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1 **First measurements of chlorine 36 (³⁶Cl) wet deposition fluxes in the vicinity**
2 **of Orano La Hague nuclear reprocessing plant in France.**

3
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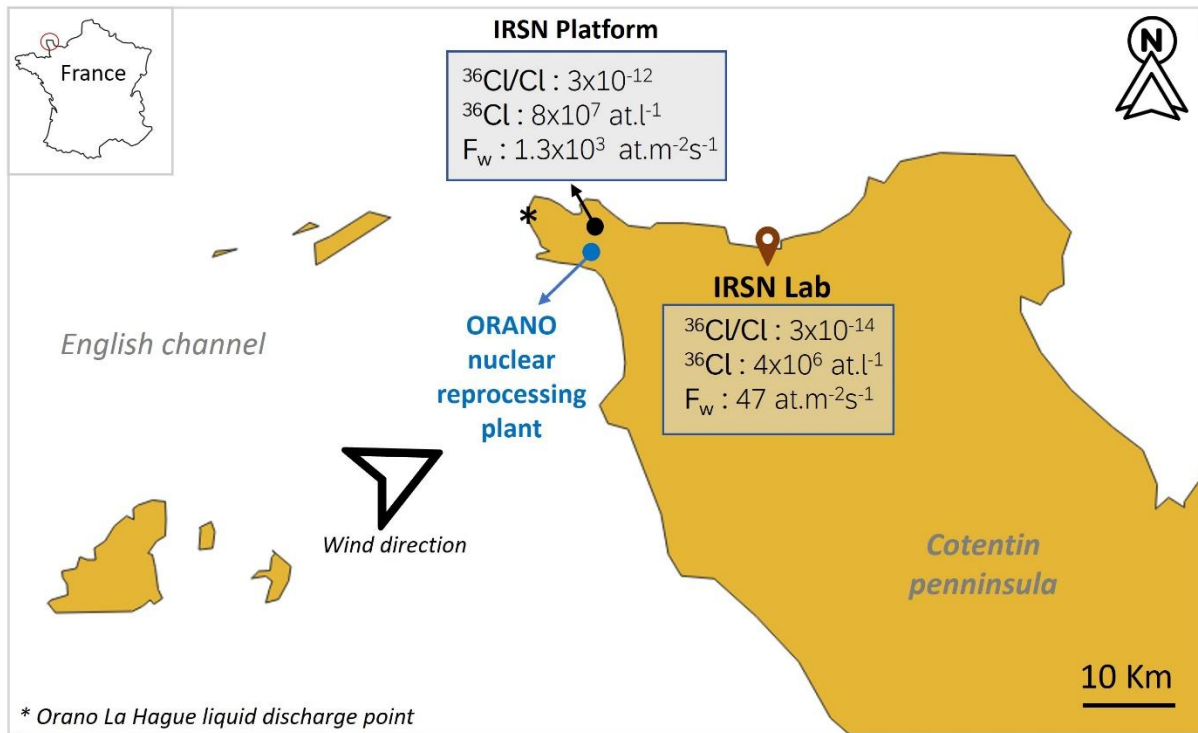
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32 Engineering for their support in this work.

39 **Graphical abstract :**



40

41

42 **Summary:**

43 Chlorine 36 (^{36}Cl) is a radionuclide of natural and anthropogenic origin, mainly used as a tracer in
44 geochemical studies. Owing to analytical constraints and its low environmental levels, knowledge of
45 ^{36}Cl behavior in the environment is still very limited. In this study, we use environmental
46 measurements to report for the first time the wet deposition fluxes of ^{36}Cl downwind an
47 anthropogenic source, the Orano nuclear reprocessing plant, which chronically emits ^{36}Cl into the
48 environment. Measurements of ^{36}Cl in rainwater samples at our study site were 1 to 2 orders of
49 magnitude above the environmental background. The isotope ratios $^{36}\text{Cl}/\text{Cl}$ of the samples and the
50 ^{36}Cl content in the rainwater averaged $2.3 \times 10^{-12} \text{ at.at}^{-1}$ and $1.7 \times 10^8 \text{ at.l}^{-1}$ respectively. A decrease in
51 these levels was observed 20 km away from the study site, outside the plant's gas plume, indicating
52 that the marking of ^{36}Cl on the study site is related to the plant discharges. Over the sampling period,
53 wet deposition fluxes at the study site averaged $3.4 \times 10^3 \text{ at.m}^{-2}\text{s}^{-1}$, with significant values measured
54 when precipitations scavenge the plant's gas plume down onto our study site. Analysis of these
55 fluxes also revealed the presence of a significant rainout phenomenon in the study area. These
56 results provide new data on the wet deposition flux of ^{36}Cl and will thus enable better assessment of
57 impact studies in a context of decommissioning or accidents involving nuclear power plants.

58

59 **Key words:**

60 Chlorine 36; Wet deposits; Nuclear fuel recycling; Dismantling nuclear power plants; Accelerator
61 Mass Spectrometry (AMS); Transfer in the environment.

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63

64 1. Introduction

65 With natural and anthropogenic origin, chlorine 36 (^{36}Cl) is a radioactive isotope (or radionuclide) of
66 chlorine (Cl) with a lifetime of around $3.01 \pm 0.02 \times 10^5$ years (Phillips, 2000). It is a pure beta emitter
67 (β^-), with low energy ($E_{max} = 708.6$ keV) and low radiotoxicity (Bastviken et al., 2013). ^{36}Cl is a
68 cosmogenic radionuclide, meaning that it is naturally produced during nuclear interactions between
69 primary and secondary cosmic ray particles with target element constituents of the terrestrial
70 environment (Bastviken et al., 2013 ; Huggle et al., 1996 ; Stone et al., 1996 ; White and Broadley,
71 2001). ^{36}Cl was massively introduced into the environment by neutron activation of ^{35}Cl present in
72 the sea as a result of nuclear testing in the Pacific. The high density of the energy released by these
73 tests triggered major upward airflows, which carried some of the ^{36}Cl produced into the atmosphere.
74 This massive introduction of ^{36}Cl is readily observable in the ^{36}Cl measurements of ice cores, and the
75 amount of ^{36}Cl generated as a result of these tests is estimated at 1,000 years of natural production
76 (Green et al., 2004 ; Heikkilä et al., 2009). However, it has recently been shown that the Cl fluxes
77 induced by these tests have returned to natural levels in Europe (Johnston and McDermott, 2008).

