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1 **First quantitative constraints on chlorine 36 dry deposition velocities on**  
2 **grassland: comparing measurements and modelling results**

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## 40 **1-Introduction:**

41 The environmental transition toward reducing the use of fossil energies is leading some countries to  
42 move increasingly towards less greenhouse gas emitting energies, particularly nuclear (Bickerstaff et  
43 al., 2008; Siqueira et al., 2019; Wang et al., 2019). However, this nuclear activity releases  
44 radionuclides into the environment during normal operation, in case of accidents or during the  
45 decommissioning of nuclear installations such as chlorine 36 (Imanaka et al., 2015; Katata et al., 2012;  
46 Le Dizès and Gonze, 2019; Linsley et al., 2005; Onda et al., 2020; Sasa et al., 2022). Mainly used for  
47 groundwater dating, chlorine 36 ( $^{36}\text{Cl}$ ,  $T_{1/2} = 301,000$  years) is a radionuclide of cosmogenic and  
48 anthropogenic origin (Phillips, 2000). It is produced naturally in the atmosphere following the  
49 spallation of argon 36 ( $^{36}\text{Ar}$ ) by cosmic rays, but also by interaction between the cosmic radiation and  
50 chlorine (Cl), calcium (Ca) and potassium (K) (Blinov et al., 2000; Stone et al., 1996; White and  
51 Broadley, 2001). In near-surface rocks and soils, it is formed by neutron activation of the chlorine 35  
52 ( $^{35}\text{Cl}$ ) present in the area (Bastviken et al., 2013; Van den Hoof and Thiry, 2012). Chlorine 36 was  
53 massively introduced into the environment by the nuclear tests carried out during the 1950s and 60s  
54 (Green et al., 2004; White and Broadley, 2001). The chlorine 36 peak generated by these tests  
55 exceeded natural production by about three orders of magnitude (Heikkilä et al., 2009; Pivot et al.,  
56 2019). However, during the 1980s, the chlorine 36 fluxes induced by these tests dropped back to  
57 natural levels (Heikkilä et al. 2009, Lazarev, 2003). Nowadays, anthropogenic chlorine 36 is produced  
58 by neutron activation of the stable chlorine present as an impurity in nuclear fuel (Calmet et al., 2001;  
59 Petrov and Pokhitonov, 2020; Sheppard et al., 1996; Van den Hoof and Thiry, 2012). It may therefore  
60 be discharged into the atmosphere in the form of gaseous waste during the decommissioning of  
61 nuclear plants or the recycling of spent nuclear fuels. In abnormal or incident situations (filters are  
62 assumed to be inefficient), chlorine 36 may also be discharged into the atmosphere in the form of  
63 aerosols (Le Dizès and Gonze, 2019; Rodríguez et al., 2006; Van den Hoof and Thiry, 2012). Once  
64 released into the atmosphere, chlorine 36 (gases and particles) can be transferred to the soil and  
65 vegetal cover by dry and wet deposition (Hurtevent et al., 2013; Scheffel et al., 1999; Sportisse, 2007).

66 However, owing to analytical constraints, studies on these depositions are very rare (Tosaki et al.,  
67 2012). Indeed, chlorine 36 levels in the environment are very low and accelerated mass spectrometry  
68 is the only technique able to detect and quantify these levels (Bouchez et al., 2015; Finkel et al., 1980;  
69 Nakata and Hasegawa, 2011; Poghosyan and Sturchio, 2015; Sheppard and Herod, 2012; Tosaki et al.,  
70 2012). Further, most studies focussed on wet deposition, considered as more significant (Keywood et  
71 al., 1998; Pupier et al., 2016; Schaeffer et al., 1960; Scheffel et al., 1999). There is little  
72 documentation on the dry deposition of chlorine 36 and the rare studies on this subject do not cover  
73 experimental measurements. However, owing to its relatively high mobility in the geosphere and its  
74 great bioavailability, the fate of chlorine 36 in the environment must be better studied to evaluate the  
75 environmental and human impacts (Le Dizès and Gonze, 2019; Penot and Gallou, 1977; Shaw et al.,  
76 2004; Sheppard et al., 1996; Van den Hoof and Thiry, 2012; White and Broadley, 2001). Moreover,  
77 dry deposition may account for a large part of the elimination of chemical substances under the form  
78 of trace in the troposphere (Wesely and Hicks, 2000).

79 The goal of this study is therefore to experimentally determine the chlorine 36 dry deposition  
80 velocities on grass, representative of grassland. Grass was chosen purposely as it is a link in the  
81 human food chain, particularly through cow's milk (Levchuk et al., 2008). These depositions are then  
82 modelled based on meteorological and micro-meteorological data and by adapting the existing models.  
83 These models are developed for gaseous and particulate chlorine 36. The deposition velocities  
84 calculated with the model will be compared with those obtained experimentally.

85

## 86 **2-Instruments and methods**

87 **2-1 Study site and sampling devices**

88 The site chosen for this study is IRSN La Hague’s Technical and Instrumental Platform (PTILH,  
 89 49°41'42.1"N, 1°52'24.2"W). It is situated 2 km north of the Orano La Hague plant (49°40'41.5"N  
 90 1°52'30.7"W) in France (Fig.1). This nuclear fuel reprocessing facility releases small quantities of  
 91 radionuclides, including chlorine 36, into the environment in the form of liquid or gaseous discharges  
 92 (GRNC, 1999). Moreover, the plant’s gaseous discharges contain krypton 85, which can be used as a  
 93 tracer to monitor plume dispersion and detect the periods during which the study site is located in the  
 94 plume (Connan et al., 2014; Leroy et al., 2010; Maro et al., 2017; Smith, 2010).

95 Young grass shoots (mixture of *Lolium perenne* 40%, *Poa pratensis* 40% and *Festuca rubra rubra*  
 96 20%) were grown at Pleurtuit (France). A one square meter stainless steel tub was prepared on the  
 97 study site for growing the grass. A mobile shelter equipped with a rain detector covered the plot during  
 98 rainy periods. Chlorine 36 wet depositions are therefore not considered in this study. The grass was  
 99 watered daily with ultrapure water (Merck Milli-Q, 18.2 MΩcm at 23°C) by a programmed watering  
 100 system. At the start of each campaign, a grass blank was taken to measure the initial chlorine 36  
 101 content. The atmospheric chlorine 36 close to the grass was sampled by an atmospheric sampler  
 102 AS3000 (SDEC, France). The sampler was equipped with a triethylenediamine (TC-30)-impregnated  
 103 active charcoal cartridge for sampling the gaseous chlorine 36 and a teflon and glass fiber composite  
 104 filter (EMFAB TX40H120-WW Pallflex) for sampling the particulate fraction. A total of 14  
 105 experimental campaigns were carried out between July 16, 2021 and March 18, 2022. These  
 106 campaigns lasted 2 weeks on average, except two of them, which lasted 1 month (Table 1).

107 In order to model the chlorine 36 dry deposition, the meteorological and micro-meteorological  
 108 parameters were measured during these campaigns. An ultrasonic anemometer (Young 81000V) at a  
 109 frequency of 10 Hz and installed 4.5m above the ground measured the direction (°), air speed  $u$  (m/s)  
 110 and friction velocity  $u^*$  (m/s), sensible heat flux  $H$ , Monin-Obukhov length  $L$  and atmospheric  
 111 stability. A weather station (Spectrum Watchdog, 2000 series) measured the temperature  $T_s$  (°C),  
 112 relative humidity RH (%), dew point  $T_r$  (°C) and total radiation SR (Wat/m<sup>2</sup>). The Krypton 85  
 113 concentrations were also continuously measured on site by a beta counter (Berthold LB123).

