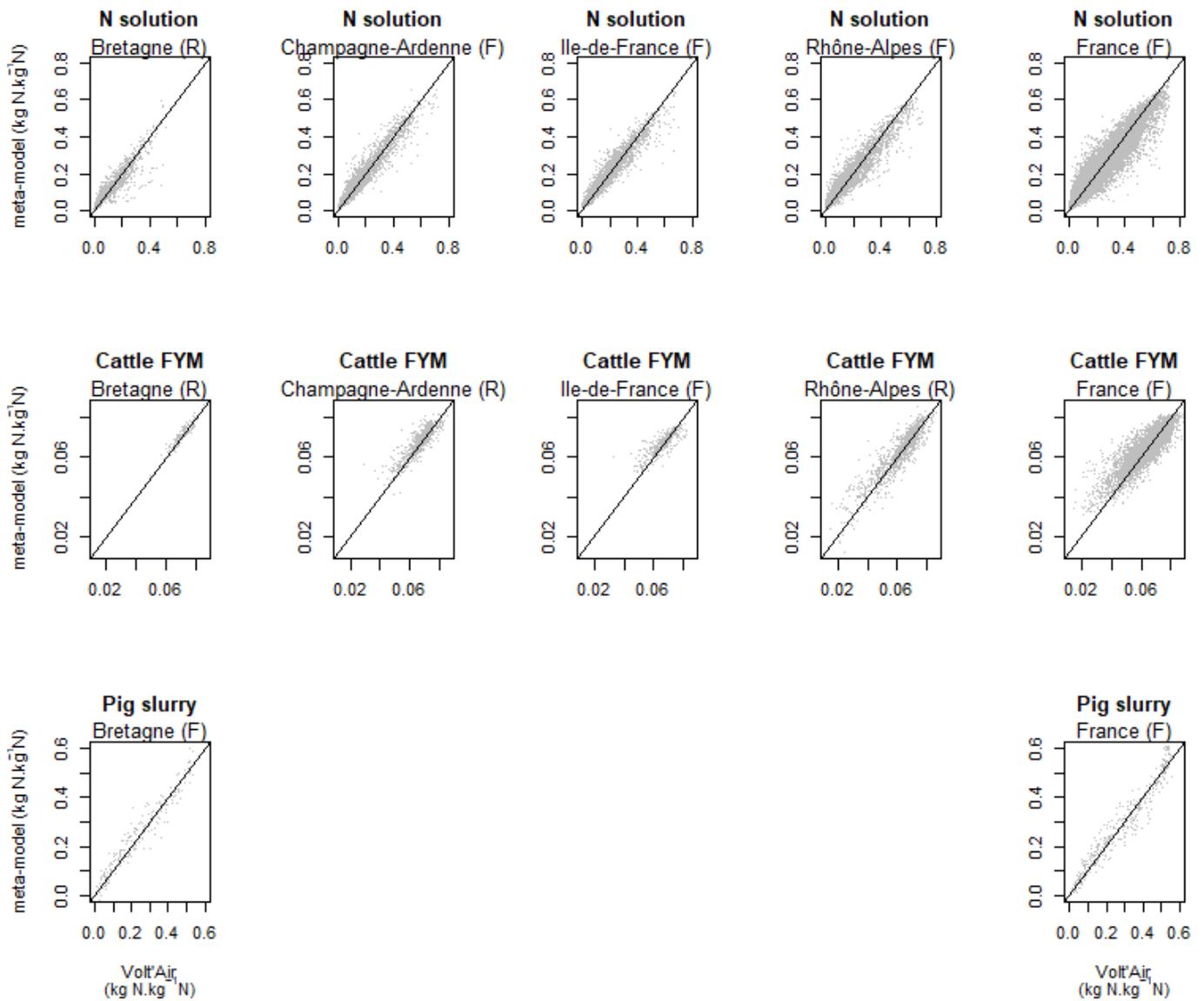
The performances of the meta-models did not differ much among regions. No single region showed systematically lower RMSEP or RRMSEP. The RMSEP tended to be higher when calculated year by year than SAR by SAR (Figure 3ab). This result was expected, because a larger number of data were removed at each iteration of the cross-validation when implemented year by year. The differences were large for N solution, small for pig slurry, and non-detectable for cattle FYM.