78 Anthropogenic ^{36}Cl is currently produced in nuclear reactor cores by the activation of stable Cl
79 present as impurities in the fuel and in the plant's construction materials. ^{36}Cl is therefore released
80 into the atmosphere in the form of gaseous effluents during fuel reprocessing operations. ^{36}Cl is also
81 likely to be released into the atmosphere during the decommissioning of nuclear facilities (Le Dizès
82 and Gonze, 2019 ; Llopart-Babot et al., 2022 ; Petrov and Pokhitonov, 2020 ; Sheppard et al., 1996).
83 In abnormal or incidental situations (filters are assumed to be ineffective), ^{36}Cl can also be released
84 into the atmosphere in particulate form. Even when emitted in gaseous form, like other
85 radionuclides, ^{36}Cl can form ultrafine particles by nucleation or condense on atmospheric aerosols
86 present in the environment (Budyka, 2000). Finally, ^{36}Cl can be emitted into the atmosphere
87 following a nuclear accident [Chernobyl example (Lazarev, 2003 ; Scheffel et al., 1999) and
88 Fukushima (Ohta et al., 2023; Sasa et al., 2022)] or, on a smaller scale, as a result of a biomass
89 combustion contaminated by nuclear industry emissions (White and Broadley, 2001).

90 Whether produced naturally or anthropogenically, ^{36}Cl is rapidly eliminated from the atmosphere
91 within a few weeks by precipitation or dry deposition (Phillips et al., 1988 ; Scheffel et al., 1999 ;
92 Sourabie et al., 2023). Studies on ^{36}Cl have been made possible mainly by advances in accelerator
93 mass spectrometry, enabling environmental concentrations to be measured and the use of ^{36}Cl for
94 groundwater tracing (Braucher et al., 2018 ; Finkel et al., 2013 ; Merchel et al., 2011). Many studies
95 have been carried out on wet deposits of ^{36}Cl providing ^{36}Cl wet deposition flux measurements
96 around the globe (Johnston and McDermott, 2008 ; Pupier et al., 2016 ; Santos et al., 2004 ; Scheffel
97 et al., 1999 ; Tosaki et al., 2012). However, these studies mainly focused on naturally produced ^{36}Cl
98 and didn't discriminate wet depositions from dry ones. In fact, compared to wet deposition, dry
99 deposition of ^{36}Cl is generally considered as non-significant but a recent study showed that dry
100 deposition can contribute significantly to total ^{36}Cl deposition (Sourabie et al., 2023). In addition,
101 since chlorine is a micronutrient necessary for life, wet deposition can therefore be considered as the
102 deposition of ^{36}Cl in a bio assimilable form by living organisms.

103 The aim of this study is therefore to determine the only-wet deposition fluxes of ^{36}Cl in fields close to
104 the Orano La Hague plant, a chronic emitter of small quantities of ^{36}Cl into environment (GRNC,
105 1999). In fact, since Milton et al. (1994), no studies have been carried out on wet deposition of ^{36}Cl in
106 the vicinity of a chronic anthropogenic source. The isotopic ratio $^{36}\text{Cl}/\text{Cl}$ in the atmosphere (gaseous
107 and particulate fraction) 2 km away from the plant is 1 to 2 orders of magnitude higher than the
108 natural background, suggesting that wet deposition fluxes of ^{36}Cl in this area are greater than those
109 generally reported in the literature (Sourabie et al., 2023). Following the study of ^{36}Cl dry deposition

110 flux on this site, the determination of ^{36}Cl wet deposition flux will therefore help to improve the
111 assessment of impact studies in a context of nuclear power plant decommissioning or accidents.