114

115 **Table 1:** Summary of the experimental campaigns

Campaigns	Start time <i>dd/mm/aaaa</i>	End time <i>dd/mm/aaaa</i>	Duration <i>days</i>	Season	Air sampled <i>m<sup>3</sup></i>
C1	16/07/2021	28/07/2021	12	Summer	404
C2	28/07/2021	09/08/2021	12		528
C3	09/08/2021	07/09/2021	29		1257
C4	07/09/2021	21/09/2021	14		606
C5	21/09/2021	06/10/2021	15	Autumn	648
C6	06/10/2021	19/10/2021	13		556
C7	19/10/2021	05/11/2021	17		737
C8	05/11/2021	23/11/2021	18		781
C9	23/11/2021	09/12/2021	16		694
C10	09/12/2021	18/01/2022	40	Winter	1000*
C11	18/01/2022	04/02/2022	17		734
C12	04/02/2022	22/02/2022	18		782
C13	22/02/2022	08/03/2022	14		609
C14	08/03/2022	18/03/2022	10		431

116 \*The atmospheric sampler is stopped after 5 weeks

## 117 2-2 Chlorine extraction

118 After sampling, the grass is dried in an oven (90°C) for one week before being crushed. The chlorine  
119 is then extracted from the samples by alkaline fusion. Alkaline fusion is a method for extracting the  
120 chlorine contained in solid matrices. It involves exposing the dry sample to high temperature in the  
121 presence of sodium hydroxide. Extraction is carried out in 2 steps, with a temperature gradient of up to  
122 450°C: chlorides with a strong affinity for basic media are firstly extracted from the sample and  
123 retained in the sodium hydroxide. The organic matter is afterwards destroyed by combustion and the  
124 sodium hydroxide is crystallized. When the grass is taken out of the oven, ultrapure water is added to  
125 the crucibles to dissolve the dry residue. This method was applied to the dried, crushed grass samples,  
126 active charcoal and aerosol filters. Before entering the oven, each sample was placed in a nickel  
127 crucible with 50 ml of sodium hydroxide 5M and spiked with a known amount of isotopically enriched  
128 stable chloride carrier (0.3g of Na<sup>35</sup>Cl (<sup>35</sup>Cl = 99.66%, Chemlab). The enriched stable chloride carrier  
129 added allows the simultaneous Cl and <sup>36</sup>Cl determination by isotope dilution. The sodium hydroxide  
130 was prepared from pellets (Normapur, VWR) dissolved in ultrapure water.

131

## 132 2-3 Sample treatment and measurement

133 The goal of the next steps is to make the sample compatible with accelerated mass spectrometry  
134 measurement, while eliminating isobaric interferences, particularly sulfur 36 (Nakata and Hasegawa,  
135 2011). After the dry residue has been dissolved, each sample is filtered to eliminate materials in  
136 suspension, particularly ash. The chlorine present in solution is afterwards precipitated into silver  
137 chloride by adding 1 milliliter of silver nitrate AgNO<sub>3</sub> 10% (analytic grade Emsure ACS, Merck) and  
138 20 milliliters of nitric acid HNO<sub>3</sub> 70% (CMOS J.T.Baker, VWR). The precipitate is recovered by  
139 filtration on a polyethersulfone filter (Millipore Express Plus, 0.45µm, 47 mm, Merck) which is  
140 collected in a tube. 10 milliliters of ammonia NH<sub>4</sub>OH 20% (Emplura, Merck) are used to dissolve the  
141 precipitate trapped on the filter and 2 milliliters of saturated barium nitrate Ba(NO<sub>3</sub>)<sub>2</sub> (analytic grade  
142 ACS, Alfa Aesar) are added to the sample tube. Adding barium makes the sulfur precipitate into  
143 barium sulfate (BaSO<sub>3</sub>). The sample is filtered after 24h, then the operation is repeated twice with 1  
144 milliliter of barium nitrate. After the third filtration of barium sulfate, 8 milliliters of nitric acid are  
145 added to each tube to precipitate the dissolved chlorides again. The samples are then cooled, stirred  
146 and centrifuged. The acid is removed, and the precipitate is washed 3 times in ultrapure water before  
147 being oven-dried at 40°C during 72h. After drying, the silver chloride precipitates were sent for  
148 chlorine 36 measurement. This measurement was performed at the French AMS national facility  
149 ASTER at CEREGE (Finkel et al., 2013). The <sup>36</sup>Cl/<sup>35</sup>Cl and the <sup>35</sup>Cl/<sup>37</sup>Cl ratios were obtained by  
150 normalization to in-house standard SM-CL-12 with an assigned <sup>36</sup>Cl/<sup>35</sup>Cl value of (1.428 ± 0.021) ×  
151 10<sup>12</sup> (Merchel et al., 2011), and assuming a natural <sup>35</sup>Cl/<sup>37</sup>Cl ratio of 3.127. <sup>36</sup>Cl and Cl concentrations  
152 in the samples were then calculated ([Appendix A](#)). These calculations are exhaustively presented by  
153 Bouchez et al., (2015). To ensure that there was no contamination with chlorine 36 during the  
154 preparation of the samples, sodium hydroxide, active charcoal and filter blanks were prepared,  
155 following the same protocol as for the samples. NIST peach leaf standards (SRM 1547) were used to  
156 determine the general efficiency of the protocol.

157

## 158 2-4 Determining deposition velocities

159 The parameter commonly used to assess deposition is the deposition velocity (V<sub>d</sub>), whose product with  
160 concentration at height z (C<sub>z</sub>) gives a mass deposition flux per unit of time (F) (Eq. (1)) (Seinfeld and  
161 Pandis, 2016).

$$V_d = -\frac{F}{c_z} \quad (1)$$

162 The chlorine 36 deposition velocities on grass were calculated with equation (1). The <sup>36</sup>Cl atmospheric  
 163 concentration corresponds to the sum of the gaseous and particulate fractions, both expressed in Bq.m<sup>-3</sup>  
 164 of air. The deposition flux (Bq.m<sup>-2</sup>s<sup>-1</sup>) was determined from the difference in concentration between  
 165 the grass blank (Bq.m<sup>-2</sup>) and the grass sample harvested at the end of the campaign (Bq.m<sup>-2</sup>) divided  
 166 by the campaign duration (s).

## 167 2-5 Parameterizing the chlorine 36 dry deposition models

### 168 2-5-1 Particulate deposition model

169 The model chosen to determine the particulate chlorine 36 dry deposition velocities is the ‘Damay-  
 170 Pellerin’ model. This model determines the particle dry deposition velocities on a substrate based on  
 171 the particle diameter, air friction velocity and atmospheric stability (Eq. (2) and (3)) (Damay et al.,  
 172 2009; Pellerin et al., 2017).

$$\frac{v_d}{u^*} = A \quad \text{in a stable or neutral atmosphere (1/L > -0.02)} \quad (2)$$

173

$$\frac{v_d}{u^*} = A \left( 1 + \left( \frac{B}{L} \right)^{\frac{2}{3}} \right) \quad \text{in an unstable atmosphere (1/L < -0.02)} \quad (3)$$

174

175 where u\* is air friction velocity (m.s<sup>-1</sup>), L is Monin-Obukhov length (m), A and B are two parameters  
 176 determined experimentally for particles of diameter between 0.2 and 1.2 μm, with dry deposition on  
 177 grassland (Connan et al., 2018; Pellerin et al., 2017).

178 An exhaustive table presenting these parameters A and B for the various particle sizes is given in  
 179 [Appendix B](#). In this study, the limits of the atmospheric stability classes were defined on the basis of  
 180 the roughness length z<sub>0</sub> of our grass area (z<sub>0</sub>=0.01m) and the 1/L ratio (Golder, 1972). The atmosphere  
 181 is defined as stable if 1/L >0.02; neutral if 1/L [-0.02~0.02]; and unstable if 1/L < -0.02 (Golder,  
 182 1972).

