



**HAL**  
open science

## First measurements of chlorine 36 ( $^{36}\text{Cl}$ ) wet deposition fluxes in the vicinity of Orano La Hague nuclear reprocessing plant in France

Deo-Gratias Sourabie, Didier Hebert, Lucilla Benedetti, Valery Guillou, Elsa Vitorge, Philippe Laguionie, Denis Maro

### ► To cite this version:

Deo-Gratias Sourabie, Didier Hebert, Lucilla Benedetti, Valery Guillou, Elsa Vitorge, et al.. First measurements of chlorine 36 ( $^{36}\text{Cl}$ ) wet deposition fluxes in the vicinity of Orano La Hague nuclear reprocessing plant in France. *Journal of Environmental Radioactivity*, 2024, 278, pp.107470. 10.1016/j.jenvrad.2024.107470 . hal-04608156

HAL Id: hal-04608156

<https://hal.science/hal-04608156v1>

Submitted on 12 Jun 2024

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

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

3  
4 Deo-Gratias Sourabie<sup>(1,2)</sup>, Didier Hebert<sup>1\*</sup>, Lucilla Benedetti<sup>2</sup>, Valery Guillou<sup>2</sup>, Elsa Vitorge<sup>3</sup>, Philippe  
5 Laguionie<sup>1</sup>, Denis Maro<sup>1</sup>

6  
7 <sup>1</sup>Institute for Radioprotection and Nuclear Safety (IRSN), PSE-ENV/STAAR/LERTA, Cherbourg-  
8 Octeville, 50130, France

9 [deo-gratias.sourabie-capgemini@irsn.fr](mailto:deo-gratias.sourabie-capgemini@irsn.fr) , [didier.hebert@irsn.fr](mailto:didier.hebert@irsn.fr) , [philippe.laguionie@irsn.fr](mailto:philippe.laguionie@irsn.fr) ,  
10 [denis.maro@irsn.fr](mailto:denis.maro@irsn.fr)

11  
12 <sup>2</sup>Univ. Aix-Marseille, CNRS, IRD, INRAE, Coll. France, UM 34 CEREGE, Aix-en-Provence, 13545, France  
13 [benedetti@cerge.fr](mailto:benedetti@cerge.fr), [guillou@cerge.fr](mailto:guillou@cerge.fr)

14  
15 <sup>3</sup>EDF – DPNT – DIPDE - DEE – Environment department, Villeurbanne, 69100, France  
16 [elsa.vitorge@edf.fr](mailto:elsa.vitorge@edf.fr)

17  
18  
19 **\* Corresponding author:**

20 E-mail address: [didier.hebert@irsn.fr](mailto:didier.hebert@irsn.fr)

21 Institute for Radiation protection and Nuclear Safety (IRSN)

22 Experimental Laboratory for Research and Expertise on Radionuclide Transfers in the Atmosphere

23 Rue Max Pol Fouchet

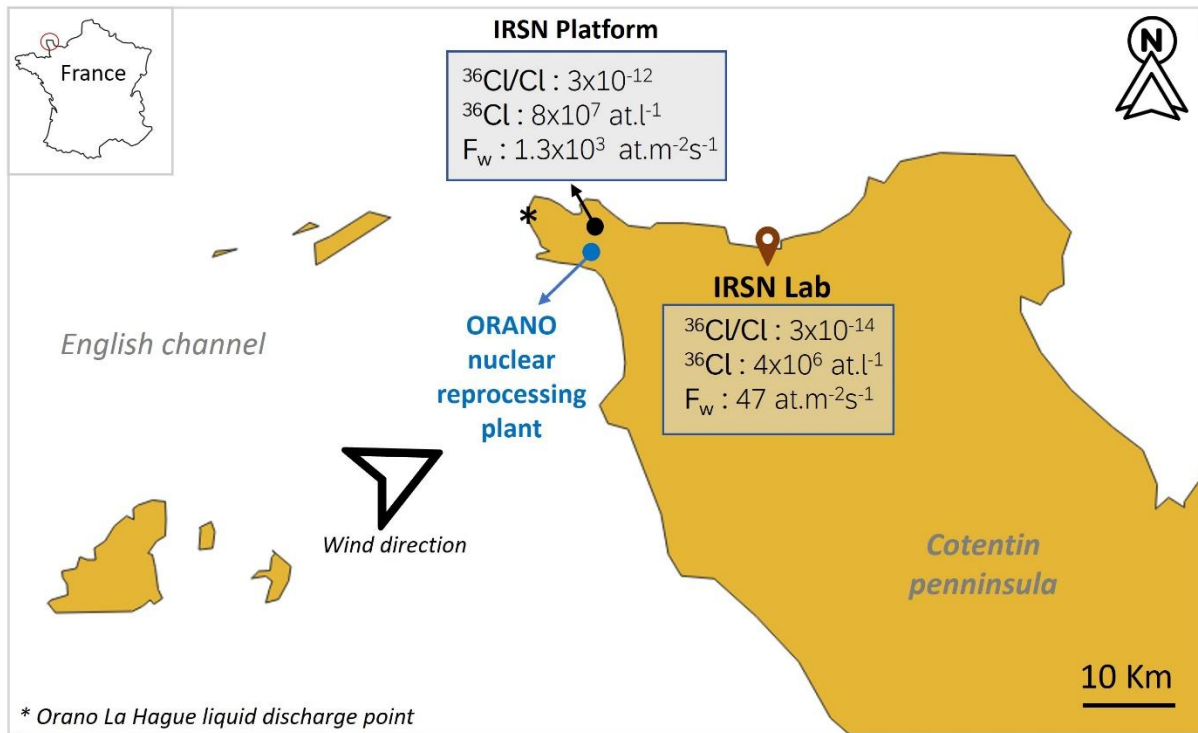
24 BP10- 50130 Cherbourg-Octeville

25 Tel: +33 (0)2 33 01 41 00

26  
27 **Acknowledgements:**

28 This work was supported by the French institute for radiation protection and nuclear safety (IRSN)  
29 and Electricite de France (EDF) under the umbrella of a research and development cooperation  
30 agreement on the safety of civil nuclear installations and their impact on health and the environment  
31 (IRSN LS21602 internal referencing). We would like to thank the LERTA team and Capgemini  
32 Engineering for their support in this work.