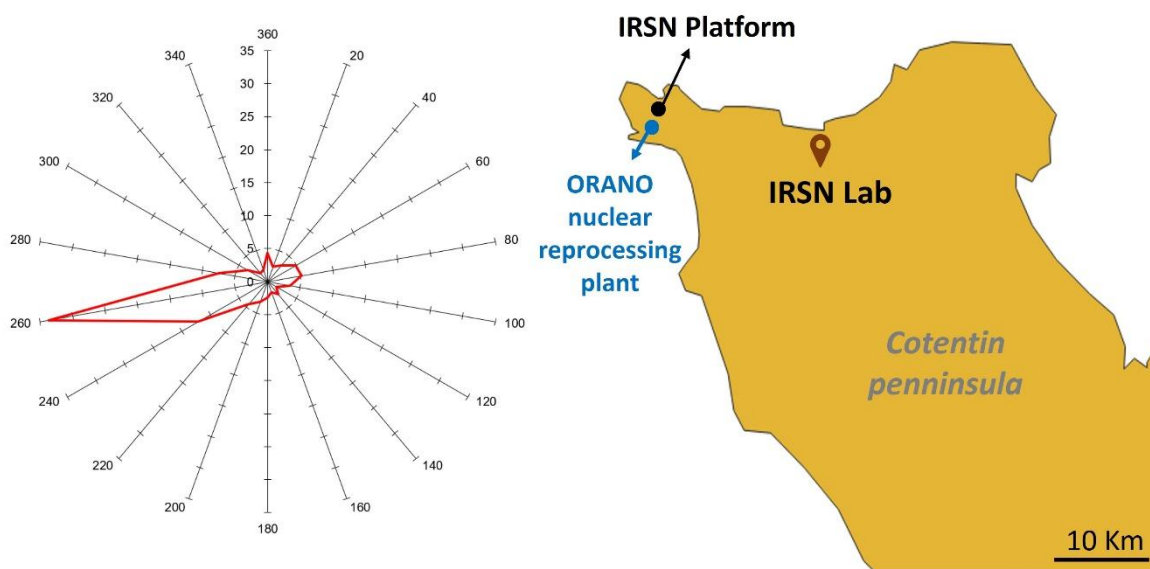
112

113 2. Materials and methods

114 2.1. Study site and sampling

115 Rainwater was sampled at the IRSN La Hague technical platform (IRSN Platform; $49^{\circ}41'42.1''\text{N}$,
116 $1^{\circ}52'24.2''\text{W}$) located 2 km north of Orano La Hague plant ($49^{\circ}40'41.5''\text{N}$, $1^{\circ}52'30.7''\text{W}$) in France and
117 2 km south of English Channel (**Figure 1**). ^{36}Cl is emitted concomitantly with krypton 85 (^{85}Kr), which
118 serves as a tracer for the plant's atmospheric plume (Connan et al., 2014). As such, ^{85}Kr is measured
119 continuously at the study site using a Berthold LB111 counter. Annual rainfall at the study site has
120 averaged 643 mm over the past 10 years, with heavier precipitation in autumn. Rainwater was
121 collected by an Eigenbrodt UNS130/E open-air rain collector equipped with an Eigenbrodt RS85
122 sensor, which only opens the water collector when precipitation was detected. This device ensures
123 that all ^{36}Cl measured in rainwater mainly comes from wet deposits. A Watchdog 2000 series
124 weather station was used to record rainfall at a frequency of 30 minutes throughout the sampling
125 period. Samples were taken over an average period of two weeks, from July 16, 2021 to July 26,
126 2022; with a break in sampling from December 09, 2021 to February 22, 2022 due to a shutdown of
127 the sampler.

128



129

130 **Fig. 1.** Wind direction frequencies from May 17 to June 1, 2022 measured at the IRSN Platform.

131 2.2. Sample processing and chlorine 36 measurement

132 Following sampling, the filtered samples underwent a series of chemical preparations to make them
133 compatible with ^{36}Cl measurement by accelerator mass spectrometry and eliminate isobaric
134 interferences such as sulfur 36 (Nakata and Hasegawa, 2011). Dissolved chlorine in rainwater is first
135 precipitated to silver chloride by adding 1 mL of silver nitrate AgNO_3 10% (analytical grade Emsure
136 ACS, Merck) and 20 mL of nitric acid HNO_3 70% (CMOS J.T.Baker, VWR). The precipitate is recovered
137 by filtration through a polyethersulfone filter (Millipore Express Plus, $0.45\mu\text{m}$, 47 mm, Merk) and
138 transferred to a tube. 10 mL of ammonia NH_4OH 20% (Emplura, Merck) is used to dissolve the
139 precipitate retained on the filter and 2 mL of saturated barium nitrate $\text{Ba}(\text{NO}_3)_2$ (analytical grade ACS,
140 Alfa Aesar) is added to the sample tube. The addition of barium causes sulfur to precipitate out as
141 barium sulfate (BaSO_3). The sample is filtered after 24 hours, then the operation is repeated twice
142 with 1 mL of barium nitrate. After the third filtration of barium sulfate, 8 mL of nitric acid are added
143 to the tube to re-precipitate dissolved chlorides. The samples are then cooled, shaken and
144 centrifuged. The acid is removed, and the precipitate washed 3 times with ultrapure water (Merck
145 Milli-Q, $18.2\text{ M}\Omega\text{cm}$ at 23°C) before being oven-dried at 40°C for 72h. After drying, the silver chloride
146 precipitates were sent for measurement of ^{36}Cl and Cl. This measurement is performed by
147 accelerator mass spectrometry (AMS) on the national instrument ASTER (Accelerator for earth
148 sciences, environment and risks) (Finkel et al., 2013). The ratios $^{36}\text{Cl}/^{35}\text{Cl}$ and $^{35}\text{Cl}/^{37}\text{Cl}$ were obtained
149 by normalization against the internal standard SM-CL-12 with an assigned $^{36}\text{Cl}/^{35}\text{Cl}$ value of $(1.428 \pm$
150 $0.021) \times 10^{-12}$ (Merchel et al., 2011) and assuming a natural $^{35}\text{Cl}/^{37}\text{Cl}$ ratio of 3.127. The ^{36}Cl and Cl
151 concentration of each sample is then calculated from the ratios $^{36}\text{Cl}/^{35}\text{Cl}$, $^{35}\text{Cl}/^{37}\text{Cl}$ and the mass of
152 doping solution (Na^{35}Cl ; $^{35}\text{Cl} = 99.66\%$, Chemlab) added to the sample. These calculations are
153 presented in detail in Bouchez et al. (2015).

154

155 **3. Results and discussion**

156 *3.1. Quality control*

157 To ensure the absence of contamination during sample preparation, reagent blanks were made. The
158 ratio $^{36}\text{Cl}/^{35}\text{Cl}$ of our blanks ($n = 3$) averaged 1.6×10^{-15} which is close to the ratio $^{36}\text{Cl}/^{35}\text{Cl}$ of blanks
159 measured by AMS at ASTER (1.3×10^{-15} ; Braucher et al., 2018). This allows us to ensure the absence of
160 ^{36}Cl contamination during the processing of our samples.

161

162 *3.2. Isotope ratios $^{36}\text{Cl}/\text{Cl}$ and ^{36}Cl content in rainwater*

163 **Table 1** shows the results of ^{36}Cl and Cl measurements on all our samples. The uncertainty
164 corresponds to the instrumental uncertainty, comprising the measurement uncertainty, the
165 uncertainty of the standard, the uncertainty of the mean of the calibration measurements and the
166 systematic error of the machine. No sampling was carried out from August 24, 2021 to September 7,
167 2021 and from April 20, 2022 to May 3, 2022 due to low rainfall.

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175 **Table 1:** ^{36}Cl measurements in rainwater samples from the study site; uncertainties is indicated by
 176 "±" in the table.