183 As emphasized by Le Dizès and Gonze (2019), during nuclear fuel recycling, particulate chlorine 36 is  
 184 only discharged in case of an incident or filter inefficiency. However, outside these incident situations,  
 185 particulate chlorine 36 may form following the deposition and/or bonding of gaseous chlorine 36  
 186 molecules on naturally occurring aerosols in the atmosphere (Budyka, 2000). Assuming that chlorine  
 187 36 in the environment behaves similarly to natural chlorine, local aerosol sampling was carried out on  
 188 the study site (Le Dizès and Gonze, 2019). The goal of this sampling operation was to determine the  
 189 particle size fraction to which natural chlorine is preferentially bonded in this area. It was carried out  
 190 on a 13-stage low-pressure impactor (LPI DEKATI) for particle diameters between 0.3 and 10 μm.

191

### 192 2-5-2 Gas deposition model

193 The model chosen for parameterizing gaseous chlorine 36 is the ‘Big-leaf’ model, which works  
 194 according to the electrical analogy. The choice of this model is based on its simplicity and its capacity  
 195 to estimate deposition velocities more reliably than more sophisticated models (Zhang et al., 2002b).  
 196 Deposition velocity is defined as the inverse of the sum of 3 resistances (Eq. (4)), namely aerodynamic  
 197 resistance (R<sub>a</sub>), quasi-laminar resistance (R<sub>b</sub>) and canopy resistance (R<sub>c</sub>) (Seinfeld and Pandis, 2016).

$$V_d = \frac{1}{R_a + R_b + R_c} \quad (4)$$

198

199 These three resistances terms represent the general properties of the lower atmosphere or surface and  
 200 must themselves be parameterized. The input data for parametrizing these resistances are given in  
 201 **Table 2** and the main formulae will be briefly presented.

202

203 Table 2: Input data for parametrizing the resistances

Parameter	Symbol	Value
Dynamic viscosity coefficient <sup>a</sup>	$\mu$	$1,8.10^{-5} \text{ Kg.m}^{-1}.\text{s}^{-1}$
Boltzmann constant <sup>a</sup>	$K_b$	$1,4.10^{-23} \text{ J.K}^{-1}$
Von Karman constant <sup>a</sup>	$K$	0,4
Cunningham's correction <sup>b</sup>	$Cu$	$1,2.10^3 \text{ Kg.m}^{-1}.\text{s}^{-1}$
Molecule diameter ( $\text{Cl}_2$ ) <sup>c</sup>	$D_p$	$2.10^{-10} \text{ m}$
Anemometer height <sup>d</sup>	$Z$	4,5 m
Leaf area index <sup>d</sup>	$LAI$	$1,5 \text{ m}^2 \text{ of grass.m}^{-2} \text{ of ground}$
Mean free path of a molecule <sup>a</sup>	$\lambda$	$6,9.10^{-8}$
Roughness Length <sup>d</sup>	$Z_0$	$10^{-2} \text{ m}$
Aerodynamic canopy resistance reference (grassland) <sup>e</sup>	$R_{ac0}$	$50 \text{ m.s}^{-1}$
Mesophyll resistance <sup>f</sup>	$R_m$	0
Minimum stomatal resistance for water vapor <sup>g</sup>	$R_i$	$120 \text{ m.s}^{-1}$ (Summer) $9999 \text{ m.s}^{-1}$ (Winter-Autumn) $240 \text{ m.s}^{-1}$ (Spring)
Air kinematic viscosity <sup>a</sup>	$\nu$	$1,5.10^{-5} \text{ m}^2.\text{s}^{-1}$

204 <sup>a</sup> Seinfeld and Pandis (2016), <sup>b</sup> Calculated data, <sup>c</sup> Greenwood and Earnshaw (2012), <sup>d</sup> Experimental  
 205 data, <sup>e</sup> Zhang et al., 2002a, <sup>f</sup> Zhang et al., 2002b, <sup>g</sup> Wesely, 1989

206 Aerodynamic resistance ( $R_a$ ) is resistance to the transport of matter through the atmospheric surface  
 207 layer (Eq. (5) and (6)). It is common to all gases.

$$R_a = \frac{1}{k u^*} \left[ 0.74 \ln \left( \frac{z}{z_0} \right) - \Psi_H \right] \quad (\text{Padro et al., 1991}) \quad (5)$$

208

209 Where

$$\Psi_H = \begin{cases} -4.7 \frac{z}{L} & (\text{stable atmosphere}) \\ 2 * 0.74 \ln \left[ \frac{(1+y)}{2} \right], \quad y = \left( 1 - 9 \frac{z}{L} \right)^{\frac{1}{2}} & (\text{unstable atmosphere}) \end{cases} \quad (6)$$

210

211  $u^*$  is air friction velocity ( $\text{m.s}^{-1}$ ),  $k$  is the Von Karman constant,  $z$  is the anemometer height (m),  $z_0$  is  
 212 roughness length (m),  $\Psi_H$  is the stability correction function for heat and  $L$  is Monin-Obukhov length.

213

214 Quasi-laminar resistance ( $R_b$ ) represents resistance to the thin air layer in contact with the leaf surface  
215 elements and varies with substance diffusivity (Eq. (7)).

216

$$R_b = \frac{z}{k u_*} \left( \frac{\nu}{D_i} \right)^{\frac{2}{3}} \quad (\text{Padro et al., 1991}) \quad (7)$$

217

218 Where

219

$$D_i = \frac{k_B T C_u}{6\pi \mu D_p} \quad (\text{Seinfeld and Pandis, 2016}) \quad (8)$$

220

221 And

222

$$C_u = 1 + \frac{\lambda}{D_p} \left( 2.54 + 0.8 \exp \left( -\frac{0.55 D_p}{\lambda} \right) \right) \quad (\text{Seinfeld and Pandis, 2016}) \quad (9)$$

223

224  $\nu$  is air kinematic viscosity,  $D_i$  is the molecular diffusivity of the species considered in air,  $k_B$  is the  
225 Boltzman constant,  $T$  is temperature in degrees kelvin,  $C_u$  is the Cunningham correction for molecules  
226 with a diameter under  $1\mu\text{m}$ ,  $\mu$  is the dynamic viscosity coefficient of air and  $D_p$  is the molecule  
227 diameter. The molecular diffusivity of molecular chlorine ( $\text{Cl}_2$ ) was calculated by means of equations  
228 (8) and (9).

229 Resistances  $R_a$  and  $R_b$  are quite similar on plant surfaces for most gases and the maximum theoretical  
230 deposition velocity of a gas can be measured as the inverse of the sum of these 2 resistances (Eq.  
231 (10)).

232

$$V_{d \max} = \frac{1}{R_a + R_b} \quad (\text{Ramsay et al., 2018}) \quad (10)$$

233

234 The surface or canopy resistance ( $R_c$ ) is generally the one which most influences deposition velocity.  
235 It can be defined as the total resistance to gas transfer at the surface and substrate (Bah, 2021).  
236 Depositions on plant surfaces are a major process for the elimination of atmospheric species, so  
237 surface resistance often reflects the efficiency of this elimination (Hewitt and Jackson, 2008). It is  
238 defined by equation (11).

$$\frac{1}{R_c} = \frac{1 - W_{st}}{R_{st} + R_m} + \frac{1}{R_{ns}} \quad (\text{L. Zhang et al., 2003}) \quad (11)$$

239 Where  $W_{st}$  is the stomatal blocking fraction,  $R_{st}$  is stomatal resistance ( $\text{s.m}^{-1}$ ),  $R_m$  is mesophyll  
240 resistance ( $\text{s.m}^{-1}$ ) and  $R_{ns}$  is non-stomatal resistance ( $\text{s.m}^{-1}$ ).

241 Stomatal resistance denotes the resistance to the transport of chemical species through the stomata and  
242 therefore the exchanges with the plant's internal tissues (Hosker and Lindberg, 1982). As the opening  
243 and closing of the stomata are conditioned by temperature and solar radiation, stomatal resistance is  
244 defined by equation (12).