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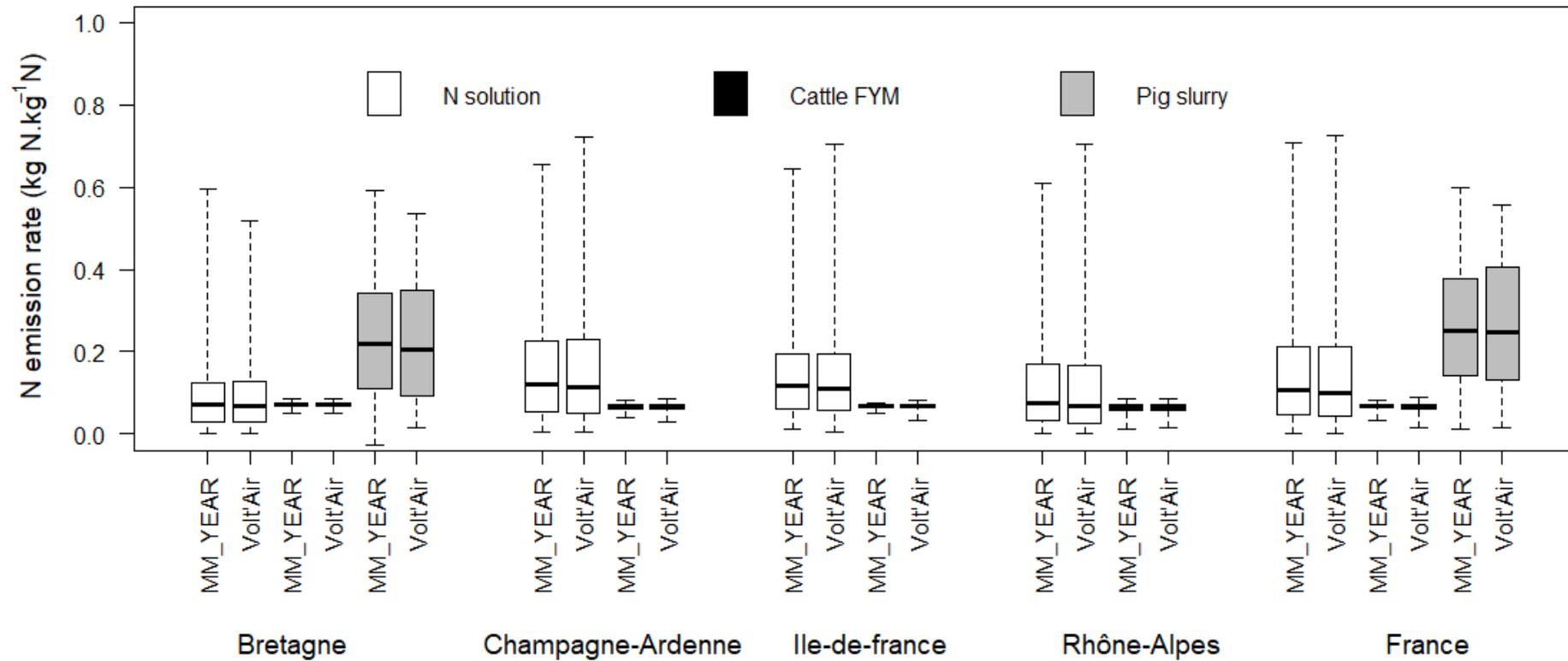
The distributions of the emission rates obtained with the selected meta-models were very similar to those obtained with Volt'Air, both at regional and national scales. This is illustrated in Figure 5 for

415 the meta-models selected by year-by-year cross-validation; results were similar with the meta-models
416 selected by SAR-by-SAR cross-validation and are therefore not shown. In Figure 5, each box-plot
417 presents the minima, 1st quartiles, medians, 3rd quartiles, and maxima calculated over the entire
418 considered site*crop*fertilization*year, using the selected meta-models and Volt'Air. Differences
419 between the meta-models and Volt'Air were very small, especially for the minima, 1st quartiles,
420 medians, and 3rd quartiles. These results indicate that the emission rate distributions produced by the
421 meta-models and by the process-based model shared similar characteristics.



422

423 Figure 4: Scatter plot of meta-models selected by year-by-year cross-validation versus Volt'Air
 424 simulated emission rates (in $\text{kg N.kg}^{-1} \text{N}$). The letters R and F stand for regional and France meta-
 425 model, respectively, for each fertilizer type.



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Figure 5: Distributions of the nitrogen emission rate calculated with the meta-models selected by year-by-year cross-validation (MM_YEAR) and with the process-based model Volt'Air (in kg.kg⁻¹ N). Each box-plot describes the minimum, 1st quartile, median, 3rd quartile and maximum values of the emission rates over the considered site*crop*fertilization*year.

3.2. Characteristics of selected meta-models

Over the five considered geographical areas (four regions plus France) and the three types of fertilizer, four different meta-models were selected by SAR-by-SAR cross-validation, versus seven by year-by-year cross-validation (Table S1 in the supplementary material). This difference is due to the number of meta-models selected for N solution and cattle FYM. For N solution, a single meta-model (the France meta-model) was indeed selected using SAR-by-SAR cross-validation for four different regions (Table S1) while the same meta-model was selected using year-by-year cross-validation for three different regions (Table S2). For cattle FYM, a single meta-model (the France meta-model) was selected using SAR-by-SAR cross-validation for three different regions (Table S1) while the same meta-model was only selected using year-by-year cross-validation for one region, Ile-de-France (Table S2).

Half of the meta-models selected by SAR-by-SAR cross-validation were Random Forest models; while the other half were ordinary linear regression models (Table S1). The two linear regression models selected by SAR-by-SAR cross-validation included quadratic and interaction terms and one of them (Meta-model 4) was fitted using a box-cox transformation of the output. The interaction *Wind speed * Cumulative rainfall* and the quadratic terms *Latitude²*, *Longitude²*, *Date of N application²*, *Mean air temperature²* and *Cumulative rainfall²* were frequently selected (see Tables S1 and S2 in the supplementary material2). The quadratic terms of “*Dose of fertilizer application*”, “*Soil organic carbon content*” and “*Maximum air temperature*” were never selected, nor was the interaction “*Mean air temperature * Cumulative rainfall*”. Among the considered statistical methods, GAM, PLSR, and LASSO were never selected whatever the type of cross-validation used.