39 **Graphical abstract :**



40

41

42 **Summary:**

43 Chlorine 36 ( $^{36}\text{Cl}$ ) 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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  downwind an  
47 anthropogenic source, the Orano nuclear reprocessing plant, which chronically emits  $^{36}\text{Cl}$  into the  
48 environment. Measurements of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  content in the rainwater averaged  $2.3 \times 10^{-12}$  at.at<sup>-1</sup> and  $1.7 \times 10^8$  at.l<sup>-1</sup> 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  $^{36}\text{Cl}$  on the study site is related to the plant discharges. Over the sampling period,  
53 wet deposition fluxes at the study site averaged  $3.4 \times 10^3$  at.m<sup>-2</sup>s<sup>-1</sup>, 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  $^{36}\text{Cl}$  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.

62

63

64        1. Introduction

65        With natural and anthropogenic origin, chlorine 36 ( $^{36}\text{Cl}$ ) 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        ( $\beta^-$ ), with low energy ( $E_{max} = 708.6$  keV) and low radiotoxicity (Bastviken et al., 2013).  $^{36}\text{Cl}$  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).  $^{36}\text{Cl}$  was massively introduced into the environment by neutron activation of  $^{35}\text{Cl}$  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  $^{36}\text{Cl}$  produced into the atmosphere.  
74        This massive introduction of  $^{36}\text{Cl}$  is readily observable in the  $^{36}\text{Cl}$  measurements of ice cores, and the  
75        amount of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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.  $^{36}\text{Cl}$  is therefore released  
80        into the atmosphere in the form of gaseous effluents during fuel reprocessing operations.  $^{36}\text{Cl}$  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),  $^{36}\text{Cl}$  can also be released  
84        into the atmosphere in particulate form. Even when emitted in gaseous form, like other  
85        radionuclides,  $^{36}\text{Cl}$  can form ultrafine particles by nucleation or condense on atmospheric aerosols  
86        present in the environment (Budyka, 2000). Finally,  $^{36}\text{Cl}$  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,  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  have been made possible mainly by advances in accelerator  
93        mass spectrometry, enabling environmental concentrations to be measured and the use of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  providing  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$   
98        and didn't discriminate wet depositions from dry ones. In fact, compared to wet deposition, dry  
99        deposition of  $^{36}\text{Cl}$  is generally considered as non-significant but a recent study showed that dry  
100        deposition can contribute significantly to total  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  in a bio assimilable form by living organisms.

103        The aim of this study is therefore to determine the only-wet deposition fluxes of  $^{36}\text{Cl}$  in fields close to  
104        the Orano La Hague plant, a chronic emitter of small quantities of  $^{36}\text{Cl}$  into environment (GRNC,  
105        1999). In fact, since Milton et al. (1994), no studies have been carried out on wet deposition of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  in this area are greater than those  
109        generally reported in the literature (Sourabie et al., 2023). Following the study of  $^{36}\text{Cl}$  dry deposition