245

$$R_{st} = r_i \{1 + [200(SR + 0,1)^{-1}]^2\} \{400[T_s(40 - T_s)]^{-1}\} \quad (\text{Wesely, 1989}) \quad (12)$$

246

247 where  $r_i$  is the minimum stomatal resistance for water vapor ( $\text{s.m}^{-1}$ ), SR is total solar radiation ( $\text{W.m}^{-2}$ )  
248 and  $T_s$  is air temperature ( $^{\circ}\text{C}$ ). The  $r_i$  values were defined by Wesely (1989) for various types of plant  
249 cover and for each season. The  $r_i$  values defined for grassland are shown in **Table 2**.

250 The stomatal blocking fraction  $W_{st}$  reflects the partial or total blocking of the stomata under certain  
251 environmental conditions (Bah, 2021). For a dry canopy, this parameter is always equal to 0. If rain or  
252 dew is present on the leaf surface, the canopy is referred to as ‘wet’ and a value is assigned to  $W_{st}$   
253 according to the total solar radiation (L. Zhang et al., 2003). In this study, given that the grass is  
254 protected from precipitation, the canopy is considered as wet only if  $T_s - T_r \leq 0$  where  $T_s$  is air  
255 temperature and  $T_r$  is dew point temperature. Throughout the experimental program, the air  
256 temperature ( $T_s$ ) was higher than the dew temperature ( $T_r$ ); the value 0 was therefore assigned to  $W_{st}$ .

257 Mesophyll resistance is resistance to the diffusion of a gas through the mesophyll of the plants. It  
258 depends on the chemical species (Bah, 2021). The  $R_m$  value is 100 for species with limited solubility  
259 and very low oxidizing power. For the other chemical species, it is 0 (Zhang et al., 2002b). Chlorine is  
260 one of the chemical species with high oxidizing power, so the value 0 was assigned to  $R_m$  in this study  
261 (Barnum and Coates, 2022).

262

263 Non-stomatal resistance  $R_{ns}$  is the resistance to gas diffusion through the leaf surface, independently of  
264 the stomata. It takes place at the cuticle (Bah, 2021). Non-stomatal resistance is determined by  
265 equation (13).

266

$$\frac{1}{R_{ns}} = \frac{1}{R_{ac} + R_g} + \frac{1}{R_{cut}} \quad (\text{L. Zhang et al., 2003}) \quad (13)$$

267

268 Where  $R_{ac}$  is the aerodynamic canopy resistance ( $\text{s.m}^{-1}$ ),  $R_{cut}$  is cuticle resistance ( $\text{s.m}^{-1}$ ) and  $R_g$  is  
269 ground resistance ( $\text{s.m}^{-1}$ ).

270 Like aerodynamic resistance, aerodynamic canopy resistance  $R_{ac}$  does not depend on the chemical  
271 species. It is conditioned by the environmental conditions, particularly the wind friction velocity  $u^*$   
272 and the leaf area index LAI (Bah, 2021). The leaf area index corresponds to the leaf area per ground  
273 area ( $\text{m}^2.\text{m}^{-2}$ ) (Sakai et al., 1997). The aerodynamic canopy resistance  $R_{ac}$  is defined by equation (14).

$$R_{ac} = \frac{R_{ac0} \times (LAI)^{\frac{1}{4}}}{u_*^2} \quad (\text{Zhang et al., 2002a}) \quad (14)$$

274

275 Where  $R_{ac0}$  is the aerodynamic canopy resistance reference value ( $\text{s.m}^{-1}$ ). It is  $50 \text{ s.m}^{-1}$  for grassland  
276 (Zhang et al., 2002a).

277 After many experiments on the dry deposition of ozone ( $\text{O}_3$ ) and sulfur dioxide ( $\text{SO}_2$ ), their cuticle  
278 resistance  $R_{cut}$  and ground resistance  $R_g$ , were precisely determined for each season. With the  
279 contrasting chemical properties of  $\text{O}_3$  and  $\text{SO}_2$ , the surface resistances of the other substances can be  
280 estimated by grading them according to solubility and oxidizing power measurements. The effective  
281 constants of the Henry law (H), which combine vapor pressure, solubility and dissociation in water,  
282 can be used to measure aqueous solubility. According to Le Dizès and Gonze (2019), the chlorine 36  
283 emitted following the reprocessing of nuclear waste takes the form of molecular chlorine ( $\text{Cl}_2$ ) or  
284 hydrochloric acid (HCl). But because of the fuel reprocessing process, gaseous chlorine discharges

rather take the form of Cl<sub>2</sub> (Bah, 2021). As Cl<sub>2</sub> has an oxidizing power close to that of ozone (E<sup>o</sup><sub>Ozone</sub> = 2V and E<sup>o</sup><sub>Chlorine</sub> = 1.36V), and their Henry constants are of the same order of magnitude (H<sup>cp</sup><sub>ozone</sub> = 1x10<sup>-4</sup> mol.m<sup>-3</sup>.Pa<sup>-1</sup> and H<sup>cp</sup><sub>chlorine</sub> = 9x10<sup>-4</sup> mol.m<sup>-3</sup>.Pa<sup>-1</sup>), the R<sub>cut</sub> values of R<sub>g</sub> chosen for the Cl<sub>2</sub> dry deposition model are therefore those of ozone (Sander, 2015; Wei et al., 2017) (Table 3).

289  
290  
291  
292

Table 3: R<sub>cut</sub> and R<sub>g</sub> values estimated for Cl<sub>2</sub> from those for O<sub>3</sub> (Zhang et al., 2002b)

	Summer	Autumn	Winter	Spring
R <sub>cut</sub> Cl <sub>2</sub>	1500	2000	6000	1500
R <sub>g</sub> Cl <sub>2</sub>	400	400	700	400

293

### 294 3- Results and discussion

#### 295 3-1 Chlorine 36 levels in the blanks and extraction efficiency

296 The blanks ratios calculated from the sodium hydroxide blanks are on average <sup>36</sup>Cl/<sup>35</sup>Cl = 8 ± 1.7x10<sup>-15</sup>  
 297 at.at<sup>-1</sup> (n=9), for active charcoal (n=4) 3.4 ± 0.3x10<sup>-14</sup> at.at<sup>-1</sup> and 6.6 ± 1x10<sup>-15</sup> at.at<sup>-1</sup> for aerosol filters  
 298 (n=4). The larger <sup>36</sup>Cl/<sup>35</sup>Cl ratio for active charcoal is not surprising, since it originates from plants and  
 299 the environmental <sup>36</sup>Cl/<sup>35</sup>Cl ratio is on the order of 10<sup>-14</sup> at.at<sup>-1</sup> (Calmet et al., 2001). All blanks  
 300 underwent chlorine extraction by alkaline fusion, and exhibit <sup>36</sup>Cl/<sup>35</sup>Cl isotopic ratios similar to those  
 301 commonly measured for process blanks at ASTER (Bouchez et al., 2015; Braucher et al., 2018; Finkel  
 302 et al., 2013; Pupier et al., 2016). This indicates that the contamination in <sup>36</sup>Cl or Cl during the during  
 303 the chemical preparation of our samples is negligible. Natural chlorine extraction and measurement in  
 304 the peach leaf analytical standards (n=4) allow calculating a mean extraction efficiency of 83 ± 2%,  
 305 confirming that alkaline fusion efficiently extracts chlorides from solid matrices (Table 4).