In the four meta-models selected by SAR-by-SAR cross-validation, a number of weather input variables (e.g., *Mean air temperature*, *Wind speed*, *Cumulative rainfall* and *Incident solar radiation*) and *Initial soil water content* were systematically selected. The *Soil organic carbon*

454 *content* was selected in one meta-model only, as was the *Region* variable, which was an input
455 candidate for three meta-models. The *Dose of fertilizer application* was never selected but it was an
456 input candidate for one meta-model only (Table S1). All other variables were selected in at least half
457 of the meta-models. The number of selected inputs ranged from 6 to 15 (if we exclude the quadratic
458 and interaction terms), depending on the meta-model considered. These numbers should be
459 compared to the total number of candidate inputs, comprised between 15 and 18 (Table 2). Meta-
460 models 1 and 3 were the most parsimonious; they included 10 selected inputs from the 18 input
461 candidates and 6 selected inputs from the 17 input candidates, respectively, i.e., about half or one
462 third of the number of candidate inputs (Table S1). Conversely, the meta-models 2 and 4 included a
463 large share of the input candidates (12/15 and 15/18, respectively). The time-period used to calculate
464 the weather inputs ranged from six to seven successive days for N solution and from one to three
465 successive days for cattle FYM; for pig slurry, it was five successive days (Tables S1 and S2).

466 The seven meta-models selected by year-by-year cross-validation shared certain similarities
467 with the four meta-models selected by SAR-by-SAR cross-validation. The inputs of these meta-
468 models (Table S2) were often the same as those reported in Table S1. *Initial soil water content*,
469 *Mean air temperature*, *Incident solar radiation*, *Wind speed* and *Cumulative rainfall* were frequently
470 selected, while the *Dose of fertilizer application*, *Soil organic carbon content*, *Region*, *Latitude*,
471 *Longitude* and *Relative air humidity*, were less frequently selected. Soil properties (*Bulk density*, *Silt*
472 *content*, *Clay content* and *Organic carbon content*) were less frequently selected in N solution meta-
473 models. The total number of selected inputs also ranged from 6 to 15 (if we exclude the quadratic
474 and interaction terms), depending on the meta-model considered. These meta-models exhibited
475 several differences compared to those selected by SAR-by-SAR cross-validation. In particular, most
476 of the meta-models selected by year-by-year cross-validation were ordinary linear regression models
477 (Table S2); RF was used by only two meta-models. All the five selected ordinary linear regression
478 models included quadratic and interaction terms (Table S2); two out of them included a box-cox

479 transformation of the output while the other three linear meta-models included untransformed output.
480 In the five selected linear regression meta-models, the interactions *Minimum air temperature* *
481 *Cumulative rainfall*, *Maximum air temperature* * *Cumulative rainfall*, and the quadratic terms *Wind*
482 *speed*², *Soil bulk density*², *Mean air temperature*², *Incident solar radiation*² and *Cumulative rainfall*²
483 were frequently selected (see Tables S1 and S2 in the supplementary material2).

484

485 The R objects of the meta-models based on linear regression and Random Forest are available
486 on request from the authors.

487

488 **4. DISCUSSION**

489 Our meta-model assessments show contrasting results. For cattle FYM and pig slurry, on the
490 one hand, meta-models provide a good approximation of Volt'Air, with an RRMSEP always lower
491 than 8% and 19% of the mean emission rates, respectively. For N solution, meta-models provide a
492 good approximation of Volt'Air for Champagne-Ardenne, Ile-de-France and France when they were
493 assessed by the SAR-by-SAR cross-validation: RRMSEP ranged from 21% to 24% of the mean
494 emission rates. The meta-models are less accurate for N solution in other situations, specifically in
495 Bretagne and Rhône-Alpes with SAR-by-SAR cross-validation, and in all the geographical areas
496 with year-by-year cross-validation; RRMSEP ranged from 31% to 40% of the mean emission rates in
497 these situations. Despite these contrasting results, our meta-models provide a good approximation of
498 the distributions of emission rates at the regional scales for all fertilizer types, including N solution
499 (Figure 5). They give realistic ranges of emission rates and could thus be used for regional NH₃
500 emission inventories.

501 The relatively poor performances of the N solution meta-models may be due to the small
502 number of soil variables selected (see below). Other variables may have a strong influence on
503 volatilization, at least for some of the considered years. Our meta-models may not fully account for
504 the soil characteristics influencing soil transfer of heat, water and ammoniacal-N species. The better
505 performance obtained for N solution by year-by-year cross validation may be due to a higher inter-
506 annual variability of volatilization for this type of fertilizer, or to the fact that more data are needed
507 to develop an accurate meta-model for this fertilizer.