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

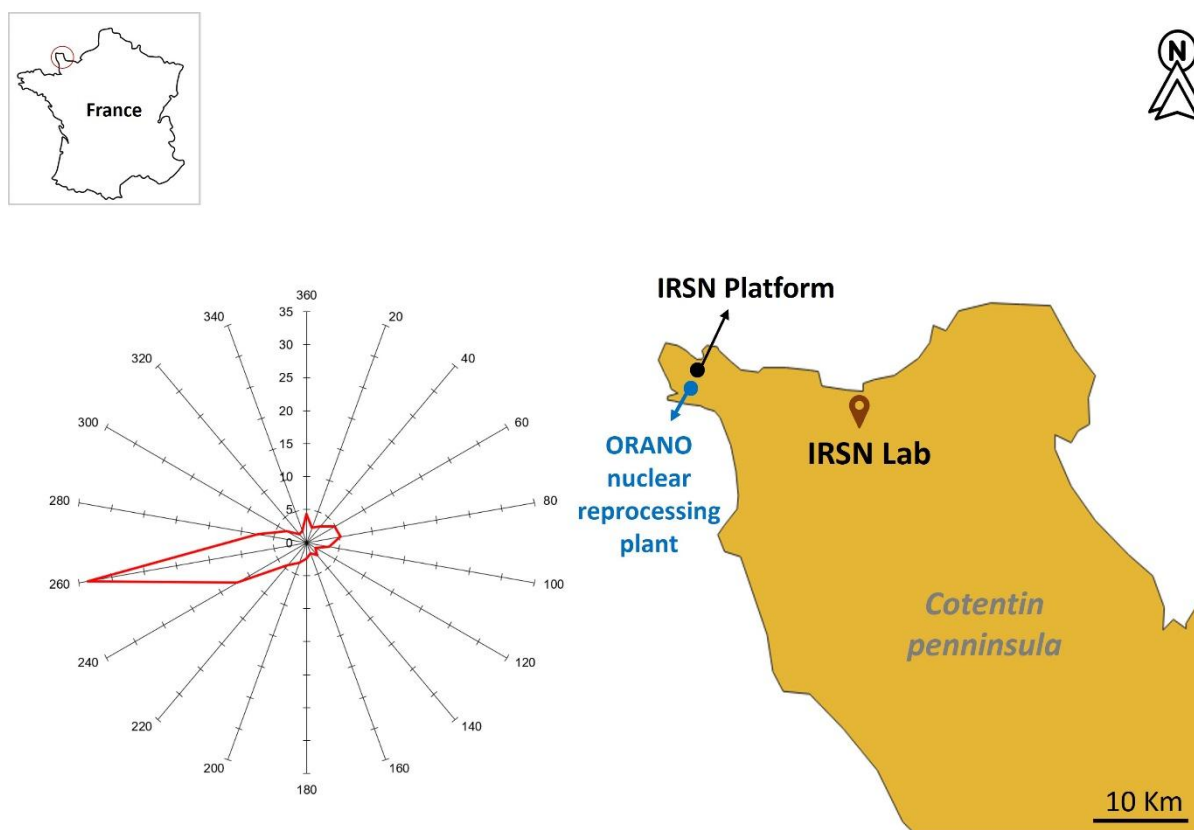
112

## 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**).  $^{36}\text{Cl}$  is emitted concomitantly with krypton 85 ( $^{85}\text{Kr}$ ), which  
118 serves as a tracer for the plant's atmospheric plume (Connan et al., 2014). As such,  $^{85}\text{Kr}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $\text{AgNO}_3$  10% (analytical grade Emsure  
136 ACS, Merck) and 20 mL of nitric acid  $\text{HNO}_3$  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  $\text{NH}_4\text{OH}$  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 ( $\text{BaSO}_3$ ). 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  $23^\circ\text{C}$ ) before being oven-dried at  $40^\circ\text{C}$  for 72h. After drying, the silver chloride  
146 precipitates were sent for measurement of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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 ( $\text{Na}^{35}\text{Cl}$ ;  $^{35}\text{Cl} = 99.66\%$ , 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  $1.6 \times 10^{-15}$  which is close to the ratio  $^{36}\text{Cl}/^{35}\text{Cl}$  of blanks  
159 measured by AMS at ASTER ( $1.3 \times 10^{-15}$ ; Braucher et al., 2018). This allows us to ensure the absence of  
160  $^{36}\text{Cl}$  contamination during the processing of our samples.

161

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

163 **Table 1** shows the results of  $^{36}\text{Cl}$  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.

168

169

170

171

172

173

174

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

Campaign no.	Start date	End date	Rainfall	$\text{Cl}^-$	$^{36}\text{Cl}/\text{Cl}$	$^{36}\text{Cl}$
			<i>mm</i>	<i>mg.l<sup>-1</sup></i>	<i>x10<sup>-12</sup></i>	<i>x10<sup>7</sup> at.l<sup>-1</sup></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  $2.1 \times 10^{-13}$  to  $5.4 \times 10^{-12}$  with an average of  
 179  $2.3 \times 10^{-12}$  (median =  $2.4 \times 10^{-12}$ ). The  $^{36}\text{Cl}/\text{Cl}$  isotopic ratio in rainwater is strongly influenced by the  
 180 atmospheric Cl content of the aera. Since  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  (**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  $30.8 \times 10^{-12}$ . 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  $1.4 \times 10^{-12}$  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).

195



196  
197  
198

**Table 2:**  $^{36}\text{Cl}$  measurements in rainwater reported in the literature.

Date	Site	$^{36}\text{Cl}/\text{Cl} (\times 10^{-15})$	$\text{at.l}^{-1} (\times 10^6)$	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*	$(1.6 - 30.8) \times 10^3$	$9.5 - 2.6 \times 10^5$	(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  $^{36}\text{Cl}$  (< 3 km)

200 In the present study,  $^{36}\text{Cl}$  levels in collected rainwater ranged from  $6.2 \pm 2.5 \times 10^7$  to  $3.9 \pm 0.5 \times 10^8$   $\text{at.l}^{-1}$   
 201 with an average of  $1.7 \pm 0.2 \cdot 10^8$   $\text{at.l}^{-1}$  (median =  $1.3 \pm 0.1 \times 10^8$   $\text{at.l}^{-1}$ ). The levels of  $^{36}\text{Cl}$  in our rainwater  
 202 samples remain mostly 1 to 2 orders of magnitude above the natural background, which is of the  
 203 order of  $10^6$   $\text{at.l}^{-1}$ . 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  $125 \cdot 10^6$   $\text{at.l}^{-1}$  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  $^{36}\text{Cl}$  content to the presence of storms, which considerably increased the drawdown of  
 208 atmospheric  $^{36}\text{Cl}$  during sampling. Several authors also report variability in  $^{36}\text{Cl}$  content in rainwater,  
 209 depending on latitude and season (Hainsworth et al., 1994 ; Tosaki et al., 2012). Natural production  
 210 of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  is introduced into the troposphere. At the same time, more cosmic rays enter the troposphere,  
 213 and the fraction of cosmogenic  $^{36}\text{Cl}$  produced in the troposphere increases (Hainsworth et al., 1994).  
 214 However, this variability cannot be observed on our results because anthropogenic sources of  $^{36}\text{Cl}$



215 overcome natural production of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  into the atmosphere in the vicinity of the study site.

218 The  $^{36}\text{Cl}$  levels measured in our samples remain lower than those of Ohta et al. (2023), who  
 219 estimated  $^{36}\text{Cl}$  levels in rainwater at  $4.7 \times 10^{10} \text{ at.l}^{-1}$  in the 6 weeks following the Fukushima accident.  
 220 The  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$ . 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  $9.5 \times 10^6$   
 225  $\text{at.l}^{-1}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  on IRSN platform is related to  
 238 the release of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  concentration in the IRSN  
 241 laboratory rainwater sample are like those in sites away from anthropogenic sources of  $^{36}\text{Cl}$ .

242 **Table 3:** Comparison of  $^{36}\text{Cl}$  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	$2,9.10^{-14}$	$4,2 \pm 0,6.10^6$
Rainwater IRSN Platform	$3,8.10^{-12}$	$8,3 \pm 0,9.10^7$

