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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 (NH<sub>3</sub>) 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 NH<sub>3</sub> 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 NH<sub>3</sub> 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 NH<sub>3</sub>  
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 NH<sub>3</sub> 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 (NH<sub>3</sub>) 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 NH<sub>3</sub> emitted is necessary to  
51 define effective strategies for reducing emissions at national and regional levels, and mitigating the  
52 potential negative impact of NH<sub>3</sub> on human health (Moldanová et al., 2011) and ecosystems  
53 (Bobbink et al., 1998; IPCC, 2006). Accurate NH<sub>3</sub> 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 NH<sub>3</sub> 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 NH<sub>3</sub> 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 NH<sub>3</sub> volatilization after N fertilizer and manure applications. This model incorporates the  
66 effect of the main agronomic, soil and meteorological factors influencing NH<sub>3</sub> 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 ( $\text{N}_2\text{O}$ )  
80 emissions (Giltrap and Ausseil, 2016; Villa-Vialaneix et al., 2012) and  $\text{CH}_4$  emissions (Katayanagi  
81 et al., 2016). Regarding  $\text{NH}_3$  emissions, this study is the first attempt to develop meta-models  
82 emulating an  $\text{NH}_3$  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  $\text{NH}_3$  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 NH<sub>3</sub> 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 NH<sub>4</sub><sup>+</sup> and NH<sub>3</sub>) 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 NH<sub>3</sub> volatilization (Garcia et al., 2011; Garcia et al., 2012; Générumont and  
100 Cellier, 1997; Voylokov et al., 2014; Le Cadre, 2004). It simulates NH<sub>3</sub> 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 NH<sub>3</sub> emission simulations  
115 performed within the CADASTRE\_NH<sub>3</sub> 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\_NH<sub>3</sub> 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 NH<sub>3</sub> 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 NH<sub>3</sub> 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; 1<sup>st</sup> (i.e., the day of fertilizer application), 2<sup>nd</sup>, 3<sup>rd</sup>, 4<sup>th</sup>, 5<sup>th</sup>, 6<sup>th</sup>, 7<sup>th</sup>,  
 186 8<sup>th</sup>, 9<sup>th</sup> and 10<sup>th</sup> 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; 1<sup>st</sup>, 2<sup>nd</sup> and 3<sup>rd</sup>, from the 1<sup>st</sup> to the 10<sup>th</sup> day, from the 11<sup>th</sup> to the 20<sup>th</sup>  
 190 day, from the 21<sup>th</sup> to the 30<sup>th</sup> 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 <sup>(a)</sup>	Categorical variable
N fertilization management	- Date of N application	- Number of days from origin 01/01/2005