508 Almost all the selected meta-models incorporate some nonlinear effects. The regression
509 models include quadratic terms and interactions, and Random Forest models deal with nonlinear
510 responses through their splitting rules. The presence of nonlinear terms in the selected meta-models
511 emphasizes the need to take nonlinear responses and interactions into account. The results of our
512 selection are consistent with those reported in previous meta-modeling studies. Regression meta-
513 models including linear, interaction, and quadratic terms and transformation of the output were
514 indeed commonly used for meta-modeling (Britz and Leip, 2009; Giltrap and Ausseil, 2016). RF
515 meta-models were also already used by Villa-Vialaneix et al. (2012) and were found to be an
516 accurate meta-modeling method for emulating models of nitrous oxide (N₂O) emission and N
517 leaching.

518 Four weather variables (*Wind speed, Air temperature, Cumulative rainfall, Incident solar*
519 *radiation*) were always selected in the meta-models regardless of the type of fertilizer and
520 geographical area. This is consistent with the Volt'Air sensitivity analyses performed by Theobald et
521 al. (2005, 2014) which showed that air temperature and wind speed are the most influential climatic
522 factors in the Volt'Air model. This result is due to the fact that Henry's Law and the acid-base
523 equilibrium constants are both expressed as exponential functions of temperature, increasing
524 temperatures leading to increased volatilization (Sommer et al., 2003). Furthermore, wind speed
525 promotes the dispersion of NH₃ in the gas phase and thus volatilization. Volt'Air is also sensitive to

526 rainfall (Génermont and Cellier, 1997); rainfall events after fertilizer application decrease NH₃
527 emissions by promoting the infiltration of slurry and soil solution containing ammoniacal N (Sanz-
528 Cobena et al., 2011; Sanz-Cobena et al., 2008; van der Weerden and Jarvis, 1997).

529 The time-periods selected to calculate the effect of the weather variables correspond to the
530 period of high volatilization for each type of fertilizer. The time-period selected for cattle FYM is the
531 shortest (less than three successive days versus five days or more for other fertilizers); there are two
532 reasons for this. First, cattle FYM has high dry matter content and is known not to infiltrate into the
533 soil: this was conceptualized in Volt'Air by adding a cattle FYM layer on the soil surface (Garcia et
534 al., 2012; Voylokov et al., 2014) whereas pig slurry and N solution are directly added to the first soil
535 layer. Second, cattle FYM has a higher pH (8.00) than pig slurry (7.50) and N solution (5.77), and
536 Volt'Air is highly sensitive to pH; the higher the pH, the higher the volatilization (Génermont and
537 Cellier, 1997; Le Cadre, 2004; Theobald et al., 2005). This information on the time-period during
538 which weather factors drive NH₃ volatilization is useful for further recommendations to adapt the
539 experimental design to capture the dynamics of NH₃ volatilization, as the time window for applying
540 abatement techniques depends on fertilizer type.

541 Soil properties were less frequently selected than weather variables in all meta-models,
542 especially for N solution (Tables S1 and S2). These variables had smaller variation ranges than the
543 meteorological variables, which probably led to their elimination during automatic variable selection.
544 Moreover, several soil properties are strongly correlated with *Soil pH* and *Initial soil water content*
545 (see Figure S1 in the supplementary material1), which were always selected in our meta-models
546 (except for cattle FYM, as explained below). As stated above, Volt'Air is highly sensitive to *Soil pH*.
547 Similarly, *Initial soil water content* is among the most influential parameters in the Volt'Air model
548 (Theobald et al., 2005). It influences the availability of volatile N on the soil surface by conditioning
549 the infiltration of the fertilizer's liquid fraction. The fact that cattle FYM stays on the soil surface and

550 does not infiltrate also explains why soil properties (especially *Soil pH*) are less frequently selected
551 for FYM meta-models.

552 The variable *Region* at the France level was selected only for N solution meta-models. For
553 cattle FYM and pig slurry, this variable was not selected in the France meta-models regardless of
554 cross-validation type but regional meta-models performed better than France meta-model. For these
555 two types of fertilizer, the meta-models account for regional characteristics through their soil and
556 weather inputs, *Latitude* and *Longitude*, and the *Dates of fertilizer application*.

557 The first advantage of using meta-models instead of Volt'Air is their extremely short
558 computation time. For example, the meta-model for N solution in France required less than 1s to
559 make 70,299 simulations whereas Volt'Air required approximately 1.5 days for the same number
560 of simulations with a parallel implementation on a Dell precision T7910 workstation with two Intel
561 Xeon 2.4 GHz processors (20 cores) and 128 GB RAM. Meta-models can thus easily be
562 implemented at large scales or in other applications requiring large amounts of data. The time
563 required to build the meta-models and to select the best meta-models according to the selection
564 method in Figure 1 was approximately three days with the R software on a desktop computer
565 (Processor 2.7 GHz, 8Go RAM). SAR-by-SAR cross-validation was the most computationally
566 expensive step, especially for calculations at the France scale where the number of SARs was 706
567 while it ranged from 25 to 76 for the four selected regions. RF and GAM required more computation
568 time than the other techniques, especially when the dataset was large, as with N solution in France.
569 Ordinary linear regression had the lowest computational cost.