244

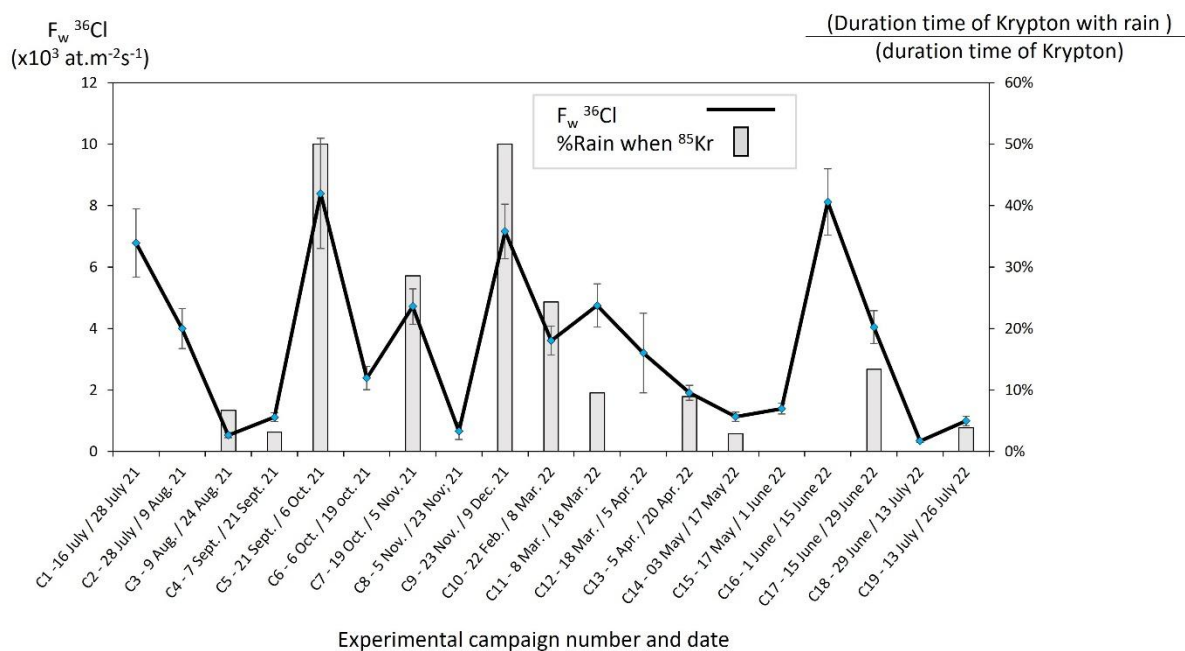
### 245 3.3. Chlorine 36 wet deposition flux

246 It is difficult to monitor changes in  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  were determined from equation (**Eq.1**) commonly used by various authors  
 250 in the literature (Hainsworth et al., 1994) :

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

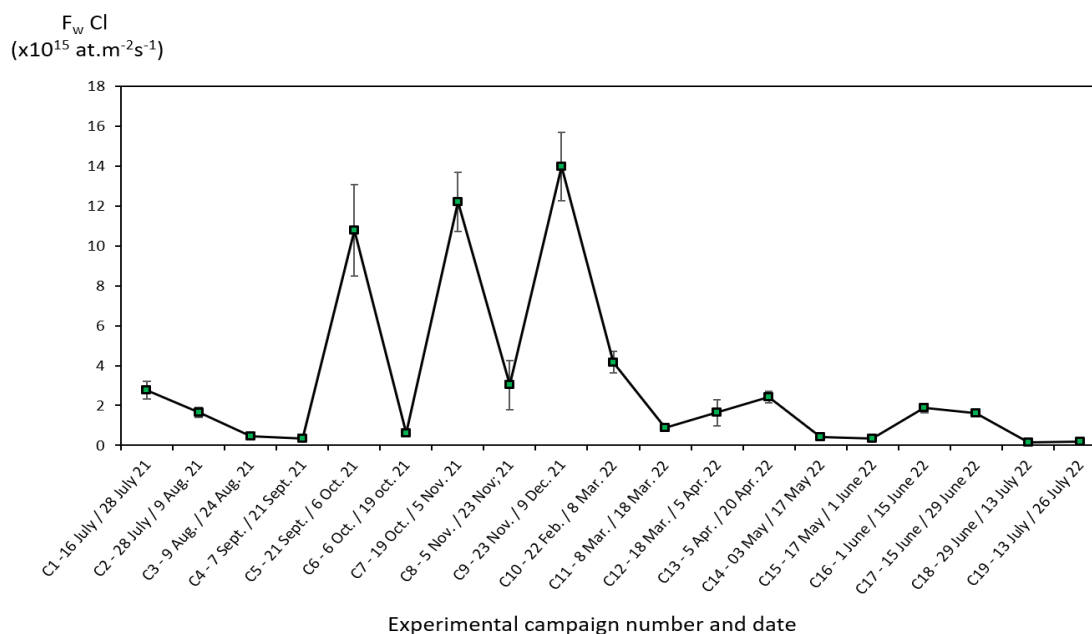
252 With [ $^{36}\text{Cl}$ ] the sample's  $^{36}\text{Cl}$  content ( $\text{at.l}^{-1}$ ),  $P$  the campaign rainfall ( $\text{l.m}^{-2}$ ) and  $d$  the campaign  
 253 duration (s).

254 As shown in **Figure 2**, wet deposition fluxes of  $^{36}\text{Cl}$  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,  $^{85}\text{Kr}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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**).



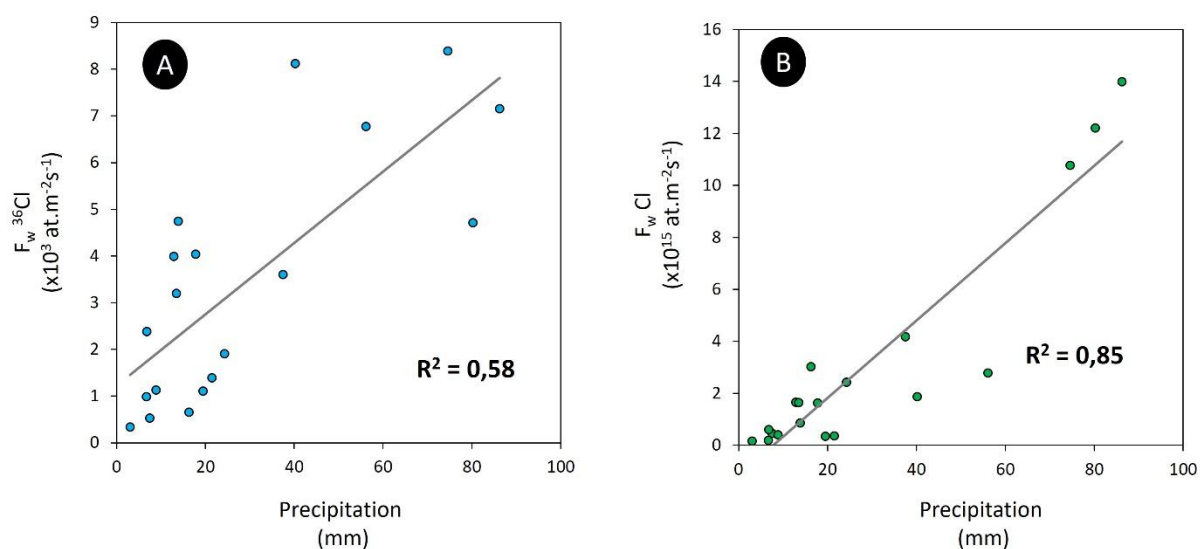
267

268

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

269 The linear regression lines between these wet deposition fluxes ( $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  on the IRSN Platform is linked to the  $^{36}\text{Cl}$  content in the air at the time of precipitations. This is