Campaign no.	Start date	End date	Rainfall	Cl^-	$^{36}\text{Cl}/\text{Cl}$	^{36}Cl
			<i>mm</i>	<i>mg.l⁻¹</i>	<i>x10⁻¹²</i>	<i>x10⁷ at.l⁻¹</i>
C1	16/07/2021	28/07/2021	56,1	3,0	2,4 ± 0,1	12,3 ± 1,7
C2	28/07/2021	09/08/2021	12,9	7,9	2,4 ± 0,1	32,3 ± 4,5
C3	09/08/2021	24/08/2021	7,5	4,6	1,1 ± 0,04	8,9 ± 1,3
C4	07/09/2021	21/09/2021	19,5	1,2	3,2 ± 0,1	6,8 ± 0,7
C5	21/09/2021	06/10/2021	74,6	11,0	0,7 ± 0,03	14,5 ± 2,9
C6	06/10/2021	19/10/2021	6,8	5,9	3,9 ± 0,1	39,2 ± 5,3
C7	19/10/2021	05/11/2021	80,2	13,2	0,3 ± 0,01	8,6 ± 0,9
C8	05/11/2021	23/11/2021	16,3	17,0	0,2 ± 0,01	6,2 ± 2,5
C9	23/11/2021	09/12/2021	86,2	13,1	0,5 ± 0,02	11,3 ± 1,2
C10	22/02/2022	08/03/2022	37,5	8,0	0,8 ± 0,04	11,6 ± 1,2
C11	08/03/2022	18/03/2022	13,9	3,1	5,4 ± 0,2	29,3 ± 3,2
C12	18/03/2022	05/04/2022	13,5	11,1	1,9 ± 0,1	36,8 ± 14,8
C13	05/04/2022	20/04/2022	24,3	7,6	0,7 ± 0,04	10,1 ± 1,1
C14	03/05/2022	17/05/2022	8,9	3,2	2,7 ± 0,1	15,3 ± 1,6
C15	17/05/2022	01/06/2022	21,5	1,3	3,8 ± 0,1	8,3 ± 0,9
C16	01/06/2022	15/06/2022	40,2	3,3	4,3 ± 0,2	24,4 ± 2,7
C17	15/06/2022	29/06/2022	17,8	6,4	2,4 ± 0,1	26,9 ± 2,9
C18	29/06/2022	13/07/2022	3,0	3,7	2,1 ± 0,1	13,4 ± 1,4
C19	13/07/2022	26/07/2022	6,7	1,8	5,3 ± 0,2	16,6 ± 2,3

177

178 Isotopic ratios $^{36}\text{Cl}/\text{Cl}$ in rainwater samples ranged from 2.1×10^{-13} to 5.4×10^{-12} with an average of
 179 2.3×10^{-12} (median = 2.4×10^{-12}). The $^{36}\text{Cl}/\text{Cl}$ isotopic ratio in rainwater is strongly influenced by the
 180 atmospheric Cl content of the aera. Since ^{36}Cl is mainly produced by cosmic rays, in inland areas the
 181 stable Cl content is so low that the $^{36}\text{Cl}/\text{Cl}$ ratio can reach 10^{-13} whereas it is 10^{-14} at the seaside
 182 (Johnston and McDermott, 2008). Despite the proximity of the sea 2 km from the study site, the
 183 $^{36}\text{Cl}/\text{Cl}$ isotope ratios measured in our samples are 1 to 2 orders of magnitude higher than those of
 184 the natural environmental background (10^{-13} - 10^{-14}), which is generally measured on sites far from
 185 anthropogenic sources of ^{36}Cl (**Table 2**). However, the $^{36}\text{Cl}/\text{Cl}$ ratio in our rainwater samples is of the
 186 same order of magnitude as those measured by Milton et al., (1994) neaby an anthropogenic source
 187 showing then the influence of Orano's plant discharges on the ratios measured. In fact, Milton et al.,
 188 (1994) measured the highest $^{36}\text{Cl}/\text{Cl}$ ratios reported in the literature at various distances from the
 189 Chalk River Nuclear Laboratory (CRL), with levels ranging from 1.6 to 30.8×10^{-12} . It is also quite
 190 interresting to notice that the $^{36}\text{Cl}/\text{Cl}$ ratio in our rainwater samples is of the same order of
 191 magnitude as the ratio of 1.4×10^{-12} measured by Sasa et al., (2022) in a monthly rainwater sample
 192 from Tsukuba during the month of the Fukushima nuclear power plant accident. Despite the distance
 193 of around 180 km from the accident site, this ratio was two orders of magnitude higher than those
 194 measured in monthly rainwater samples before and after this event (Sasa et al., 2022).

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Table 2: ³⁶Cl measurements in rainwater reported in the literature.

Date	Site	³⁶ Cl/Cl (x10 ⁻¹⁵)	at.l ⁻¹ (x10 ⁶)	Source
1979	La Jolla, United States	58	2.5	(Finkel et al., 1980)
1988	Several sites in Israel	1.8 - 50	0.34 - 125	(Herut et al., 1992)
1991-1992	St. Mary's County, United States	3.8 - 568	1.2 (average)	(Hainsworth et al., 1994)
1999-1993	Several sites around Chalk River Nuclear Laboratories, Canada*	(1.6 – 30.8) x10 ³	9.5 – 2.6 x10 ⁵	(Cornett et al., 1997; Milton et al., 1994)
1999-2000	Seville, Spain	4 - 300	0.12 – 22.7	(Santos et al., 2004)
2004-2007	Tsukuba, Japan	13 - 116	0.28 – 1.76	(Tosaki et al., 2012)
2010-2011	Several sites in Europe	21 - 746	1.63 – 9.92	(Johnston and McDermott, 2008)
2010-2011	Tsukuba, Japan	18 - 1420	0.1 - 18.3	(Sasa et al., 2022)
2012-2013	Montiers, France	177	3.2	(Grapeloup et al., 2023)
2012-2014	Montiers, France	40 - 900	0.8 – 8.9	(Pupier et al., 2016)
2021-2022	La Hague, France*	217 - 5486	62 - 390	This study

199 *Sites with a nearby anthropogenic source of ³⁶Cl (< 3 km)

200 In the present study, ³⁶Cl levels in collected rainwater ranged from $6.2 \pm 2.5 \times 10^7$ to $3.9 \pm 0.5 \times 10^8$ at.l⁻¹
 201 with an average of $1.7 \pm 0.2 \cdot 10^8$ at.l⁻¹ (median = $1.3 \pm 0.1 \times 10^8$ at.l⁻¹). The levels of ³⁶Cl in our rainwater
 202 samples remain mostly 1 to 2 orders of magnitude above the natural background, which is of the
 203 order of 10⁶ at.l⁻¹. This natural background value is the one generally measured in rainwater by
 204 various authors at sites around the world (Table 2). However, a content of $125 \cdot 10^6$ at.l⁻¹ similar to the
 205 average content of our samples was measured by Herut et al (1992). Finding no correlation with site
 206 location, rainfall amount, topography or distance from the sea, Herut et al. (1992) attributed the
 207 higher ³⁶Cl content to the presence of storms, which considerably increased the drawdown of
 208 atmospheric ³⁶Cl during sampling. Several authors also report variability in ³⁶Cl content in rainwater,
 209 depending on latitude and season (Hainsworth et al., 1994 ; Tosaki et al., 2012). Natural production
 210 of ³⁶Cl is indeed latitudinally dependent, with higher production near the geomagnetic poles (Masarik
 211 and Beer, 1999 ; Poluianov et al., 2016). Similarly, when the tropopause rises in spring, stratospheric
 212 ³⁶Cl is introduced into the troposphere. At the same time, more cosmic rays enter the troposphere,
 213 and the fraction of cosmogenic ³⁶Cl produced in the troposphere increases (Hainsworth et al., 1994).
 214 However, this variability cannot be observed on our results because anthropogenic sources of ³⁶Cl

215 overcome natural production of ^{36}Cl influenced by latitude and season. The variability observed
 216 between the different campaigns in our study is therefore rather influenced by rainfall and Orano's
 217 releases of ^{36}Cl into the atmosphere in the vicinity of the study site.