570 The second advantage of using the meta-models is that they need fewer inputs than the total
571 number required for running Volt'Air. In our case study, Volt'Air required 16 types of input variable,
572 and five of them (i.e., the weather variables) needed to be provided at an hourly time-step for one
573 month (Section 2.1) leading to at most a total of 3,611 input variables. The use of Volt'Air is thus

574 relatively costly in terms of data acquisition. The total number of selected inputs in the meta-models
575 ranged from 6 to 15 (if we exclude the quadratic and interaction terms). In the meta-models, weather
576 inputs need to be supplied over at most seven days.

577 The meta-models produced here combine qualities of both the process-based model Volt'Air
578 and the empirical models developed and used for calculating NH₃ volatilization in the field
579 (Génermont, 2014). Our meta-models include a small number of input variables and have a very
580 short computation time. But they do not suffer from the same drawbacks as purely empirical models
581 based on a limited number of experimental datasets. The domain of validity of our meta-models is
582 related to the domain of validity of Volt'Air and the extent of the dataset used to build the meta-
583 models. Thus we can expect a larger domain of validity than for the empirical models which
584 generally focus on specific conditions, for example, that of Menzi (Menzi et al., 1998), which was
585 constructed for cattle slurry applied to grassland with a splash plate, a total ammoniacal nitrogen
586 (TAN) content of 0.75 g. kg⁻¹, a mean temperature ranging from 0 to 25°C, mean relative humidity
587 of air ranging from 0.5 to 0.9 % and no rain. The model constructed by Misselbrook et al. (2004) and
588 adjusted by Chadwick et al. (2005) does not account for the effect of wind speed on NH₃
589 volatilization from mineral fertilizers.

590 Our meta-models offer a good alternative to the default emission factors proposed by the
591 EMEP guidebook for the official French NH₃ emission inventories (CITEPA, 2017; EMEP/EEA,
592 2016), because of their relative simplicity compared to process-based models. The meta-models
593 could also be connected to crop models (e.g., CERES-EGC, Gabrielle et al., 2006; STICS, Brisson et
594 al., 2003; SYST'N, Dupas et al., 2015; Parnaudeau et al., 2012) to improve decision support tools
595 used for fertilization management.

596 The meta-modelling approach presented here is generic. It can be implemented again with
597 new sets of simulations provided by an upgraded version of Volt'Air, in new geographical areas, for

598 new management techniques (i.e., abatement techniques), or for new types of fertilizer. Our approach
599 can thus be used to produce a variety of meta-models suitable for a diversity of situations and
600 applications.

601 **References**

- 602 Abdi, H. (2010). Partial least squares regression and projection on latent structure regression (PLS
603 regression). *Wiley Interdisciplinary Reviews: Computational Statistics* 2, 97–106. DOI:
604 10.1002/wics.051.
- 605 AGRESTE (2006). "Méthodologie Enquête sur les pratiques culturelles en 2006". *Agreste Chiffres et*
606 *Données Agriculture N°200*. 11 pp.
- 607 AGRESTE (2010). "Pratiques culturelles 2006". *AGRESTE Les Dossiers N° 8 - JUILLET 2010*. 77
608 pp.
- 609 AGRESTE (2014). "Enquête Pratiques culturelles 2011: Principaux résultats". *AGRESTE Les*
610 *Dossiers N° 21 - JUILLET 2014*. 70 pp.
- 611 Akaike, H. (1974). A new look at the statistical model identification. *IEEE Transactions on*
612 *Automatic Control* 19, 716-723. DOI: 10.1109/TAC.1974.1100705.
- 613 Bobbink, R., Hornung, M., and Roelofs, J. G. M. (1998). The effects of air-borne nitrogen pollutants
614 on species diversity in natural and semi-natural European vegetation. *Journal of Ecology* 86,
615 717-738. DOI: 10.1046/j.1365-2745.1998.8650717.x.
- 616 Breiman, L. (2001). Random forests. *Mach Learn*, 45, 5-32. DOI:
617 <http://dx.doi.org/10.1023/A:1010933404324>.
- 618 Brisson N., Garya C., Justes E., Roche R., Mary B., Ripoche D., Zimmer D., Sierra J., Bertuzzi P.,
619 Burger P., Bussièrre F., Cabidoche Y.M., Cellier P., Debaeke P., Gaudillère J.P., Hénault C.,
620 Maraux F., Seguin B., and H., S. (2003). An overview of the crop model STICS. *European*
621 *Journal of Agronomy* 13, 309-332. DOI: [https://doi.org/10.1016/S1161-0301\(02\)00110-7](https://doi.org/10.1016/S1161-0301(02)00110-7).
- 622 Britz, W., and Leip, A. (2009). Development of marginal emission factors for N losses from
623 agricultural soils with the DNDC–CAPRI meta-model. *Agriculture, Ecosystems &*
624 *Environment* 133, 267-279. DOI: 10.1016/j.agee.2009.04.026.
- 625 Chadwick D.R. (2005). Emissions of ammonia, nitrous oxide and methane from cattle manure heaps:
626 effect of compaction and covering. *Atmospheric Environment*, Vol 39, 4, 787-799. DOI:
627 <https://doi.org/10.1016/j.atmosenv.2004.10.012>
- 628 CITEPA (2017a). "Inventaire des émissions de polluants atmosphériques et de gaz à effet de serre en
629 France - Séries sectorielles et analyses étendues - Format SECTEN." *Centre Interprofessionnel*
630 *Technique d'Etude de la Pollution Atmosphérique*. 320 pp.