281 confirmed by the fact that the presence of  $^{85}\text{Kr}$  (attesting the presence of  $^{36}\text{Cl}$  emitted by Orano on  
 282 the study site) during a rainy episode increases the quantity of  $^{36}\text{Cl}$  deposited. This can be observed in  
 283 the wet deposition fluxes of  $^{36}\text{Cl}$  measured in campaigns n°5, 7 and 9. The particularly high wet  
 284 deposition fluxes of  $^{36}\text{Cl}$  in campaigns n°1, 2 and 16 cannot, however, be explained by the presence  
 285 of the plume  $^{36}\text{Cl}$ . 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  $^{36}\text{Cl}$  drawdown in the cloud or rainout in  
 288 addition to the washout. We already know that following liquid discharges of  $^{36}\text{Cl}$  into the sea near  
 289 the IRSN Platform, atmospheric  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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,  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  content  
 298 on IRSN Platform and at IRSN Lab. However, unlike Cl, the incorporation of  $^{36}\text{Cl}$  into the cloud  
 299 depends on the quantities of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$ , notably that of Hainsworth  
 302 et al. (1994), the majority of studies on wet deposition of  $^{36}\text{Cl}$  use rainwater samplers subjected to  
 303 dry deposition during non-rainy periods. Nevertheless, the wet deposition fluxes of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  measured at our site are however equivalent to the dry deposition  
 312 fluxes of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $1.32 \times 10^{15}$   
 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  $2.00 \times 10^{15} \text{ at}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ).

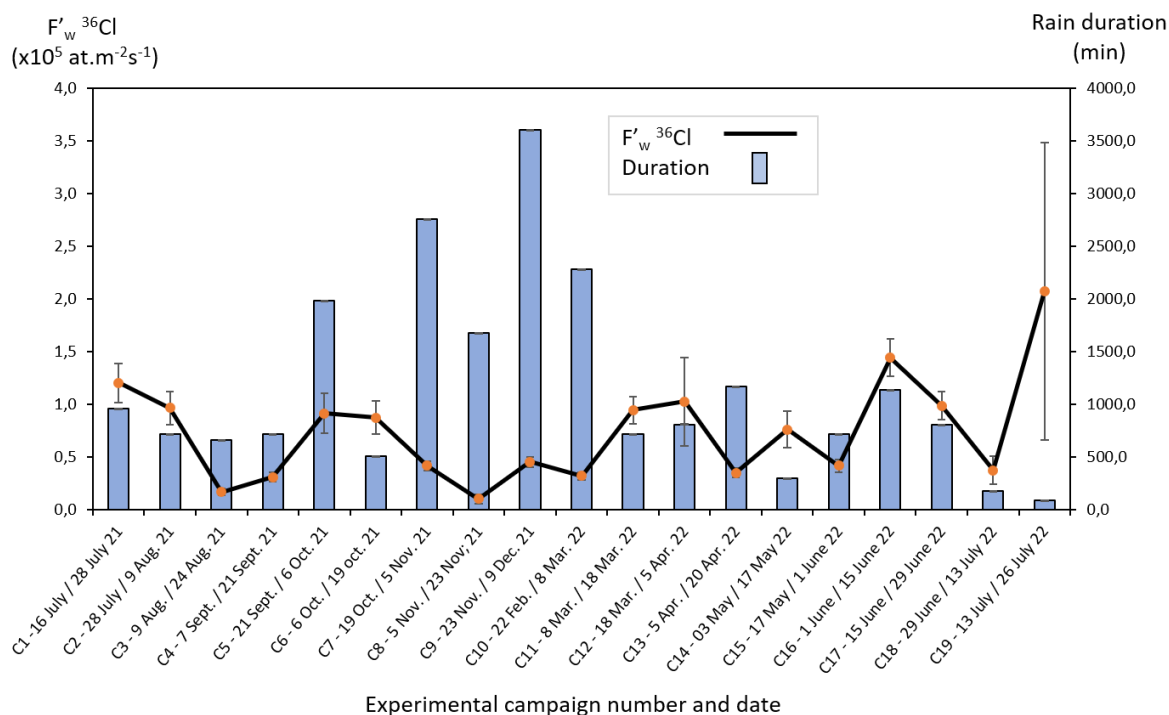
319 Although rarely used, wet deposition can also be described through deposition flux expressed over  
 320 the duration of precipitation. Wet deposition fluxes of  $^{36}\text{Cl}$  over the duration of precipitation refer to  
 321 the amount of  $^{36}\text{Cl}$  washed down at the time of precipitations, rather than over the total duration of  
 322 the campaign. Wet deposition fluxes of  $^{36}\text{Cl}$  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):

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

325 With [ $^{36}\text{Cl}$ ] the sample's  $^{36}\text{Cl}$  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**,  $^{36}\text{Cl}$  wet deposition fluxes reported to precipitations duration range from  $1.0 \pm$   
 328  $0.4 \times 10^4$  and  $2.0 \pm 1.4 \times 10^5$  at.m $^{-2}$ s $^{-1}$  with an average of  $7.4 \pm 1.9 \times 10^4$  at.m $^{-2}$ s $^{-1}$  (median =  $7.5 \pm 1.7 \times 10^4$   
 329 at.m $^{-2}$ 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  $^{36}\text{Cl}$  over rainfall duration.

335

336 Overall, the wet deposition fluxes of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$   $F'_w$  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  $^{36}\text{Cl}$  wet deposition for campaigns with presence of  $^{85}\text{Kr}$   
 348 during rain events. However, based on our measurements, we think that in the presence of the  
 349 plume,  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$  wet deposition on our site.