218 The ^{36}Cl levels measured in our samples remain lower than those of Ohta et al. (2023), who
 219 estimated ^{36}Cl levels in rainwater at $4.7 \times 10^{10} \text{ at.l}^{-1}$ in the 6 weeks following the Fukushima accident.
 220 The ^{36}Cl levels measured in our samples also remain lower than those measured 250 m from the CRL,
 221 which reached $2.6 \times 10^{11} \text{ at.l}^{-1}$ (Milton et al., 1994). It should be noted however that these levels
 222 measured in rainwater by Milton et al. (1994) were linked to CRL discharges of ^{36}Cl . In fact,
 223 measurements taken away from the CRL were strongly related to the distance from the point of
 224 discharge. Levels in rainwater were around $2.6 \times 10^9 \text{ at.l}^{-1}$ at 3 km, $5.6 \times 10^7 \text{ at.l}^{-1}$ at 10 km and 9.5×10^6
 225 at.l^{-1} at 140 km. Nevertheless, this reduction in concentrations was not linear and depended strongly
 226 on speed and wind direction. Drawing a parallel with our study and the distance between our study
 227 site and the Orano plant (2 km), rainwater sampling was carried out at the IRSN Laboratory (IRSN
 228 Lab; $49^{\circ}38'04.8''\text{N}$, $1^{\circ}38'45.6''\text{W}$) located 20 km away from the IRSN Platform, synchronized with the
 229 sampling period running from May 17 to June 1 2022, using an automatic rain collector. As shown in
 230 **Figure 1**, wind direction during this period was predominantly south-westerly.

231 The position of the IRSN Lab, around 20 km from the IRSN Platform and outside the gas plume over
 232 the period under consideration, enabled us to estimate changes in ^{36}Cl levels outside the influence of
 233 Orano plant's gaseous emissions. **Table 3** compares the results obtained from this rainwater sample
 234 with those obtained from the IRSN Platform. Rainfall at IRSN Lab was 15 mm over the period in
 235 consideration, which is of the same order of magnitude as that measured at IRSN platform (21 mm).
 236 But, the ^{36}Cl content in the rainwater sampled at the IRSN laboratory is an order of magnitude lower
 237 than that taken at the IRSN Platform indicating that the marking of ^{36}Cl on IRSN platform is related to
 238 the release of ^{36}Cl by the nearby plant. However, since observation period showing the data in Table
 239 3 is limited, there are still some uncertainties about the representativity of this experiment. It is
 240 nevertheless interesting to note that both, the $^{36}\text{Cl}/\text{Cl}$ ratio and the ^{36}Cl concentration in the IRSN
 241 laboratory rainwater sample are like those in sites away from anthropogenic sources of ^{36}Cl .

242 **Table 3:** Comparison of ^{36}Cl levels in rainwater collected at the study site (IRSN Platform) and at the
 243 IRSN Laboratory (IRSN Lab).

Sample	$^{36}\text{Cl}/\text{Cl}$	$^{36}\text{Cl} (\text{at.l}^{-1})$
Rainwater IRSN Lab	$2,9 \cdot 10^{-14}$	$4,2 \pm 0,6 \cdot 10^6$
Rainwater IRSN Platform	$3,8 \cdot 10^{-12}$	$8,3 \pm 0,9 \cdot 10^7$

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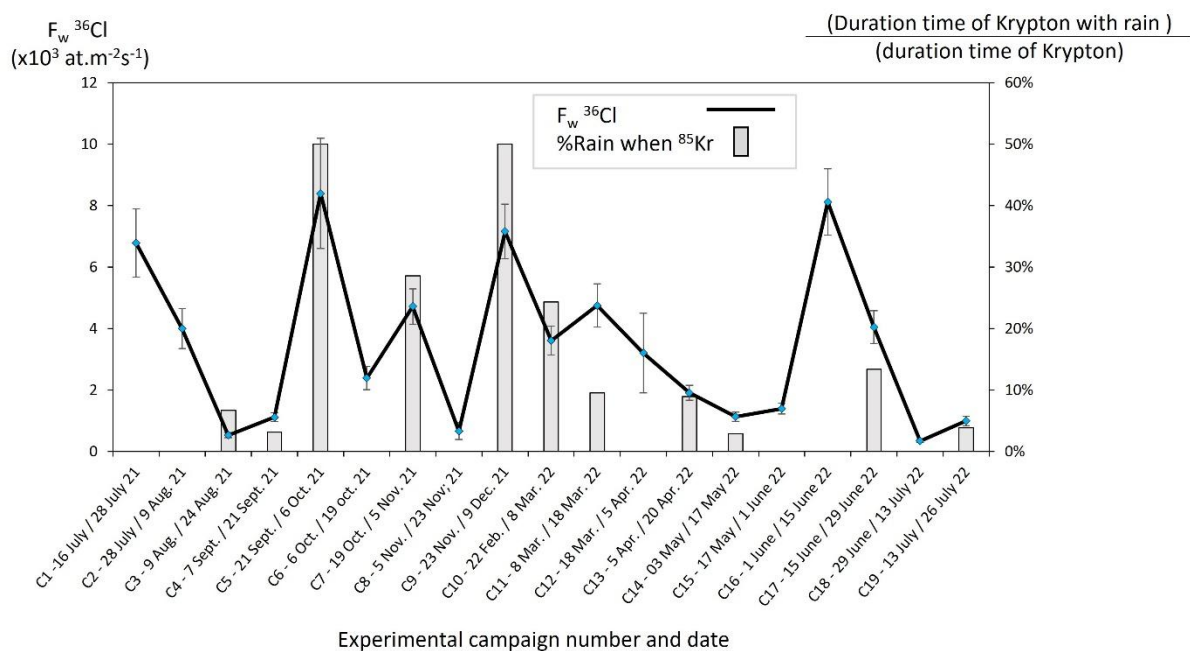
245 3.3. Chlorine 36 wet deposition flux