306 Table 4 : Chlorine extraction efficiency of the NIST peach leaf standard SRM 1547\*

Cl (mg.g <sup>-1</sup> )	Δ Cl (mg.g <sup>-1</sup> )	Yield
0,298	0,027	83%
0,292	0,027	81%
0,326	0,012	90%
0,280	0,006	78%

307 \*Chlorine level = 0.361 ± 0.014 mg.g<sup>-1</sup>

308

#### 309 3-2 Chlorine 36 in the environment

310 Table 5 lists the chlorine 36/total chlorine (<sup>36</sup>Cl/Cl<sub>t</sub>) calculated isotopic ratios for each sample from the  
 311 AMS measurements and provide the mean krypton activity over the period covered by the  
 312 corresponding sample.

313

314 Table 5: Mean krypton activity (Bq.m<sup>-3</sup>) and <sup>36</sup>Cl/Cl<sub>t</sub> isotopic ratio of the samples (10<sup>-14</sup>)

Campaigns	Krypton (Bq.m <sup>-3</sup> )	<sup>36</sup> Cl/Cl <sub>t</sub> (10 <sup>-14</sup> )			
		Gaz	Particulate	Grass blank	Grass sample
C1	0.6	39.1 ± 2.2	86.6 ± 7.0	2.4 ± 0.4	5,7± 0.9
C2	408.9	346.8 ± 19.1	209.4 ± 10.2	2.2 ± 0.4	8.0 ± 1.1

C3	194.0	415.7 ± 17.6	352.4 ± 14.2	2.5 ± 0.4	14.5 ± 3.8
C4	789.3	735.8 ± 41.6	317.0 ± 16.6	2.5 ± 0.4	11.2 ± 1.7
C5	345.1	993.0 ± 52.0	212.2 ± 11.2	2.6 ± 0.4	16.1 ± 2.8
C6	840.4	972.3 ± 39.2	969.4 ± 36.1	3.6 ± 0.6	28.4 ± 4.1
C7	574.3	1104.1 ± 44.3	334.4 ± 13.2	19.6 ± 3.2	40.4 ± 7.1
C8	451.1	770.3 ± 30.8	268.2 ± 10.9	4.7 ± 0.6	23.5 ± 3.3
C9	188.4	228.8 ± 9.9	104.0 ± 4.4	32.5 ± 3.4	35.4 ± 4.5
C10	1782.5	5019.3 ± 244.6	1636.6 ± 81.5	3.5 ± 0.5	76.7 ± 9.9
C11	68.7	122.3 ± 6.9	361.4 ± 19.1	3.3 ± 0.4	5.2 ± 0.8
C12	505.4	694.7 ± 37.4	161.8 ± 9.7	2.2 ± 0.4	7.7 ± 1.1
C13	4155.5	1612.4 ± 84.5	449.2 ± 23.4	2.3 ± 0.4	7.6 ± 1.0
C14	2080.2	1732.2 ± 87.1	1310.2 ± 60.6	2.6 ± 0.4	12.5 ± 1.9

315

### 316 3-2-1 Atmospheric chlorine 36

317 The  $^{36}\text{Cl}/\text{Cl}_t$  isotopic ratio in the atmosphere is between  $4 \times 10^{-13}$  and  $1.7 \times 10^{-11}$  at.at<sup>-1</sup> for gaseous  
318 fraction and between  $8.7 \times 10^{-13}$  and  $1.6 \times 10^{-11}$  at.at<sup>-1</sup> for the particulate fraction. These isotopic ratios are  
319 1-2 orders of magnitude greater than that of the natural atmospheric  $^{36}\text{Cl}/\text{Cl}_t$  ratio estimated to be  $7 \times 10^{-13}$   
320 at.at<sup>-1</sup> (Phillips et al., 1988). However, it must be kept in mind that the natural  $^{36}\text{Cl}/\text{Cl}_t$  isotopic ratio  
321 is between  $10^{-15}$  and  $10^{-12}$  at.at<sup>-1</sup> and varies with the geographical location and the stable chlorine  
322 content in the aera (Bastviken et al., 2013; Daillant et al., 2009; Lazarev, 2003). Since our study site is  
323 close to the sea and therefore subjected to stable chlorine marine inputs, this would yield ratio close  
324 the low range thus an  $^{36}\text{Cl}/\text{Cl}_t$  isotopic ratios reaching  $10^{-11}$  strongly suggest chlorine 36 input from a  
325 non-natural source in our study area. These ratios are similar to those found in the soils of on a site in  
326 Japan which housed four nuclear reactors and one nuclear fuel reprocessing plant (Bastviken et al.,  
327 2013). In our study, the atmospheric chlorine 36 concentrations varied from  $3 \pm 0.5$  to  $165.3 \pm 9$   
328 nBq.m<sup>-3</sup> (average =  $50 \pm 2.8$  nBq.m<sup>-3</sup>) for the gaseous fraction and  $0.5 \pm 0.03$  to  $83.2 \pm 2.2$  nBq.m<sup>-3</sup>  
329 (average =  $19 \pm 0.5$  nBq.m<sup>-3</sup>) for the particulate fraction (Fig. 2). When the chlorine 36 contents are  
330 related to the krypton 85 ( $^{85}\text{Kr}$ ) levels, it is noticed that the campaigns with high mean krypton 85  
331 contents exhibit the highest levels of gaseous and particulate chlorine 36. However, the correlation  
332 between krypton and chlorine 36 remains low for the gaseous fraction ( $r^2 = 0.40$ ) and even more so for  
333 the particulate fraction ( $r^2 = 0.25$ ). (Fig. 3)

334 Two reasons can explain this low correlation: firstly, the high reactivity of chlorine once released into  
335 the atmosphere, unlike krypton which is only subjected to dispersion. Secondly, because chlorine is  
336 washed out by rain when the plume is crossed by rainfall before it reaches the study site (Appendix  
337 C). The low correlation of krypton with particulate chlorine 36 is not surprising since it is not directly  
338 released via the plant stacks, unlike gaseous chlorine 36.

339 The partition between the gaseous and particulate fractions is an important input for understanding  
340 deposition processes, so it was determined for each of the campaigns. The  $^{36}\text{Cl}_{\text{gaseous}} / ^{36}\text{Cl}_{\text{particulate}}$  ratio  
341 is an average of 3, which corresponds to the expectations for measuring the near field radionuclides of  
342 the source term (Sportisse, 2007). This  $^{36}\text{Cl}_{\text{gaseous}} / ^{36}\text{Cl}_{\text{particulate}}$  ratio rises to 10 for campaign 4 and  
343 remains greater than or equal to 2 for all the campaigns, except for campaign 11 where this ratio is 0.5.  
344 Atmospheric chlorine 36 on our study site is therefore mostly present in gaseous form. This result is  
345 similar to the conclusions of Bah (2021) on the partition between the gaseous and particulate fractions  
346 of iodine 129 ( $^{129}\text{I}$ ) on this site. However, chlorine 36 levels remain less than those of iodine 129,  
347 which were between 5 and 110  $\mu\text{Bq.m}^{-3}$  for the gaseous fraction and between 0.8 and 27  $\mu\text{Bq.m}^{-3}$  for  
348 the particulate fraction (Bah, 2021).

349

### 350 **3-2-2 Chlorine 36 in grass**

351 The  $^{36}\text{Cl}/\text{Cl}_t$  isotopic ratio of the grass blanks was between  $2 \times 10^{-14}$  and  $3 \times 10^{-13}$  at.at<sup>-1</sup> while that of the  
352 grass samples harvested at the end of the campaigns were between  $5 \times 10^{-14}$  and  $7 \times 10^{-13}$  at.at<sup>-1</sup>.  
353 Generally, except for campaigns 7 and 9, the grass blanks had an  $^{36}\text{Cl}/\text{Cl}_t$  isotopic ratio on the order of  
354  $10^{-14}$  at.at<sup>-1</sup> similar to that of the natural background noise for plants (Calmet et al., 2001). The chlorine  
355 36 concentrations in the grass varied from  $0.2 \pm 0.04$  to  $4.5 \pm 0.09$   $\mu\text{Bq.g}^{-1}$  (mean =  $1 \pm 0.04$   $\mu\text{Bq.g}^{-1}$ )  
356 for the grass blanks and from  $0.7 \pm 0.09$  to  $10 \pm 0.21$   $\mu\text{Bq.g}^{-1}$  (mean =  $4 \pm 0.26$   $\mu\text{Bq.g}^{-1}$ ) for the grass  
357 samples (Fig. 4).