352

#### 353 4. Conclusions

354 Through 19 experimental campaigns,  $^{36}\text{Cl}$  was measured in rainwater for the first time downwind of  
355 the Orano La Hague plant, a chronic emitter of small quantities of  $^{36}\text{Cl}$  into the environment. The  
356 levels of  $^{36}\text{Cl}$  measured in rainwater, of the order of  $10^8 \text{ at.l}^{-1}$ , 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  $^{36}\text{Cl}$  showed the importance of  $^{36}\text{Cl}$  deposition when the plant plume is  
359 washed out by precipitation, but also highlighted the presence of wet deposition by rainout of  $^{36}\text{Cl}$ .  
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  $^{36}\text{Cl}$  do not  
362 present any radiological risk. Nevertheless, these results will serve to improve our knowledge on the  
363 fate of  $^{36}\text{Cl}$  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  $^{36}\text{Cl}$ .

367

#### 368 References:

- 369 Bastviken, D., Svensson, T., Sandén, P., Kylin, H., 2013. Chlorine cycling and fates of  $^{36}\text{Cl}$  in terrestrial  
370 environments. Swedish Nuclear Fuel and Waste Management Co.
- 371 Bouchez, C., Pupier, J., Benedetti, L., Deschamps, P., Guillou, V., Keddadouche, K., Aumaître, G.,  
372 Arnold, M., Bourlès, D., 2015. Isotope Dilution-AMS technique for  $^{36}\text{Cl}$  and Cl determination  
373 in low chlorine content waters. *Chem. Geol.* 404, 62–70.  
374 <https://doi.org/10.1016/j.chemgeo.2015.03.022>
- 375 Braucher, R., Keddadouche, K., Aumaître, G., Bourlès, D.L., Arnold, M., Pivot, S., Baroni, M., Scharf,  
376 A., Rugel, G., Bard, E., 2018. Chlorine measurements at the 5MV French AMS national facility  
377 ASTER: Associated external uncertainties and comparability with the 6MV DREAMS facility.  
378 *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. At.* 420, 40–45.  
379 <https://doi.org/10.1016/j.nimb.2018.01.025>
- 380 Budyka, A.K., 2000. Phase transformations of iodine and other volatile radionuclides in free  
381 atmosphere. *J. Aerosol Sci., European Aerosol Conference 2000* 31, 480–481.  
382 [https://doi.org/10.1016/S0021-8502\(00\)90493-2](https://doi.org/10.1016/S0021-8502(00)90493-2)
- 383 Connan, O., Solier, L., Hébert, D., Maro, D., Lamotte, M., Voiseux, C., Laguionie, P., Cazimajou, O., Le  
384 Cavelier, S., Godinot, C., Morillon, M., Thomas, L., Percot, S., 2014. Near-field krypton-85  
385 measurements in stable meteorological conditions around the AREVA NC La Hague  
386 reprocessing plant: estimation of atmospheric transfer coefficients. *J. Environ. Radioact.* 137,  
387 142–149. <https://doi.org/10.1016/j.jenvrad.2014.07.012>
- 388 Cornett, R.J., Andrews, H.R., Chant, L.A., Davies, W.G., Greiner, B.F., Imahori, Y., Koslowsky, V.T.,  
389 Kotzer, T., Milton, J.C.D., Milton, G.M., 1997. Is  $^{36}\text{Cl}$  from weapons' test fallout still cycling in  
390 the atmosphere? *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. At.*,  
391 *Accelerator Mass Spectrometry* 123, 378–381. [https://doi.org/10.1016/S0168-](https://doi.org/10.1016/S0168-583X(96)00733-1)  
392 [583X\(96\)00733-1](https://doi.org/10.1016/S0168-583X(96)00733-1)
- 393 Fiévet, B., Bailly du Bois, P., Voiseux, C., Godinot, C., Cazimajou, O., Solier, L., De Vismes Ott, A.,  
394 Cossonnet, C., Habibi, A., Fleury, S., 2020. A comprehensive assessment of two-decade  
395 radioactivity monitoring around the Channel Islands. *J. Environ. Radioact.* 223–224, 106381.  
396 <https://doi.org/10.1016/j.jenvrad.2020.106381>
- 397 Finkel, R., Arnold, M., Aumaître, G., Benedetti, L., Bourlès, D., Keddadouche, K., Merchel, S., 2013.  
398 Improved  $^{36}\text{Cl}$  performance at the ASTER HVE 5MV accelerator mass spectrometer national