246 It is difficult to monitor changes in ^{36}Cl wet deposition relying only on concentrations measured in
 247 rainwater. Wet deposition should therefore be expressed in terms of wet deposition flux, in order to
 248 standardize all campaigns on this basis and make a more objective comparison. Wet deposition
 249 fluxes (F_w , $\text{at.m}^{-2}\text{s}^{-1}$) of ^{36}Cl were determined from equation (**Eq.1**) commonly used by various authors
 250 in the literature (Hainsworth et al., 1994) :

251
$$F_w = \frac{[^{36}\text{Cl}] \times P}{d} \text{ (Eq.1)}$$

252 With [^{36}Cl] the sample's ^{36}Cl content (at.l^{-1}), P the campaign rainfall (l.m^{-2}) and d the campaign
 253 duration (s).

254 As shown in **Figure 2**, wet deposition fluxes of ^{36}Cl at our study site range from $3.3 \pm 0.4 \times 10^2$ and
 255 $8.3 \pm 1.7 \cdot 10^3 \text{ at.m}^{-2}\text{s}^{-1}$ with an average of $3.4 \pm 0.5 \cdot 10^3 \text{ at.m}^{-2}\text{s}^{-1}$ (median = $3.2 \pm 1.3 \times 10^3 \text{ at.m}^{-2}\text{s}^{-1}$). As
 256 described above, ^{85}Kr serves as a tracer for the atmospheric plume at the Orano plant and is
 257 therefore measured continuously at the study site. This continuous measurement makes it possible
 258 to determine the periods when the plume is present over the study site. In order to assess the
 259 importance of the plume scavenging on ^{36}Cl deposition fluxes, the percentage of the plume subject
 260 to leaching has been calculated for all campaigns.

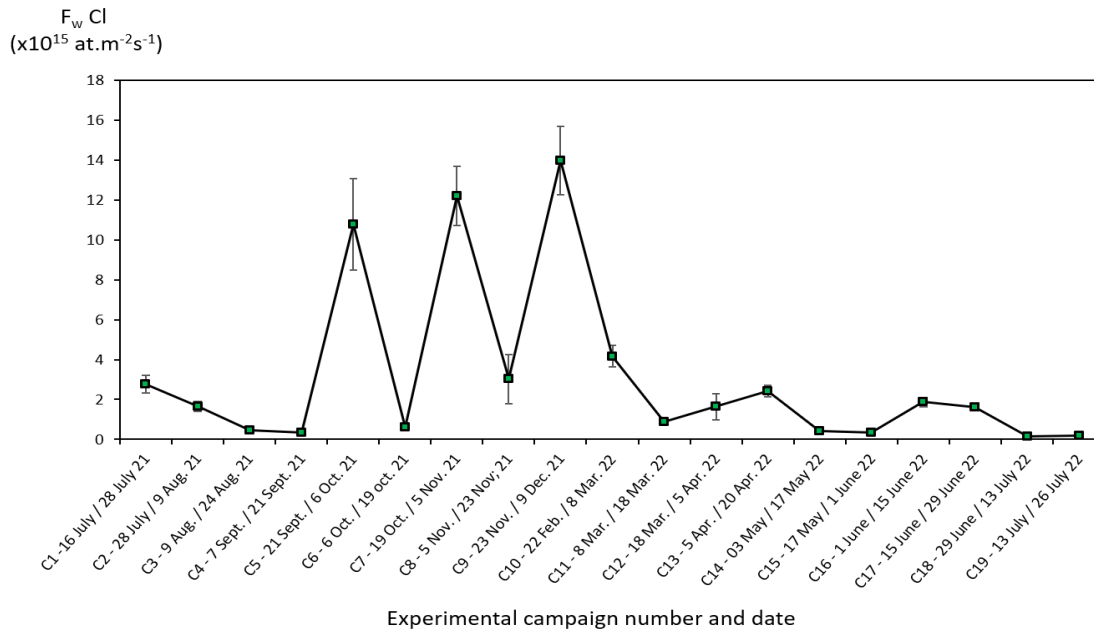


261

262 **Fig. 2.** Wet deposition fluxes of ^{36}Cl and percentages of rain events over the duration of the plume's
 263 presence, determined using krypton 85 measurements at the study site.

264

265 The Cl wet deposition flux ranges from $1.6 \pm 0.1 \times 10^{14}$ and $1.4 \pm 0.1 \times 10^{16} \text{ at.m}^{-2}\text{s}^{-1}$ with an average of
 266 $3.1 \pm 0.5 \times 10^{15} \text{ at.m}^{-2}\text{s}^{-1}$ (median = $1.6 \pm 0.2 \times 10^{15} \text{ at.m}^{-2}\text{s}^{-1}$) (**Figure 3**).



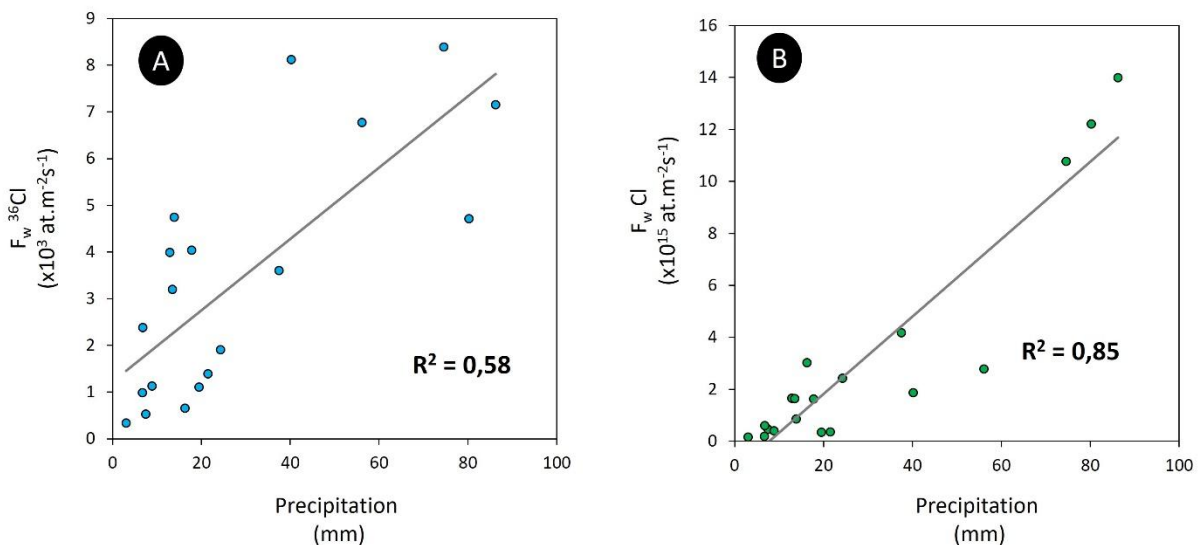
267

268

Fig. 3. Cl wet deposition flow on the study site.