358 Even if the chlorine 36 levels are higher in the grass blanks for campaigns 7 and 9, the chlorine 36  
359 content measured in the grass at the end of the campaign always remained higher. The dry deposition  
360 fluxes of chlorine 36 on grass can then be calculated and the deposition velocities deduced from this.  
361 This increase in chlorine 36 content during the campaign is only related to the absorption of  
362 atmospheric chlorine 36, because the grass grows in aluminum tubs without contact with the study site  
363 ground. Indeed, studies showed that the leaf absorption of chlorine was rapid and significant,  
364 especially when the root inputs are restricted (Le Dizès and Gonze, 2019). The chlorine 36 levels  
365 measured in the grass samples are similar to those determined in the same region in grass ( $5.5$   $\mu\text{Bq.g}^{-1}$ )  
366 <sup>1</sup>). However, they remain higher than the chlorine 36 contents measured in vegetables ( $0.18$   $\mu\text{Bq.g}^{-1}$ ),  
367 oats ( $0.77$   $\mu\text{Bq.g}^{-1}$ ), barley ( $0.57$   $\mu\text{Bq.g}^{-1}$ ) and wheat ( $0.62$   $\mu\text{Bq.g}^{-1}$ ) (Calmet et al., 2001).

368

### 369 **3-3 Chlorine 36 dry deposition fluxes and velocities on grass**

#### 370 **3-3-1 Dry deposition fluxes**

371 Anthropogenic chlorine 36 dry deposition fluxes after nuclear tests are poorly documented. This is  
372 explained by the fact that they are generally considered negligible compared to wet deposition fluxes  
373 and complex to determine on an experimental basis. In our study, chlorine 36 dry deposition fluxes on  
374 grass were between  $1 \times 10^{-2}$  and  $4 \times 10^{-1}$   $\text{nBq.m}^{-2}\text{s}^{-1}$  (Fig. 5).

375 As the deposition flux variability between the campaigns is related to chlorine 36 capture by the grass,  
376 the factors influencing this deposition are chlorine availability in the atmosphere but also the  
377 meteorological and seasonal conditions influencing this capture (Spicer and Fox, 2021).

378 In the following, the experimental wet deposition fluxes taken from the literature will be considered as  
379 total depositions, because very often water collectors are subjected to dry depositions outside rainy  
380 periods. The dry deposition fluxes on grass determined in our study are far greater than the  
381 cosmogenic chlorine 36 global deposition flux, which is about  $1.7 \times 10^{-3}$   $\text{nBq.m}^{-2}\text{s}^{-1}$  (Poluianov et al.,  
382 2016). Note that the natural (dry and wet) chlorine 36 deposition flux varies with the season and the  
383 geographical position, particularly latitude. The highest values are observed in tropical areas with  
384 average calculated fluxes around  $6 \times 10^{-3}$   $\text{nBq.m}^{-2}\text{s}^{-1}$ , whereas they average around  $4.1 \times 10^{-3}$   $\text{nBq.m}^{-2}\text{s}^{-1}$   
385 for polar areas (Masarik and Beer, 2009; Poluianov et al., 2016). However, the chlorine 36 deposition  
386 fluxes measured are generally higher than the simulations for cosmogenic production (Scheffel et al.,  
387 1999). In France, a study measured the chlorine 36 total deposition flux on rain samples from a site  
388 located at the same latitude than the one of our study site. The chlorine 36 deposition fluxes were  
389 between  $3.2 \times 10^{-3}$  and  $2.8 \times 10^{-2}$   $\text{nBq.m}^{-2}\text{s}^{-1}$  (Pupier et al., 2016). The mean dry deposition flux  
390 determined in the present study is  $1.7 \times 10^{-1}$   $\text{nBq.m}^{-2}\text{s}^{-1}$ , which is higher than the wet deposition fluxes  
391 usually measured by previous studies (Keywood et al., 1998; Scheffel et al., 1999). Nonetheless, they  
392 remain lower than the deposition flux induced by the bomb, which was about  $7.3 \times 10^{-1}$   $\text{nBq.m}^{-2}\text{s}^{-1}$   
393 during the following decade (Heikkilä et al., 2009).

394

395 **3-3-2 Experimental deposition velocities**

396 Deposition velocity reflects the velocity at which an element is transferred from the atmosphere to the  
 397 plant either through absorption (gases), or adsorption (gases and particles). It depends on the substrate,  
 398 but also on parameters related to site meteorology and atmospheric turbulence (Hearn et al., 2013).  
 399 The total deposition velocities of chlorine 36 determined in the present study are between  $1 \times 10^{-3}$  and  
 400  $6 \times 10^{-3} \text{ m.s}^{-1}$  with a mean velocity of  $3 \times 10^{-3} \text{ m.s}^{-1}$  (Fig. 6).

401 Nonetheless, given that the chlorine depositions are related to seasonality, it seems wiser to calculate  
 402 mean values for each season. The mean deposition velocities per season are  $4.6 \times 10^{-3}$ ,  $4.4 \times 10^{-3}$  and  
 403  $1.4 \times 10^{-3} \text{ m.s}^{-1}$  for summer, fall and winter, respectively. No campaign was performed during spring, so  
 404 no mean velocity can be determined for this season. In the literature, the stable chlorine deposition  
 405 velocities for terrestrial ecosystems are primarily documented for hydrogen chloride. According to the  
 406 work done by Harrison et al., (1989), the hydrogen chloride deposition velocities on grass are between  
 407  $4.3 \times 10^{-3}$  and  $2 \times 10^{-2} \text{ m.s}^{-1}$ , in line with the estimations of Spicer and Fox, (2021) which were between  
 408  $3 \times 10^{-3}$  and  $3.2 \times 10^{-2} \text{ m.s}^{-1}$ . This study estimates the molecular chlorine deposition velocities at between  
 409  $2 \times 10^{-3}$  and  $6.3 \times 10^{-2} \text{ m.s}^{-1}$  on the ground and between  $1.8 \times 10^{-2}$  and  $2.1 \times 10^{-2} \text{ m.s}^{-1}$  on alfalfa (Sehmel,  
 410 1980). The deposition velocity of particulate chlorine is said to have a mean value of  $3.6 \times 10^{-3} \text{ m.s}^{-1}$   
 411 (Anatolaki and Tsitouridou, 2007). The chlorine 36 velocities obtained in the present study are not  
 412 comparable with those taken from the literature, as the latter relate either to gaseous chlorine or to  
 413 particulate chlorine. However, the velocities determined in the present study are the total of the  
 414 chlorine 36 gaseous chlorine and particulate chlorine velocities, with both fractions measured in the  
 415 atmosphere. From a radio-ecological standpoint, the chlorine 36 dry deposition velocities on grass  
 416 remain comparable with those of the widely studied radionuclides, particularly iodine 131 (0.26 – 0.57  
 417  $\text{cm.s}^{-1}$ ), iodine 129 (0.12-0.36  $\text{cm.s}^{-1}$ ), caesium 137 (0.2-0.5  $\text{cm.s}^{-1}$ ), caesium 134 (0.21-0.48  $\text{cm.s}^{-1}$ )  
 418 and caesium 136 (0.25-0.54  $\text{cm.s}^{-1}$ ) (Bah, 2021; Masanori and Shuichi, 2014). As chlorine is an  
 419 element spread widely across the surface of the globe and essential for living organisms, the chlorine  
 420 36 dry deposition velocities obtained in this way will help to provide better understanding of the  
 421 mechanisms of transfer to plants. More generally, they will help to improve the characterization of the  
 422 transfers of radionuclides to the environment and to man. The chlorine 36 dry deposition velocities  
 423 obtained in this study will also allow better evaluation of the impact studies in the event of nuclear  
 424 power plant decommissioning or accident for better integration of the dosimetric impact and reduction  
 425 of the related uncertainties.