399 facility. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. At., Proceedings of  
400 the Twelfth International Conference on Accelerator Mass Spectrometry, Wellington, New  
401 Zealand, 20-25 March 2011 294, 121–125. <https://doi.org/10.1016/j.nimb.2012.05.019>  
402 Finkel, R.C., Nishiizumi, K., Elmore, D., Ferraro, R.D., Gove, H.E., 1980.  $^{36}\text{Cl}$  in polar ice, rainwater and  
403 seawater. Geophys. Res. Lett. 7, 983–986. <https://doi.org/10.1029/GL007i011p00983>  
404 Grapeloup, C., Cornu, S., Giraud, X., Pupier, J., Team, A., Guillou, V., Ciffroy, P., Cabana, B.L.,  
405 Couegnas, C., Hatté, C., Benedetti, L., 2023.  $^{36}\text{Cl}$ , a new tool to assess soil carbon dynamics.  
406 Sci. Rep. 13, 15085. <https://doi.org/10.1038/s41598-023-41555-x>  
407 Green, J.R., Cecil, L.D., Synal, H.-A., Santos, J., Kreutz, K.J., Wake, C.P., 2004. A high resolution record  
408 of chlorine-36 nuclear-weapons-tests fallout from Central Asia. Nucl. Instrum. Methods Phys.  
409 Res. Sect. B Beam Interact. Mater. At., Proceedings of the Ninth International Conference on  
410 Accelerator Mass Spectrometry 223–224, 854–857.  
411 <https://doi.org/10.1016/j.nimb.2004.04.157>  
412 GRNC, 1999. GRNC, Groupe Radioécologie Nord Cotentin, Work group no.1 report: inventory of  
413 radioactive waste from nuclear installations. [WWW Document]. URL [http://www.gep-](http://www.gep-nucleaire.org/norcot/gepnc/sections/travauxgep/premiere_mission5339/volume_1_publie_en/view)  
414 [nucleaire.org/norcot/gepnc/sections/travauxgep/premiere\\_mission5339/volume\\_1\\_publie\\_](http://www.gep-nucleaire.org/norcot/gepnc/sections/travauxgep/premiere_mission5339/volume_1_publie_en/view)  
415 [en/view](http://www.gep-nucleaire.org/norcot/gepnc/sections/travauxgep/premiere_mission5339/volume_1_publie_en/view) (accessed 2.16.23).  
416 Hainsworth, L.J., Mignerey, A.C., Helz, G.R., Sharma, P., Kubik, P.W., 1994. Modern chlorine-36  
417 deposition in southern Maryland, U.S.A. Nucl. Instrum. Methods Phys. Res. Sect. B Beam  
418 Interact. Mater. At. 92, 345–349. [https://doi.org/10.1016/0168-583X\(94\)96032-1](https://doi.org/10.1016/0168-583X(94)96032-1)  
419 Heikkilä, U., Beer, J., Feichter, J., Alfimov, V., Synal, H.-A., Schotterer, U., Eichler, A., Schwikowski, M.,  
420 Thompson, L., 2009.  $^{36}\text{Cl}$  bomb peak: comparison of modeled and measured data.  
421 Atmospheric Chem. Phys. 9, 4145–4156. <https://doi.org/10.5194/acp-9-4145-2009>  
422 Herut, B., Starinsky, A., Katz, A., Paul, M., Boaretto, E., Berkovits, D., 1992.  $^{36}\text{Cl}$  in chloride-rich  
423 rainwater, Israel. Earth Planet. Sci. Lett. 109, 179–183. [https://doi.org/10.1016/0012-](https://doi.org/10.1016/0012-821X(92)90082-7)  
424 [821X\(92\)90082-7](https://doi.org/10.1016/0012-821X(92)90082-7)  
425 Huggle, D., Blinov, A., Stan-Sion, C., Korschinek, G., Scheffel, C., Massonet, S., Zerle, L., Beer, J.,  
426 Parrat, Y., Gaeggeler, H., Hajdas, W., Nolte, E., 1996. Production of cosmogenic  $^{36}\text{Cl}$  on  
427 atmospheric argon. Planet. Space Sci. 44, 147–151. [https://doi.org/10.1016/0032-](https://doi.org/10.1016/0032-0633(95)00085-2)  
428 [0633\(95\)00085-2](https://doi.org/10.1016/0032-0633(95)00085-2)  
429 Johnston, V., McDermott, F., 2008. The distribution of meteoric Cl-36 in precipitation across Europe  
430 in spring 2007. Earth Planet. Sci. Lett. 275, 154–164.  
431 <https://doi.org/10.1016/j.epsl.2008.08.021>  
432 Keywood, M.D., Fifield, L.K., Chivas, A.R., Cresswell, R.G., 1998. Fallout of chlorine 36 to the Earth's  
433 surface in the southern hemisphere. J. Geophys. Res. Atmospheres 103, 8281–8286.  
434 <https://doi.org/10.1029/97JD03125>  
435 Knies, D.L., Elmore, D., Sharma, P., Vogt, S., Li, R., Lipschutz, M.E., Petty, G., Farrell, J., Monaghan,  
436 M.C., Fritz, S., Agee, E., 1994.  $^7\text{Be}$ ,  $^{10}\text{Be}$ , and  $^{36}\text{Cl}$  in precipitation. Nucl. Instrum. Methods  
437 Phys. Res. Sect. B Beam Interact. Mater. At. 92, 340–344. [https://doi.org/10.1016/0168-](https://doi.org/10.1016/0168-583X(94)96031-3)  
438 [583X\(94\)96031-3](https://doi.org/10.1016/0168-583X(94)96031-3)  
439 Lazarev, V., 2003. The cosmogenic and anthropogenic  $^{36}\text{Cl}$  in the environment. Technische  
440 Universität München.  
441 Le Dizès, S., Gonze, M.A., 2019. Behavior of  $^{36}\text{Cl}$  in agricultural soil-plant systems: A review of  
442 transfer processes and modelling approaches. J. Environ. Radioact. 196, 82–90.  
443 <https://doi.org/10.1016/j.jenvrad.2018.10.011>  
444 Llopart-Babot, I., Vasile, M., Dobney, A., Boden, S., Bruggeman, M., Leermakers, M., Qiao, J.,  
445 Warwick, P., 2022. On the determination of  $^{36}\text{Cl}$  and  $^{129}\text{I}$  in solid materials from nuclear  
446 decommissioning activities. J. Radioanal. Nucl. Chem. [https://doi.org/10.1007/s10967-022-](https://doi.org/10.1007/s10967-022-08327-9)  
447 [08327-9](https://doi.org/10.1007/s10967-022-08327-9)  
448 Masarik, J., Beer, J., 1999. Simulation of particle fluxes and cosmogenic nuclide production in the  
449 Earth's atmosphere. J. Geophys. Res. Atmospheres 104, 12099–12111.  
450 <https://doi.org/10.1029/1998JD200091>



451 Merchel, S., Bremser, W., Alfimov, V., Arnold, M., Aumaître, G., Benedetti, L., Bourlès, D.L., Caffee,  
452 M., Fifield, L.K., Finkel, R.C., Freeman, S.P.H.T., Martschini, M., Matsushi, Y., Rood, D.H., Sasa,  
453 K., Steier, P., Takahashi, T., Tamari, M., Tims, S.G., Tosaki, Y., Wilcken, K.M., Xu, S., 2011.  
454 Ultra-trace analysis of  $^{36}\text{Cl}$  by accelerator mass spectrometry: an interlaboratory study. *Anal.*  
455 *Bioanal. Chem.* 400, 3125–3132. <https://doi.org/10.1007/s00216-011-4979-2>

456 Milton, G.M., Andrews, H.R., Causey, S.E., Chant, L.A., Cornett, R.J., Davies, W.G., Greiner, B.F.,  
457 Koslowsky, V.T., Imahori, Y., Kramer, S.J., McKay, J.W., Milton, J.C.D., 1994. Chlorine-36  
458 dispersion in the Chalk River area. *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact.*  
459 *Mater. At.* 92, 376–379. [https://doi.org/10.1016/0168-583X\(94\)96037-2](https://doi.org/10.1016/0168-583X(94)96037-2)