269 The linear regression lines between these wet deposition fluxes (^{36}Cl and Cl) as a function of rainfall
 270 are shown in **Figure 4**.

271



272

273 **Fig. 4.** Correlation coefficient and linear regression line for (A) wet deposition flux of ^{36}Cl as a function
 274 of precipitation; (B) wet deposition flux of Cl as a function of precipitation.

275

276 Based on the linear regression lines for wet deposition fluxes as a function of precipitation, it can be
 277 seen that the deposition flux of ^{36}Cl appears to be less dependent on rainfall than that for Cl.

278 This no doubt explains why Cl wet deposition fluxes are particularly high for campaigns with the
 279 highest rainfall (examples of campaigns n° 5, 7 and 9). On the other hand, the wet deposition flux of
 280 ^{36}Cl on the IRSN Platform is linked to the ^{36}Cl content in the air at the time of precipitations. This is

281 confirmed by the fact that the presence of ^{85}Kr (attesting the presence of ^{36}Cl emitted by Orano on
282 the study site) during a rainy episode increases the quantity of ^{36}Cl deposited. This can be observed in
283 the wet deposition fluxes of ^{36}Cl measured in campaigns n°5, 7 and 9. The particularly high wet
284 deposition fluxes of ^{36}Cl in campaigns n°1, 2 and 16 cannot, however, be explained by the presence
285 of the plume ^{36}Cl . The deposition fluxes measured in these 3 campaigns cannot therefore be
286 explained by the washout alone. Apart from the washout, the wet deposition fluxes measured over
287 these campaigns can only be explained by the presence of a ^{36}Cl drawdown in the cloud or rainout in
288 addition to the washout. We already know that following liquid discharges of ^{36}Cl into the sea near
289 the IRSN Platform, atmospheric ^{36}Cl of marine origin was measured on the IRSN Platform in the
290 absence of the gaseous plume from Orano plant (Sourabie et al., 2023). However, these liquid
291 discharges of ^{36}Cl have different frequencies from atmospheric discharges and the quantities are not
292 measured. Based on marine dispersion models, we also know that radionuclide dispersion following
293 liquid discharges from Orano La Hague can extend over kilometers (Fiévet et al., 2020). Even if it is a
294 weak possibility, as it happens for Cl, ^{36}Cl can also be incorporated into the cloud many kilometers
295 upstream of the study site, and then washed down by precipitation. The presence of wet deposition
296 by rainout in addition to wet deposition by washout would thus explain the significant deposition
297 fluxes found in campaigns n°1, 2 and 16. It could also explain the differences observed in ^{36}Cl content
298 on IRSN Platform and at IRSN Lab. However, unlike Cl, the incorporation of ^{36}Cl into the cloud
299 depends on the quantities of ^{36}Cl discharged into the sea by Orano plant which are subjected to
300 strong variations in space and time.

301 Apart from a few studies focusing exclusively on the wet deposits of ^{36}Cl , notably that of Hainsworth
302 et al. (1994), the majority of studies on wet deposition of ^{36}Cl use rainwater samplers subjected to
303 dry deposition during non-rainy periods. Nevertheless, the wet deposition fluxes of ^{36}Cl determined
304 at our study site are generally an order of magnitude higher than those measured by Santos et al.
305 (2004) in Seville (from 5 to 1250 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$); by Keywood et al. (1998) in Australia (15 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ on
306 average); by Tosaki et al. (2012) in Tsukuba (between 8 and 173 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$); by Pupier et al. (2016) and
307 Grapeloup et al. (2023) at Montier-sur-Saulx (from 45 to 396 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$); by Hainsworth et al. (1994) in
308 Maryland (38 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ on average); by Knies et al. (1994) in West Lafayette (between 10 and 180
309 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$); by Johnston and McDermott (2008) at sites across Europe (between 11 and 153 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$);
310 and by Scheffel et al. (1999) on sites spread across the globe (between 25 and 755 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$).

311 The wet deposition fluxes of ^{36}Cl measured at our site are however equivalent to the dry deposition
312 fluxes of ^{36}Cl on grass previously measured at the same site over the same period of time by Sourabie
313 et al (2023). Indeed, the average wet deposition flux of ^{36}Cl measured in this study ($3.1\times 10^3 \text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$)
314 is similar to the average dry deposition flux of ^{36}Cl on grass ($2.3\times 10^3 \text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$). Although largely above
315 natural background levels, these environmental levels of ^{36}Cl do not present any radiological risk to
316 the environment or humans. Concerning Cl wet deposition fluxes, due to the proximity of the sea,
317 they are higher than those determined by Pupier et al. (2016) 390 km from the sea (0.07 to 1.32×10^{15}
318 $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$) and by Tosaki et al. (2012) 50 km from the sea (0.27 to $2.00\times 10^{15} \text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$).