426  
 427 **3-4 Dry deposition models**

428 **Table 6** gives the mean meteorological and micro-meteorological data recorded throughout each of  
 429 the campaigns. The frequency of acquisition of these data was 30 min. These data correspond to the  
 430 input data for the particulate and gaseous chlorine 36 deposition models. Accordingly, particulate and  
 431 gaseous chlorine 36 deposition velocities were determined for each 30 min time plot. Based on the  
 432 concentrations measured in the atmosphere, quantities of chlorine 36 deposited on grass were  
 433 calculated for each 30 min time plot and for each of the fractions. Given that during precipitation  
 434 periods the atmospheric concentrations are reduced by the scavenging effect, the chlorine 36  
 435 atmospheric concentrations during rain are set at 0.

436  
 437 Table 6: Meteorological and micro-meteorological parameters of the campaigns

Campaign	$u^*$ ( $\text{m.s}^{-1}$ )	Atmospheric stability			Relative humidity	Solar radiations ( $\text{W.m}^{-2}$ )	Temperature ( $^{\circ}\text{C}$ )	Dew point ( $^{\circ}\text{C}$ )
		%stable	%neutral	%instable				

C1	0,36	21,1%	48,0%	30,9%	87,7%	271,9	18,5	15,8
C2	0,53	15,7%	63,6%	20,7%	92,1%	238,4	15,6	13,8
C3	0,44	9,7%	82,2%	8,1%	89,9%	195,1	16,5	14,3
C4	0,34	31,4%	30,6%	38,0%	91,1%	150,2	16,5	14,6
C5	0,58	11,7%	82,2%	6,1%	87,1%	131,2	14,4	11,7
C6	0,32	29,9%	61,1%	9,0%	90,4%	105,5	13,6	11,5
C7	0,60	8,7%	90,6%	0,7%	86,4%	66,8	11,8	9,1
C8	0,41	11,5%	85,0%	3,5%	88,3%	39	10,7	8,3
C9	0,68	25,4%	74,6%	0,0%	82,6%	27	7,5	4,2
C10	0,56	8,0%	91,1%	0,9%	91,6%	27,2	8,2	6,4
C11	0,45	11,4%	85,9%	2,7%	89,8%	40,8	6,7	4,7
C12	0,73	3,5%	95,2%	1,3%	89,2%	58,1	8,2	6,0
C13	0,57	21,0%	70,0%	9,0%	84,2%	97,2	7,3	4,2
C14	0,52	17,1%	76,2%	6,7%	87,3%	116,8	9,1	6,5

438

### 439 3-4-1 Particulate chlorine 36 dry deposition model

440 The stable chlorine contents measured in the atmospheric aerosol on the study site are shown in [Fig. 7](#).

441 The particle size range for which chlorine is most present on the study site is therefore between 0.32  
442 and 0.77  $\mu\text{m}$ . To define the particle size most representative of this range in the <<Damay-Pellerin>>  
443 model, a geometrical particle diameter of 0.48  $\mu\text{m}$  was defined in the model. The A and B parameters  
444 determined for this particle size are respectively  $1.6 \times 10^{-3}$  and -11 (Damay et al., 2009; Pellerin et al.,  
445 2017). The dry deposition velocities of particulate chlorine 36 calculated from the micro-  
446 meteorological data are between  $5.9 \times 10^{-5}$  and  $8.82 \times 10^{-4} \text{ m.s}^{-1}$  (mean =  $1.8 \times 10^{-4} \text{ m.s}^{-1}$ ) ([Fig. 8](#)).

447 The mean deposition velocity calculated by the model is similar to that defined as reference deposition  
448 velocity for the accumulation mode aerosols, i.e.  $1 \times 10^{-4} \text{ m.s}^{-1}$  (Sportisse, 2007). The deposition  
449 velocities on grass calculated for this particle size are less than those calculated for depositions on  
450 corn, which were between  $1.22 \times 10^{-4}$  and  $6.79 \times 10^{-3} \text{ m.s}^{-1}$  (Damay et al., 2009). This highlights the  
451 influence of the nature and properties of the plant cover on particle deposition velocities. However, the  
452 parameters most influencing the particle deposition velocity are those relating to micro-meteorology  
453 and atmospheric turbulence. These include wind friction velocity  $u^*$  and atmospheric stability,  
454 calculated from sensible heat flux H (Pellerin et al., 2017). Deposition velocity is closely related to  
455  $u^*$ , so particulate chlorine 36 depositions are greater for higher  $u^*$  values. It was nonetheless shown  
456 that periods of atmospheric instability induce greater deposition velocities (Connan et al., 2018). This  
457 explains the deposition velocities calculated for campaigns 1 and 4, which are one order of magnitude  
458 above that of the other campaigns. Indeed, the atmosphere was unstable for about 30% of the time  
459 during these campaigns ([Table 6](#)). The quantity of particulate chlorine 36 deposited on grass during a  
460 campaign nonetheless remains dependent on the particulate chlorine 36 atmospheric concentration.

### 461 3-4-2 Gaseous chlorine 36 dry deposition model

462 The mean values of resistances  $R_a$ ,  $R_b$  and  $R_c$  used to calculate the deposition velocities are  
463 represented in [Fig. 9](#). The gaseous chlorine 36 deposition velocities calculated by the model are  
464 between  $3.4 \times 10^{-4}$  and  $4.8 \times 10^{-3} \text{ m.s}^{-1}$  (mean =  $2.3 \times 10^{-3} \text{ m.s}^{-1}$ ). These deposition velocities are higher in  
465 summer, reduced during the fall and lower in winter ([Fig. 8](#)). This is related to the opening of the  
466 stomata during the summer, which reduces the stomatal resistance  $R_{st}$  and therefore the surface  
467 resistance. These stomata gradually close during the fall and are totally closed in winter, hence the  
468 increase in canopy resistance (Zhang et al., 2002b). Accordingly, outside summer and spring, when  
469 the stomata are fully open, the deposition velocity on grass primarily depends on non-stomatal

470 resistance ( $R_{ns}$ ). Further, the highest values of the cuticle ( $R_{cut}$ ) and ground ( $R_g$ ) resistances occur in  
471 winter, which even further increases the  $R_{ns}$  values. Given that molecular chlorine is more soluble  
472 than ozone, the chlorine specific  $R_{cut}$  and  $R_g$  values should be lower. As a result, the deposition  
473 velocities calculated by the model are slightly underpredicted. Indeed, a study of gas depositions on  
474 alfalfa showed that dichloride was deposited more rapidly than ozone (Hill, 1971).

475 Apart from the season, the parameters most influencing the gas deposition velocity are the wind  
476 friction velocity ( $u^*$ ) and the leaf area index (LAI) (Zhang et al., 2002b). The work carried out by Bah  
477 et al., 2020 indeed showed the close correlation between  $R_c$  and the gas deposition velocity ( $V_d$ ). This  
478 means that higher  $u^*$  values entail greater deposition velocities. However, the  $R_{cut}$  and  $R_g$  values used  
479 in this study were estimated based on those for ozone and these values were determined according to  
480 the season. This slightly reduces the influence of  $u^*$  on the  $V_d$  of the model because, for the  $R_{ns}$   
481 calculation,  $u^*$  is only a factor in the  $R_{ac}$  calculation. Regarding LAI, the larger it becomes, the more  
482 the uptake of gas by the plant is increased. This is related to the increase in the capture area, but also to  
483 the increased number of stomata. So, for identical meteorological conditions, the gas deposition  
484 velocities on superior plants will be greater than on grass (Sehmel, 1980).