460 Nakata, K., Hasegawa, T., 2011. Improvement of pre-treatment method for  $^{36}\text{Cl}/\text{Cl}$  measurement of  
461  $\text{Cl}$  in natural groundwater by AMS. *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact.*  
462 *Mater. At.* 269, 300–307. <https://doi.org/10.1016/j.nimb.2010.11.022>

463 Ohta, T., Fifield, L.K., Palcsu, L., Tims, S.G., Pavetich, S., Mahara, Y., 2023. Record of  $^3\text{H}$  and  $^{36}\text{Cl}$  from  
464 the Fukushima nuclear accident recovered from soil water in the unsaturated zone at  
465 Koriyama. *Sci. Rep.* 13, 19672. <https://doi.org/10.1038/s41598-023-46853-y>

466 Petrov, B.F., Pokhitonov, Yu.A., 2020. Content of Long-Lived Radionuclides of Carbon-14 and  
467 Chlorine-36 in Reactor Graphite and in the Biosphere (Is there a Problem with Carbon-14 and  
468 Chlorine-36 when It Comes to the Processing of Reactor Graphite?). *Radiochemistry* 62, 138–  
469 140. <https://doi.org/10.1134/S106636222001018X>

470 Phillips, F.M., 2000. Chlorine-36, in: Cook, P.G., Herczeg, A.L. (Eds.), *Environmental Tracers in*  
471 *Subsurface Hydrology*. Springer US, Boston, MA, pp. 299–348. [https://doi.org/10.1007/978-](https://doi.org/10.1007/978-1-4615-4557-6_10)  
472 [1-4615-4557-6\\_10](https://doi.org/10.1007/978-1-4615-4557-6_10)

473 Phillips, F.M., Mattick, J.L., Duval, T.A., Elmore, D., Kubik, P.W., 1988. Chlorine 36 and tritium from  
474 nuclear weapons fallout as tracers for long-term liquid and vapor movement in desert soils.  
475 *Water Resour. Res.* 24, 1877–1891. <https://doi.org/10.1029/WR024i011p01877>

476 Poluianov, S.V., Kovaltsov, G.A., Mishev, A.L., Usoskin, I.G., 2016. Production of cosmogenic isotopes  
477  $^7\text{Be}$ ,  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{22}\text{Na}$ , and  $^{36}\text{Cl}$  in the atmosphere: Altitudinal profiles of yield functions. *J.*  
478 *Geophys. Res. Atmospheres* 121, 8125–8136. <https://doi.org/10.1002/2016JD025034>

479 Pupier, J., Benedetti, L., Bouchez, C., Bourlès, D., Leclerc, E., Thiry, Y., Guillou, V., 2016. Monthly  
480 record of the  $\text{Cl}$  and  $^{36}\text{Cl}$  fallout rates in a deciduous forest ecosystem in NE France in 2012  
481 and 2013. *Quat. Geochronol.* 35, 26–35. <https://doi.org/10.1016/j.quageo.2016.04.002>

482 Santos, F.J., López-Gutiérrez, J.M., García-León, M., Schnabel, C., Synal, H.-A., Suter, M., 2004.  
483 Analysis of  $^{36}\text{Cl}$  in atmospheric samples from Seville (Spain) by AMS. *Nucl. Instrum. Methods*  
484 *Phys. Res. Sect. B Beam Interact. Mater. At.*, Proceedings of the Ninth International  
485 Conference on Accelerator Mass Spectrometry 223–224, 501–506.  
486 <https://doi.org/10.1016/j.nimb.2004.04.094>

487 Sasa, K., Ochiai, Y., Tosaki, Y., Matsunaka, T., Takahashi, T., Matsumura, M., Sueki, K., 2022. Chlorine-  
488  $^{36}\text{Cl}$  deposition at Tsukuba, Japan, after the Fukushima Daiichi Nuclear Power Plant accident.  
489 *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. At.* 532, 73–77.  
490 <https://doi.org/10.1016/j.nimb.2022.10.003>

491 Scheffel, C., Blinov, A., Massonet, S., Sachsenhauser, H., Stan-Sion, C., Beer, J., Synal, H.A., Kubik,  
492 P.W., Kaba, M., Nolte, E., 1999.  $^{36}\text{Cl}$  in modern atmospheric precipitation. *Geophys. Res.*  
493 *Lett.* 26, 1401–1404. <https://doi.org/10.1029/1999GL900249>

494 Sheppard, S.C., Johnson, L.H., Goodwin, B.W., Tait, J.C., Wuschke, D.M., Davison, C.C., 1996. Chlorine-  
495  $^{36}\text{Cl}$  in nuclear waste disposal—1. Assessment results for used fuel with comparison to  $^{129}\text{I}$   
496 and  $^{14}\text{C}$ . *Waste Manag.* 16, 607–614. [https://doi.org/10.1016/S0956-053X\(97\)00001-9](https://doi.org/10.1016/S0956-053X(97)00001-9)

497 Sourabie, D.-G., Hebert, D., Benedetti, L., Vitorge, E., Lourino-Cabana, B., Guillou, V., Maro, D., 2023.  
498 First quantitative constraints on chlorine 36 dry deposition velocities on grassland:  
499 Comparing measurements and modelling results. *J. Environ. Radioact.* 268–269, 107264.  
500 <https://doi.org/10.1016/j.jenvrad.2023.107264>

501 Stone, J.O., Allan, G.L., Fifield, L.K., Cresswell, R.G., 1996. Cosmogenic chlorine-36 from calcium  
502 spallation. *Geochim. Cosmochim. Acta* 60, 679–692. [https://doi.org/10.1016-](https://doi.org/10.1016/0016-7037(95)00429-7)  
503 [7037\(95\)00429-7](https://doi.org/10.1016/0016-7037(95)00429-7)  
504 Tosaki, Y., Tase, N., Sasa, K., Takahashi, T., Nagashima, Y., 2012. Measurement of the <sup>36</sup>Cl deposition  
505 flux in central Japan: natural background levels and seasonal variability. *J. Environ. Radioact.*  
506 106, 73–80. <https://doi.org/10.1016/j.jenvrad.2011.11.010>  
507 White, P.J., Broadley, M.R., 2001. Chloride in Soils and its Uptake and Movement within the Plant: A  
508 Review. *Ann. Bot.* 88, 967–988. <https://doi.org/10.1006/anbo.2001.1540>  
509  
510