- 631 CITEPA (2017b). "National inventories of air emissions in France: organisation and methodology."
632 Centre Interprofessionnel Technique d'Etude de la Pollution Atmosphérique. OMINEA report
633 - 14th edition. 838 pp.
- 634 Conti S., O'Hagan A. (2010). Bayesian emulation of complex multi-output and dynamic computer
635 models. *Journal of Statistical Planning and Inference* 140, 640-650. DOI :
636 10.1016/j.jspi.2009.08.006
- 637 Dupas, R., Parnaudeau, V., Reau, R., Jeuffroy, M.-H., Durand, P., and Gascuel-Oudou, C. (2015).
638 Integrating local knowledge and biophysical modeling to assess nitrate losses from cropping
639 systems in drinking water protection areas. *Environmental Modelling & Software* 69, 101-110.
640 DOI: 10.1016/j.envsoft.2015.03.009.
- 641 Efron, B. (1983). Estimating the error rate of a prediction rule: improvement on cross-validation. *J.*
642 *Amer. Statist. Assoc.*, 78(382):316–331.
- 643 EMEP/EEA (2016a). "Air pollutant emission inventory guidebook 2016. Part B, Chapter 3.B
644 Manure management." European Monitoring and Evaluation Programme/European
645 Environment Agency. 62 pp.
- 646 EMEP/EEA (2016b). "Air pollutant emission inventory guidebook 2016. Part B, Chapter 3.D Crop
647 production and agricultural soils 2016." European Monitoring and Evaluation
648 Programme/European Environment Agency. 41 pp
- 649 FAO, IIASA, ISRIC, ISS-CAS, and JRC (2012). "Harmonized World Soil Database (Version 1.2),"
650 FAO, Rome, Italy and IIASA, Laxenburg, Austria.
- 651 Gabrielle B., Laville P., Duval O., Nicoullaud B., Germon J-C., et al. (2006). Process-based
652 modeling of nitrous oxide emissions from wheat-cropped soils at the subregional scale. *Global*
653 *Biogeochemical Cycles*, American Geophysical Union, 20, pp.GB4018.
654 <10.1029/2006GB002686>. <hal-00017134v3>.
- 655 Garcia, L., Bedos, C., Générumont, S., Braud, I., and Cellier, P. (2011). Assessing the ability of
656 mechanistic volatilization models to simulate soil surface conditions: a study with the Volt'Air
657 model. *Sci Total Environ* 409, 3980-92. DOI: 10.1016/j.scitotenv.2011.05.003.
- 658 Garcia, L., Générumont, S., Bedos, C., Simon, N. N., Garnier, P., Loubet, B., and Cellier, P. (2012).
659 Accounting for Surface Cattle Slurry in Ammonia Volatilization Models: The Case of
660 Volt'Air. *Soil Science Society of America Journal* 76, 2184. DOI: 10.2136/sssaj2012.0067.
- 661 Générumont, S. (2014). Ammonia volatilisation after application of fertilisers and organic products:
662 Potential for updating emission factors. In "Proceedings International Fertiliser Society 762".
663 IFS International Fertiliser Society (no ISSN 1466-1314), Cambridge (UK), 12/12/2014, 44 pp.
- 664 Générumont, S., and Cellier, P. (1997). A mechanistic model for estimating ammonia volatilization
665 from slurry applied to bare soil. *Agricultural and Forest Meteorology* 88 145-167. DOI:
666 10.1016/S0168-1923(97)00044-0.