485

### 486 3-4-3 Comparison of model and measured depositions velocities

487 The total deposition velocities ( $V_d$ ) calculated with the previous models are between  $3.48 \times 10^{-4}$  and  
488  $4.88 \times 10^{-3} \text{ m.s}^{-1}$  (Fig. 10). Overall, the  $V_d$  values calculated by the model ( $V_{d \text{ mod}}$ ) differ by less than  
489 one order of magnitude from the  $V_d$  values obtained experimentally ( $V_{d \text{ exp}}$ ). Without considering the  
490 uncertainties on the  $V_{d \text{ exp}}$ , the largest differences were observed in campaigns 7, 8, 9 and 11, with  $V_d$   
491  $\text{exp} / V_{d \text{ mod}}$  ratios of 2, 4, 3 and 5 respectively. For the other campaigns, the  $V_{d \text{ mod}}$  values show a  
492 difference of less than a factor of 2 from the  $V_{d \text{ exp}}$  and are within the uncertainty zone. The  $V_{d \text{ mod}}$   
493 values vary little within the same season, unlike the  $V_{d \text{ exp}}$ . The result is therefore a low correlation ( $R^2$   
494 = 0.3) between these two velocities. However, this correlation increases when the uncertainties of the  
495 experimental  $V_d$  values are integrated ( $R^2 = 0.7$ ).

496 The high  $V_{d \text{ exp}}/V_{d \text{ mod}}$  ratio observed in campaign 11 can be explained by variability in the particulate  
497 chlorine 36 diameter in the medium. This diameter was in fact fixed in the model following a single  
498 measurement which may not reflect the aerosol profile of the study site over the whole range of  
499 campaigns. Campaign 11 is indeed the only campaign where the particulate chlorine 36 concentration  
500 is greater than that of the gaseous fraction, making the deposition velocity of the model more sensitive  
501 to that of the particles. Unlike  $V_{d \text{ mod}}$ , the  $V_{d \text{ exp}}$  of campaigns 7 to 9 are quite high with respect to the  
502 season and the state of closure of the stomata for this period. Campaigns 7 and 9 feature large  
503 uncertainties on the experimental deposition velocities arising from an already high chlorine 36  
504 content in the grass blank. Their experimental deposition velocities could be lower and therefore  
505 closer to the model predictions. Part of the high  $V_{d \text{ exp}}/V_{d \text{ mod}}$  ratio in campaign 8 can be explained by  
506 the micro-meteorological conditions of this campaign. Indeed, very low  $u^*$  values in a stable  
507 atmosphere lead to very high canopy resistances, reducing the deposition velocities calculated by the  
508 gas model (Leiming Zhang et al., 2003). Obviously, this does not explain the size of the difference.  
509 Apart from the meteorological and micro-meteorological parameters, the factor liable to increase  
510 chlorine uptake by the plant is leaf surface wetness. Indeed, it has been proved that a wet leaf surface  
511 considerably reduced surface resistance, increasing gas deposition (Leiming Zhang et al., 2003). As it  
512 happens, under certain conditions water droplets may form on the grass surface even if the ambient  
513 temperature is greater than the dew point temperature. During the night, when relative humidity is  
514 high (>95%), the solid parts close to the ground may have a temperature lower than that of air, causing  
515 water to condense on their surfaces. It cannot be ruled out that the leaf surface was wetted at some  
516 time during campaign 8, keeping in mind that these conditions were observed just before the krypton  
517 85 peak on the study site. The chlorine 36 uptake may therefore have been more significant at this  
518 time, explaining the higher deposition velocity. Nonetheless, it must be remembered that dry

519 deposition is a complex process affected by the meteorological conditions, biological factors and  
 520 physical-chemical properties of the species under consideration. All these factors are sources of  
 521 uncertainty, making it difficult to perfectly model the depositions (Leiming Zhang et al., 2003). Even  
 522 for homogeneous atmospheric and surface conditions, a bad description of the plant cover properties  
 523 or plant hydric stress may lead to large differences between the measured and observed deposition  
 524 velocities (Wesely and Hicks, 2000). Despite these uncertainties, the model developed in this study  
 525 provides an accurate estimate of the chlorine 36 dry deposition velocities on grass, based on  
 526 atmospheric chlorine 36 concentrations and the meteorological and micro-meteorological parameters.

527

### 528 **3-4-4 Comparison of gaseous and particulate chlorine 36 deposition fluxes**

529 With the deposition fluxes experimentally determined in this study, it is not possible to discriminate  
 530 between the input due to gaseous chlorine 36 and that resulting from particulate chlorine 36. The  
 531 calculated  $V_{d\text{ mod}}$  are of the same order of magnitude as the  $V_{d\text{ exp}}$ , so the deposition models can be used  
 532 to evaluate the depositions for each fraction. It was observed that for the whole set of campaigns, the  
 533 gaseous chlorine 36 depositions represent 97% on average, against 3% for particulate chlorine 36  
 534 (Table 7). This is not surprising, given that the gaseous chlorine 36 concentrations are much higher in  
 535 the atmosphere and the  $V_d$  calculated by the model are one order of magnitude above those calculated  
 536 for the particles. Additionally, the diffusivity of the gases is greater than that of the particles (Telly  
 537 Bah et al., 2020). On this basis, the gas deposition model alone can supply a good estimate of the total  
 538 chlorine 36 deposition on grass.

539 Table 7: Experimental dry deposition fluxes of chlorine 36 on grass compared with the deposition  
 540 fluxes calculated by the model

Campaign	Experimental deposits of $^{36}\text{Cl}$ ( $\mu\text{Bq.m}^2$ )	Model		
		Modelled deposits of $^{36}\text{Cl}$ ( $\mu\text{Bq.m}^2$ )	% Gaz	% Particulate
C1	16.5 ± 7.0	12.4	96.8%	3.2%
C2	151.9 ± 27.5	135.4	99.2%	0.8%
C3	419.6 ± 103.0	320.5	95.9%	4.1%
C4	228.8 ± 37.8	245.7	98.0%	2.0%
C5	209.8 ± 44.2	254.9	99.6%	0.4%
C6	457.9 ± 66.1	486.8	99.8%	0.2%
C7	369.3 ± 169.1	135.1	98.8%	1.2%
C8	332.8 ± 52.6	78.5	99.3%	0.7%
C9	111.5 ± 78.8	29.7	98.0%	2.0%
C10	1303.7 ± 344.7	729.9	96.4%	3.6%
C11	41.1 ± 23.6	8.0	88.3%	11.7%
C12	88.1 ± 35.0	55.8	95.8%	4.2%
C13	126.9 ± 43.2	120.9	98.1%	1.9%
C14	163.3 ± 55.6	122.0	96.3%	3.7%

541

542

### 543 **4-Conclusions**

544 In this study, we determined dry deposition velocities of chlorine 36 on grass for the first time.  
 545 Through 14 experimental campaigns conducted downwind from the Orano La-Hague plant, chlorine  
 546 36 was characterized in the atmosphere and its transfers to grass quantified. The dry deposition  
 547 velocities on grass were between  $1 \times 10^{-3}$  and  $6 \times 10^{-3}$  m.s<sup>-1</sup> and the highest values for the campaigns were



548 recorded during the summer. Atmospheric chlorine uptake by the grass was very high, even if levels  
549 remained well below the regulatory thresholds. Our results showed that these deposition velocities  
550 were dependent on the season, but also on meteorological and micro-meteorological conditions. The  
551 chlorine 36 dry deposition model proposed in this study provides a good estimate of deposition  
552 velocity but requires improvement to reduce the differences with the observed velocities.  
553 Nonetheless, the chlorine 36 dry deposition fluxes and velocities determined in this study will help to  
554 improve knowledge on the fate of chlorine 36 and its transfer to the environment.

555

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559

560

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