	- Dose of application <sup>(b)</sup>	- kg N.ha <sup>-1</sup>

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

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## 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  $y_i^\lambda$ , 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  $\lambda$  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 <sup>(1)</sup>	
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 <sup>(1)</sup> 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  $\text{NH}_3$  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.

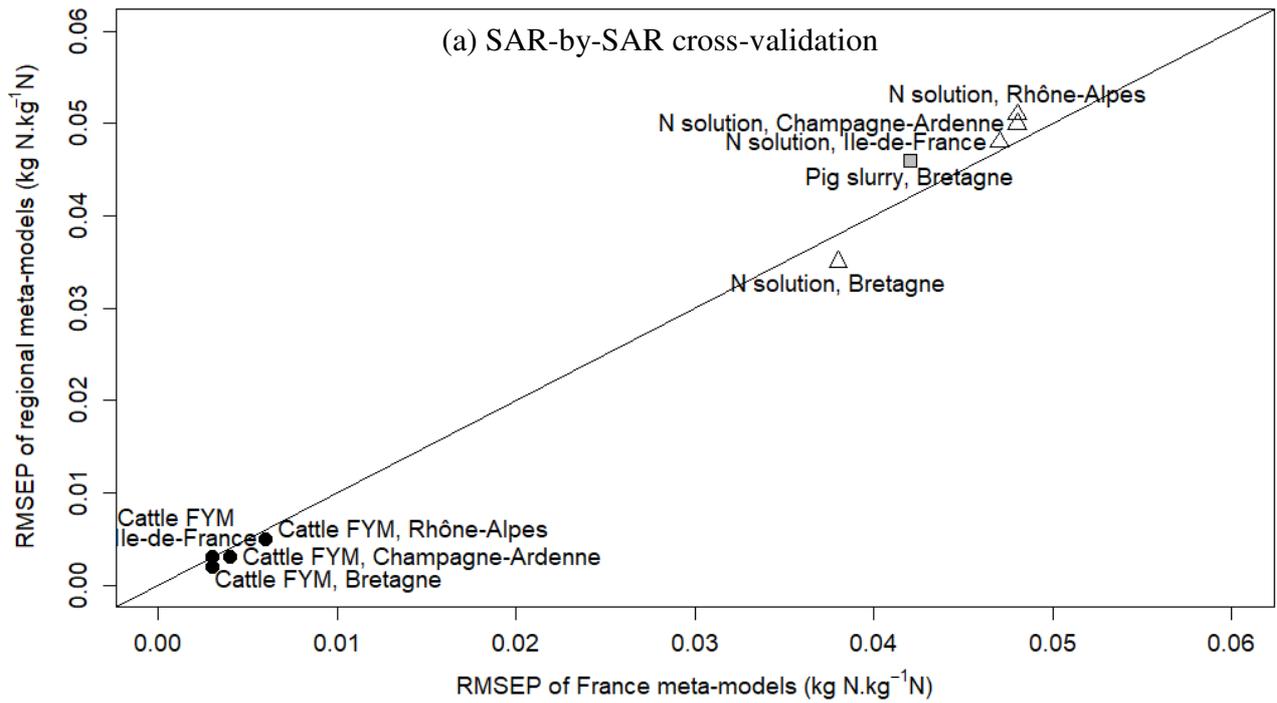
## 3. RESULTS

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

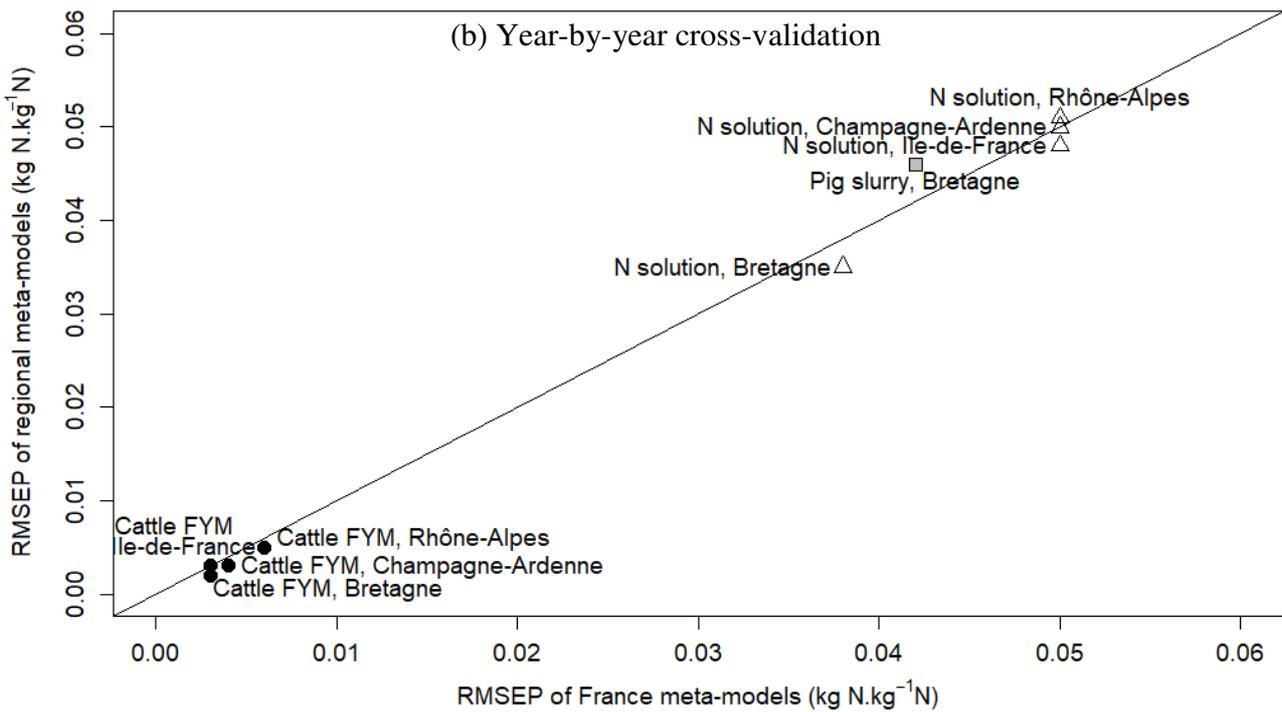
Over all meta-models (before selection), values of RMSEP range from 0.002 to 0.729 kg N.kg<sup>-1</sup> N and values of RRMSEP range from 3% to 755% of the mean emission rates. For N solution, values of RMSEP range from 0.033 to 0.729 kg N.kg<sup>-1</sup> N and values of RRMSEP range from 24 to 755 % of the mean emission rates. For cattle FYM and pig slurry, values of RMSEP range from 0.002 to 0.074 kg N.kg<sup>-1</sup> N and 0.045 to 0.598 kg N.kg<sup>-1</sup> 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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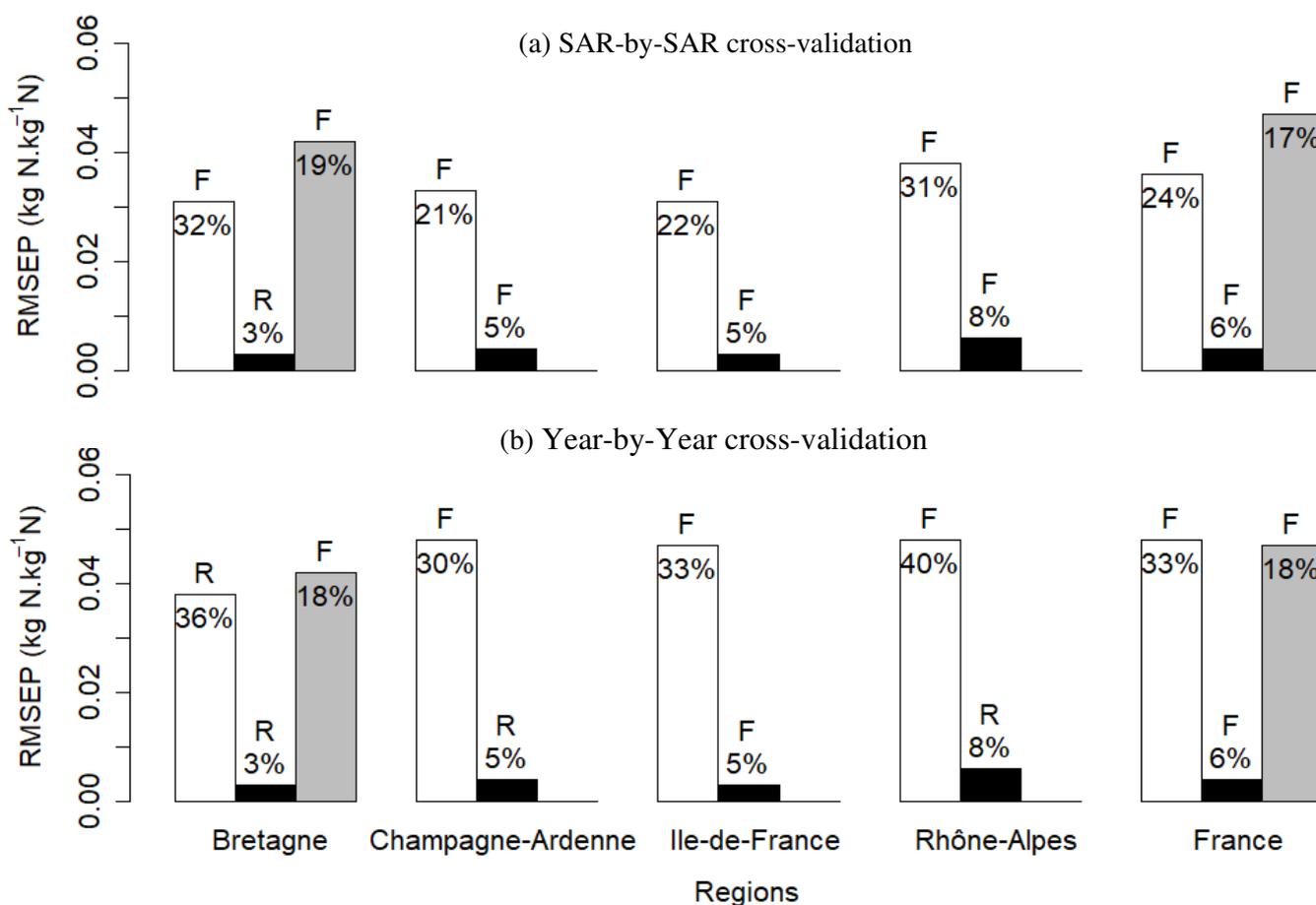
386 **Figure 2:** Comparison of RMSEP by (a) SAR-by-SAR cross-validation and (b) year-by-year cross-  
 387 validation, obtained for each region and each type of fertilizer with meta-models fitted at the regional  
 388 and France scales.