- 667 G nermont, S., Gilliot, J.-M., Ramanantenasoa, M. M. J., Maury, O., Mignolet, C., Personne, E.,
668 Loubet, B., Bedos, C., and Cellier, P. (2015). "R alisation de cadastres dynamiques des
669  missions d'ammoniac li es   la fertilisation azot e aux  chelles r gionale et nationale (Spatial
670 and temporal high-resolution inventory of ammonia emissions from agricultural soils over
671 France at regional and national scales)." Convention ADEME n  1081C0031. Final report. 90
672 pp + annexes.
- 673 G nermont S., Ramanantenasoa M.M.J., Dufosse K., Maury O., Mignolet C., Gilliot J.-M. (2018)
674 Data on spatio-temporal representation of mineral N fertilization and manure N application as
675 well as ammonia volatilization in French regions for the crop year 2005/06. Data in Brief. DOI:
676 <https://doi.org/10.1016/j.dib.2018.09.119>.
- 677 Giltrap, D. L., and Ausseil, A. G. (2016). Upscaling NZ-DNDC using a regression based meta-model
678 to estimate direct N₂O emissions from New Zealand grazed pastures. *Sci Total Environ* 539,
679 221-30. DOI: 10.1016/j.scitotenv.2015.08.107.
- 680 Hafner S.D., Pacholski A., Bittman S., Burchill W., Bussink W., Chantigny M., Carozzi M.,
681 G nermont S., H ni C., Hansen M.N., Huijsmans J., Hunt D., Kupper T., Lanigan G., Loubet
682 B., Misselbrook T., Meisinger J.J., Neftel A., Nyord T., Pedersen S.V., Sintermann J.,
683 Thompson R.B., Vermeulen B., Vestergaard A.V., Voylokov P., Williams J.R., and S.G., S. (in
684 press). The ALFAM2 database on ammonia emission from field-applied manure: Description
685 and illustrative analysis. *Agricultural and Forest Meteorology*, DOI:
686 <https://doi.org/10.1016/j.agrformet.2017.11.027>.
- 687 Huijsmans, J. F. M., Holterma, H., Vermeulen, G. D., Stolk, A. J., and van Pul (2014). "Simulating
688 emission of ammonia after liquid manure application on arable land; Preliminary performance
689 assessment of the Volt'Air model for manure application conditions in the Netherlands ". *Plant
690 Research International*, Wageningen, The Netherlands. 32 pp.
- 691 IPCC (2006). "IPCC Guidelines for National Greenhouse Gas Inventories. Volume 4 – Agriculture,
692 Forestry and Other Land Use. Chapter 10: Emissions from Livestock and Manure
693 Management." IPCC. 87 pp.
- 694 JRC (2008). MARS Bulletin Vol 16 No 6.
- 695 Katayanagi, N., Fumoto, T., Hayano, M., Takata, Y., Kuwagata, T., Shirato, Y., Sawano, S., Kajiura,
696 M., Sudo, S., Ishigooka, Y., and Yagi, K. (2016). Development of a method for estimating total
697 CH₄ emission from rice paddies in Japan using the DNDC-Rice model. *Sci Total Environ* 547,
698 429-40. DOI: 10.1016/j.scitotenv.2015.12.149.
- 699 Le Cadre, E. (2004). Mod lisation de la volatilisation d'ammoniac en interaction avec les processus
700 chimiques et biologiques du sol: le mod le Volt'Air, Thesis. ABIES, UMR INRA INAPG
701 Environnement et Grandes Cultures de Grignon, INAPG. 315 pp.
- 702 Makowski D. et al. (2015). A statistical analysis of ensembles of crop model responses to climate
703 change factors. *Agriculture and Forest Meteorology* 214–215, 483–493. DOI:
704 <https://doi.org/10.1016/j.agrformet.2015.09.013>

- 705 Martin, P., Ronfort, C., Laroutis, D., Souchère, V., and Sebillotte, C. (2014). Cost of best
706 management practices to combat agricultural runoff and comparison with the local
707 populations' willingness to pay: Case of the Austreberthe watershed (Normandy, France). *Land*
708 *Use Policy* 38, 454-466. DOI: 10.1016/j.landusepol.2013.12.014.
- 709 Menzi, H., Katz, P. E., Fahrni, M., Neftel, A., and Frick, R. (1998). A simple empirical model based
710 on regression analysis to estimate ammonia emissions after manure application. *Atmospheric*
711 *Environment* 32, 301-307. DOI: [https://doi.org/10.1016/S1352-2310\(97\)00239-2](https://doi.org/10.1016/S1352-2310(97)00239-2).
- 712 Mignolet, C., Schott, C., and Benoit, M. (2007). Spatial dynamics of farming practices in the Seine
713 basin: methods for agronomic approaches on a regional scale. *Sci Total Environ* 375, 13-32.
714 DOI: 10.1016/j.scitotenv.2006.12.004.
- 715 Misselbrook, T. H., Sutton, M. A., and Scholefield, D. (2004). A simple process-based model for
716 estimating ammonia emissions from agricultural land after fertilizer applications. *Soil Use and*
717 *Management* 20, 365-372. DOI: 10.1111/j.1475-2743.2004.tb00385.x.
- 718 Moldanová, J., Grennfelt, P., Jonsson, Å., Simpson, D., Spranger, T., Aas, W., Munthe, J., and Rabl,
719 A. (2011). Chapter 18. Nitrogen as a threat to European air quality. In "The European Nitrogen
720 Assessment. Sources, Effects and Policy Perspectives" (M. A. Sutton, C. M. Howard, J. W.
721 Erisman, G. Billen, A. Bleeker, P. Grennfelt, H. V. Grinsven and B. Grizzetti, eds.).
722 Cambridge University Press, 405-433 pp
- 723 Panagos, P., Van Liedekerke, M., Jones, A., and Montanarella, L. (2012). European Soil Data
724 Centre: Response to European policy support and public data requirements. *Land Use Policy*
725 29, 329-338. DOI: 10.1016/j.landusepol.2011.07.003.
- 726 Parnaudeau, V., Reau, R., and Dubrulle, P. (2012). A tool to assess N losses towards environment at
727 the cropping system scale: the software Syst'N (in French). *Innovations Agronomiques* 21, 59-
728 70.
- 729 Ramanantenasoa, M. M. J., Gilliot, J.-M., Mignolet, C., Bedos, C., Mathias, E., Eglin, T., Makowski,
730 D., and Générmont, S. (2018). A new framework to estimate spatio-temporal ammonia
731 emissions due to nitrogen fertilization in France. *Science of The Total Environment* 645, 205-
732 219.
- 733 Razavi S. , Tolson B.A., Burn D.H. (2012). Review of surrogate modeling in water resources. *Water*
734 *Resour. Res.*, 48 W07401.
- 735 Sanz-Cobena, A., Misselbrook, T., Camp, V., and Vallejo, A. (2011). Effect of water addition and
736 the urease inhibitor NBPT on the abatement of ammonia emission from surface applied urea.
737 *Atmospheric Environment* 45, 1517-1524. DOI: 10.1016/j.atmosenv.2010.12.051.
- 738 Sanz-Cobena, A., Misselbrook, T. H., Arce, A., Mingot, J. I., Diez, J. A., and Vallejo, A. (2008). An
739 inhibitor of urease activity effectively reduces ammonia emissions from soil treated with urea
740 under Mediterranean conditions. *Agriculture, Ecosystems & Environment* 126, 243-249. DOI:
741 10.1016/j.agee.2008.02.001.