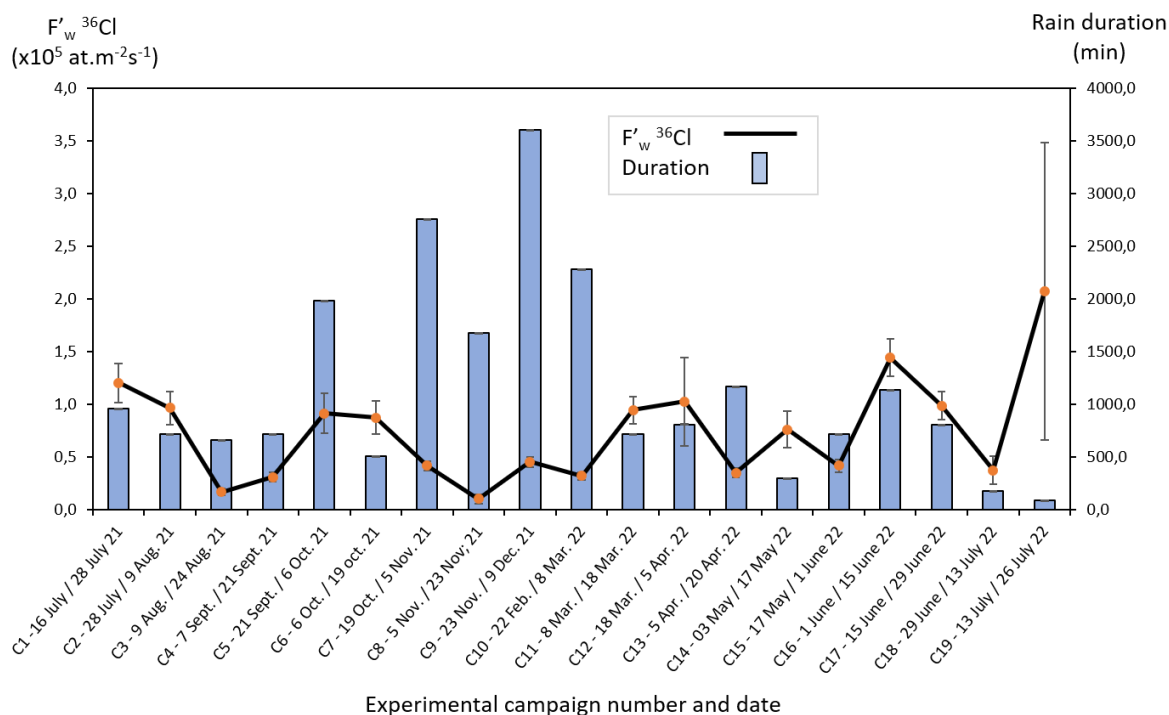
319 Although rarely used, wet deposition can also be described through deposition flux expressed over
320 the duration of precipitation. Wet deposition fluxes of ^{36}Cl over the duration of precipitation refer to
321 the amount of ^{36}Cl washed down at the time of precipitations, rather than over the total duration of
322 the campaign. Wet deposition fluxes of ^{36}Cl expressed over the duration of precipitations (F'_w ; $\text{at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$)
323 were determined from equation (Eq.2):

324
$$F'_w = \frac{[^{36}\text{Cl}] \times P}{dt} \text{ (Eq.2)}$$

325 With [^{36}Cl] the sample's ^{36}Cl content (at.l^{-1}), P the campaign rainfall (l.m^{-2}) and d' the total rainfall
 326 duration (s).

327 As shown in **Figure 5**, ^{36}Cl wet deposition fluxes reported to precipitations duration range from $1.0 \pm$
 328 0.4×10^4 and $2.0 \pm 1.4 \times 10^5 \text{ at.m}^{-2}\text{s}^{-1}$ with an average of $7.4 \pm 1.9 \times 10^4 \text{ at.m}^{-2}\text{s}^{-1}$ (median = $7.5 \pm 1.7 \times 10^4$
 329 $\text{at.m}^{-2}\text{s}^{-1}$). As a reminder, rainfall at the study site is acquired at a frequency of 30 minutes. The
 330 duration of rainfall is therefore calculated by multiplying the number of chronicles in which rainfall
 331 was measured by the duration of the chronicle (30 minutes).

332



333

334 **Fig. 5.** Rainfall duration and wet deposition flux of ^{36}Cl over rainfall duration.

335

336 Overall, the wet deposition fluxes of ^{36}Cl expressed over the duration of precipitation (d') is 1 to 2
 337 orders of magnitude greater than that reported to the entire duration of campaign (d) and dry
 338 deposition fluxes of ^{36}Cl on grass previously measured at the same site over the same period of time
 339 by Sourabie et al (2023). This reflects the amount of chlorine 36 that is captured and delivered to the
 340 ground by raindrops. However, it can be noticed that this flux varies between the different
 341 campaigns. For example, the ^{36}Cl F'_w of campaign n°2 is an order of magnitude higher than that of
 342 campaign n°8 despite their similar rainfall. The absence of plume washout during these two
 343 campaigns shows that wet deposition by rainout is neither systematic nor constant over time. The
 344 differences observed between these campaigns without plume washout may be related to
 345 differences in the cloud generation process, but this is unlikely to explain the large flux difference
 346 between campaigns n°2 and n°8. Due to the large uncertainties on rainout, it is quite difficult to
 347 distinguish between rainout and washout in ^{36}Cl wet deposition for campaigns with presence of ^{85}Kr
 348 during rain events. However, based on our measurements, we think that in the presence of the
 349 plume, ^{36}Cl wet deposition by washout is greater than that by rainout. However, the relatively
 350 punctual presence of the plume at our site (on average 10% of the time) and the high rainfall in the
 351 region make rainout a significant contributor to ^{36}Cl wet deposition on our site.

352

353 4. Conclusions

354 Through 19 experimental campaigns, ^{36}Cl was measured in rainwater for the first time downwind of
355 the Orano La Hague plant, a chronic emitter of small quantities of ^{36}Cl into the environment. The
356 levels of ^{36}Cl measured in rainwater, of the order of 10^8 at.l^{-1} , were 1 to 2 orders of magnitude above
357 the natural background, demonstrating the influence of the plant's discharges on our site. The study
358 of wet deposition fluxes of ^{36}Cl showed the importance of ^{36}Cl deposition when the plant plume is
359 washed out by precipitation, but also highlighted the presence of wet deposition by rainout of ^{36}Cl .
360 However, these results remain specific to our study site and are linked to the proximity of chronic
361 discharge sources (atmospheric and marine). Moreover, these environmental levels of ^{36}Cl do not
362 present any radiological risk. Nevertheless, these results will serve to improve our knowledge on the
363 fate of ^{36}Cl in the environment. They will also enable better assessment of impact studies in a context
364 of decommissioning or nuclear power plant accidents, so that dosimetric impact can be better
365 considered and associated uncertainties reduced. These results could moreover serve as an
366 experimental basis for the development and validation of wet deposition models for ^{36}Cl .

367

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