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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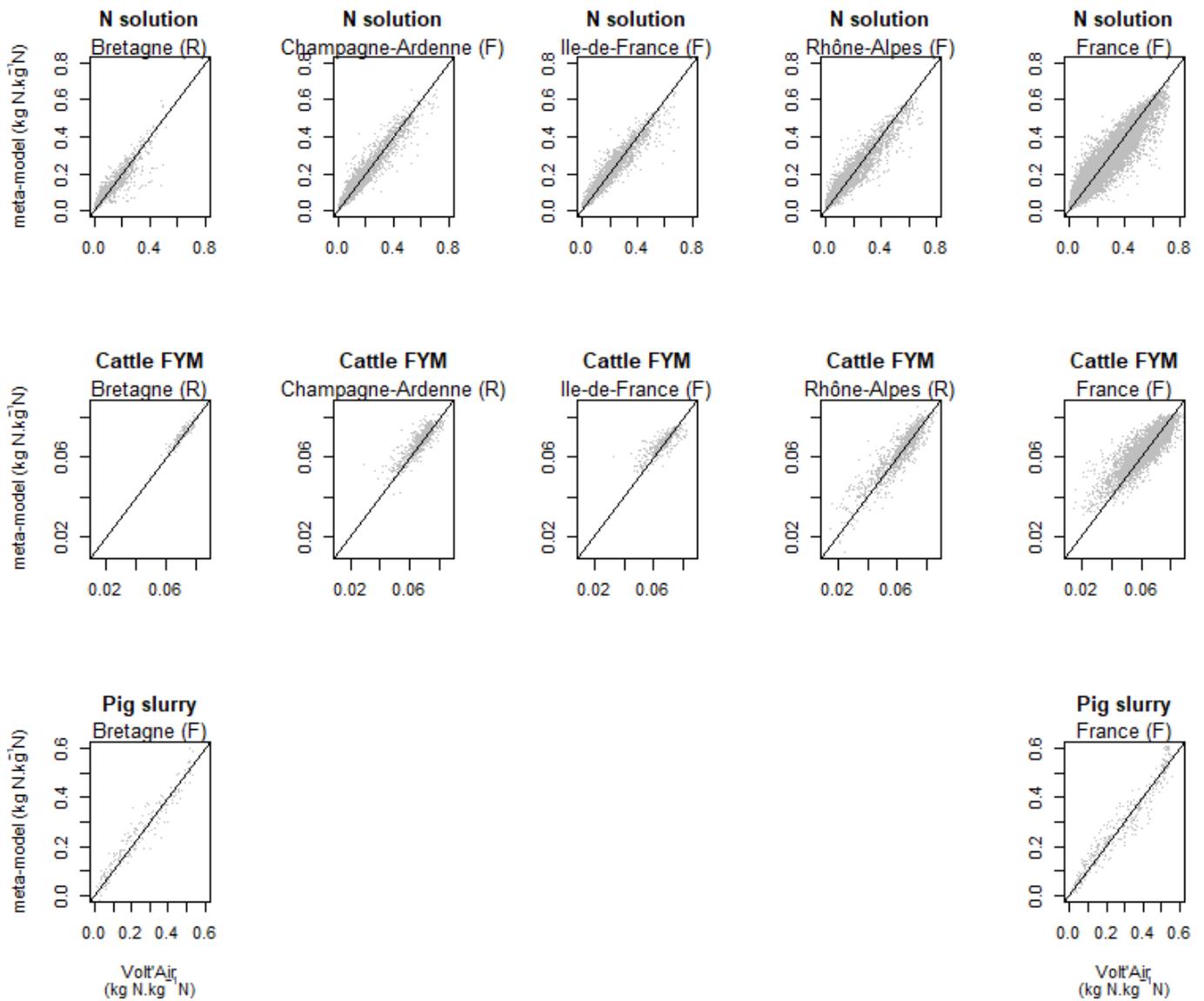
The performances of the meta-models did not differ much among regions. No single region showed systematically lower RMSEP or RRMSEP. The RMSEP tended to be higher when calculated year by year than SAR by SAR (Figure 3ab). This result was expected, because a larger number of data were removed at each 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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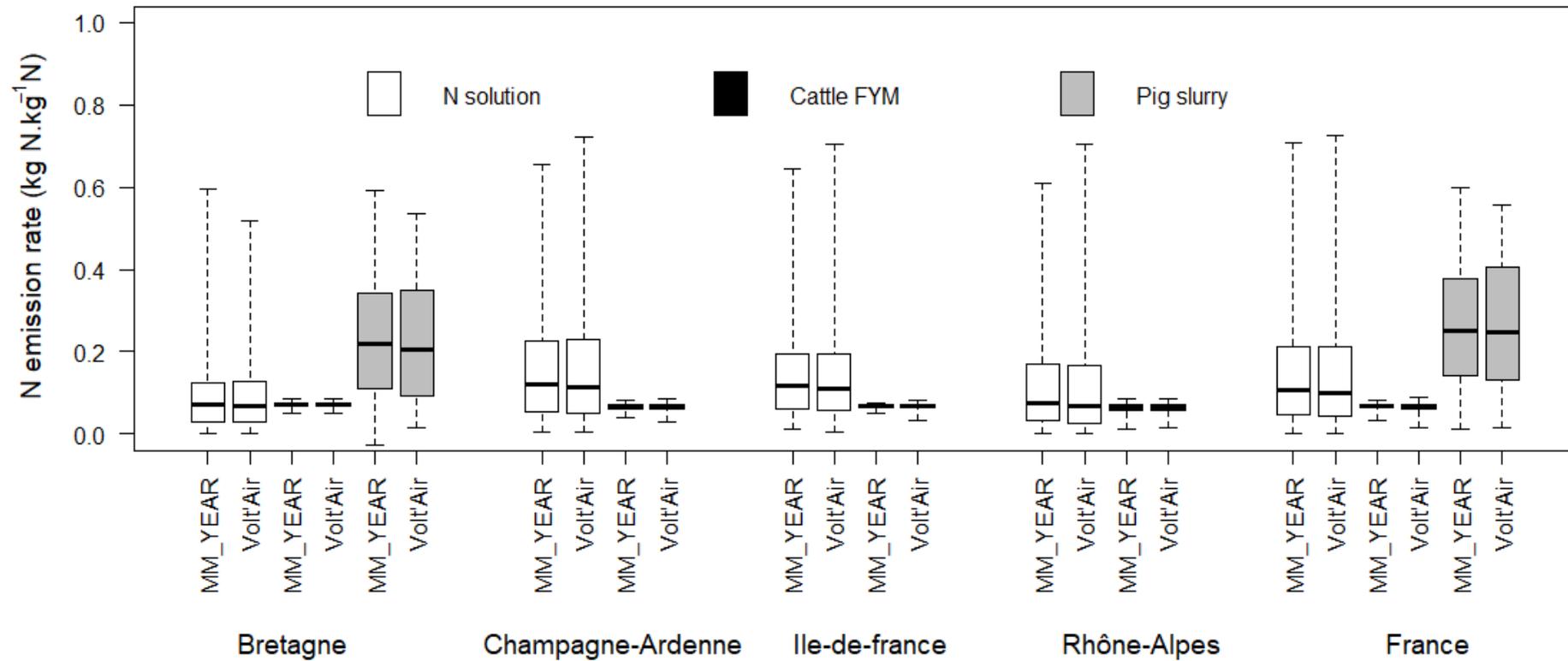
The distributions of the emission rates obtained with the selected meta-models were very similar to those obtained with Volt'Air, both at regional and national scales. This is illustrated in Figure 5 for

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<sup>-1</sup> N). Each box-plot describes the minimum, 1<sup>st</sup> quartile, median, 3<sup>rd</sup> 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 *Latitude<sup>2</sup>*, *Longitude<sup>2</sup>*, *Date of N application<sup>2</sup>*, *Mean air temperature<sup>2</sup>* and *Cumulative rainfall<sup>2</sup>* 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*<sup>2</sup>, *Soil bulk density*<sup>2</sup>, *Mean air temperature*<sup>2</sup>, *Incident solar radiation*<sup>2</sup> and *Cumulative rainfall*<sup>2</sup>  
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 NH<sub>3</sub>  
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 (N<sub>2</sub>O) 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 NH<sub>3</sub> 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  $\text{NH}_3$   
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  $\text{NH}_3$  volatilization is useful for further recommendations to adapt the  
539 experimental design to capture the dynamics of  $\text{NH}_3$  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 NH<sub>3</sub> 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<sup>-1</sup>, 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 NH<sub>3</sub>  
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 NH<sub>3</sub> 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.

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