- 742 Sintermann, J., Neftel, A., Ammann, C., Häni, C., Hensen, A., Loubet, B., and Flechard, C. R.
743 (2012). Are ammonia emissions from field-applied slurry substantially over-estimated in
744 European emission inventories? *Biogeosciences* 9, 1611-1632. DOI: 10.5194/bg-9-1611-2012.
- 745 Smith, E., Gordon, R., Bourque, C., Campbell, A., Genermont, S., Rochette, P., and Mkhabela, M.
746 (2009). Simulated management effects on ammonia emissions from field applied manure.
747 *Journal of Environmental Management* 90, 2531–2536. DOI: 10.1016/j.jenvman.2009.01.012.
- 748 Søgaard, H. T., Sommer, S. G., Hutchings, N. J., Huijsmans, J. F. M., Bussink, D. W., and
749 Nicholson, F. (2002). Ammonia volatilization from field-applied animal slurry- the ALFAM
750 model. *Atmospheric Environment* 36, 3309-3319. DOI 10.1016/S1352-2310(02)00300-X.
- 751 Sommer, S. G., Générumont, S., Cellier, P., Hutchings, N. J., Olesen, J. E., and Morvan, T. (2003).
752 Processes controlling ammonia emission from livestock slurry in the field. *European Journal of*
753 *Agronomy* 19, 465-486. DOI: 10.1016/s1161-0301(03)00037-6.
- 754 Theobald M.R., Makowski D., Bedos C., Ramanantenasoa J., Générumont S. (2014) An ammonia
755 emission model for fertiliser applications suitable for use in climate change scenarios, *Open*
756 *Science Conference Integreting impacts of air pollution and climate change on ecosystems,*
757 *Budapest (Hongrie), 01-03/10/2014.*
- 758 Theobald, M. R., Williams, A. G., Rosnolbet, J., Campbell, A., Cumby, T. R., Demmers, T. G. M.,
759 Loubet, B., Parsons, D. J., Gabrielle, B., Nemitz, E., Générumont, S., Le Cadre, E., Dragosits,
760 U., Van Oijen, M., Cellier, P., and Sutton, M. A. (2005). An assessment of how process
761 modelling can be used to estimate agricultural ammonia emissions and the efficacy of
762 abatement techniques. Project Report to Defra. 27 pp.
- 763 Tibshirani, R. (1996). Regression shrinkage and selection via the lasso. *Journal of the Royal*
764 *Statistical Society. Series B* 58, 267-288. Stable URL: <http://www.jstor.org/stable/2346178>.
- 765 UNIFA (2015). *Livraison d'engrais & d'amendements minéraux basiques. Mémento de la statistique*
766 *2013-2014.* 38 pp.
- 767 van der Weerden, T. J., and Jarvis, S. C. (1997). Ammonia emission factors for N fertilizers applied
768 to two contrasting grassland soils. *Environmental Pollution* 95, 205-211. DOI:
769 [https://doi.org/10.1016/S0269-7491\(96\)00099-1](https://doi.org/10.1016/S0269-7491(96)00099-1).
- 770 Villa-Vialaneix, N., Follador, M., Ratto, M., and Leip, A. (2012). A comparison of eight
771 metamodeling techniques for the simulation of N₂O fluxes and N leaching from corn crops.
772 *Environmental Modelling & Software* 34, 51-66. 10.1016/j.envsoft.2011.05.003.
- 773 Voylokov, P., Flura, D., Décuq, C., Durand, B., Labat, C., Houot, S., and Générumont, S. (2014).
774 Accounting for fym and composts in ammonia volatilization models: The case of volt'air. In
775 "Open Science Conference Integreting impacts of air pollution and climate change on
776 ecosystems", Budapest (Hongrie).
- 777 Wood, S. N. (2017). *Generalized Additive Models: An Introduction with R, Second Edition.* CRC
778 Press, 476 pp. ISBN 9781498728331.

779 Zaman, M., Saggar, S., and Stafford, A. D. (2013). Mitigation of ammonia losses from urea applied
780 to a pastoral system: The effect of nBTPT and timing and amount of irrigation. Proceedings of
781 the New Zealand Grassland Association 75, 209-214.

782 **Web references**

783 <http://agreste.agriculture.gouv.fr/definitions/zonages/>. Last accessed 19th April 2018.