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Determination of the scavenging coefficients of gaseous and particulate chlorine 36 by rain

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Chlorine radioactive isotope

Long-lived radionuclide ($t = 301,000$ years)

$$\text{Activity} = 1,22 \times 10^9 \text{ Bq.g}^{-1}$$



*High solubility
and
bioavailability*

(Bastviken et al., 2013)



*Natural
and
anthropogenic
sources*

(Pupier et al., 2016)



*Lack of data on his
fate and
dosimetric impact*

(Tanaka and Thiry, 2019)
(Le Dizès and Gonze, 2019)

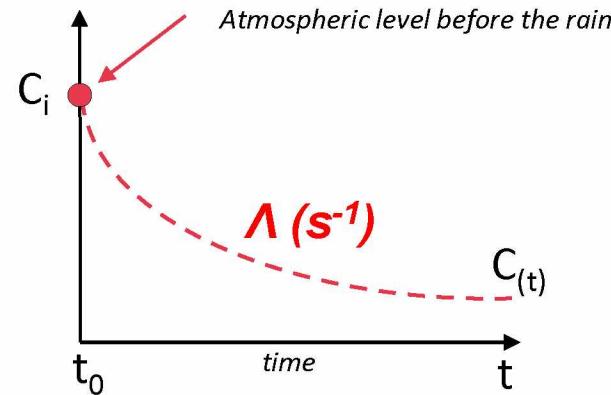
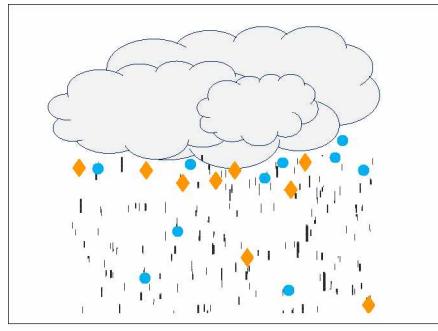


*Wet and dry
deposition*

(Lazarev, 2003)

The scavenging coefficient (λ) reflects the amount of gas or particulate matter arriving on the ground via rainfall and allows a deposition flux to be determined (Laguionie et al., 2014)

- ◆ Particles
- Gaseous species



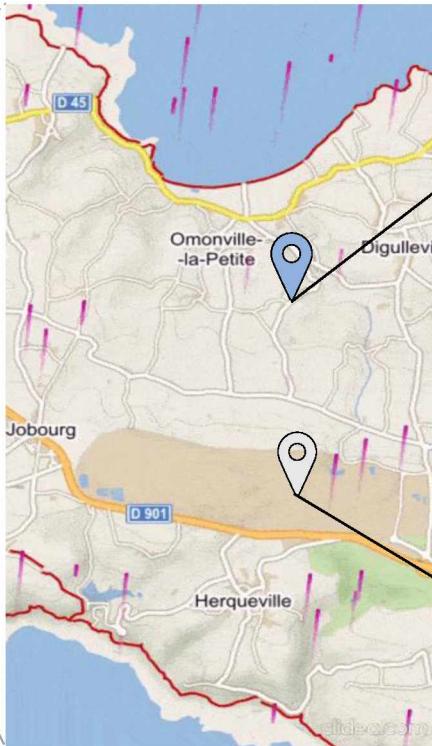
The scavenging coefficient depends on parameters such as aerodynamic diameter (*particles*), solubility and Henry's constant (*gases*), rain intensity, drop size, below cloud height...

(Duhanyan and Roustan, 2011)

So, the objective of this study was to determine experimentally the scavenging coefficients of particulate and gaseous ^{36}Cl by rain

Study site

- Chronic emission of small quantities of chlorine 36 and krypton 85 by Orano La-Hague plant
- $^{36}\text{Cl}/\text{Cl}_{\text{tot}}$ ratio measurement on PTILH (particles, gases, grass) 100 times higher than natural background
- **Unique site for the experimental study of chlorine 36 deposits**



Windy data ; 17/10/2021



Technical
Plateform
IRSN La
Hague
(PTILH)



Nuclear
waste
reprocessing
center
Orano La
Hague

Aerosol sampling in chlorine 36 plume (*Krypton 85 as tracer*)

Objective : Determine the size fraction of chlorine 36 emitted by Orano La-Hague plant

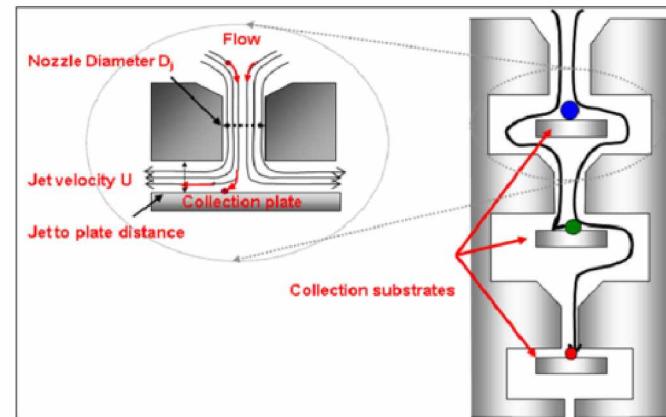
Specifications

Particle collection area	Ø 25 mm
Particle size range	30 nm - 10 μm
	0-10 μm with filter stage
Sample flow rate	10 or 30 l/min
Impactor low pressure	100 mbar
Max temperature	200 °C
Weight	5 kg
Material	Stainless steel (AISI 316)

13 stages



Low Pressure Impactor (LPI) DEKATI



Working principle of LPI DEKATI
(Verma et al., 2016)

Determination of particles scavenging coefficients by double tracking



**Electrical Low Pressure
Impactor (ELPI[®]+) DEKATI**

*It measures particles concentration and
size distribution (14 size fractions) in
real-time at 10 Hz sampling rate*



**Optical disdrometer OTT
Parsivel**

*Measures particle size and
velocity of solid and liquid
precipitation, rain intensity,...
in real time*

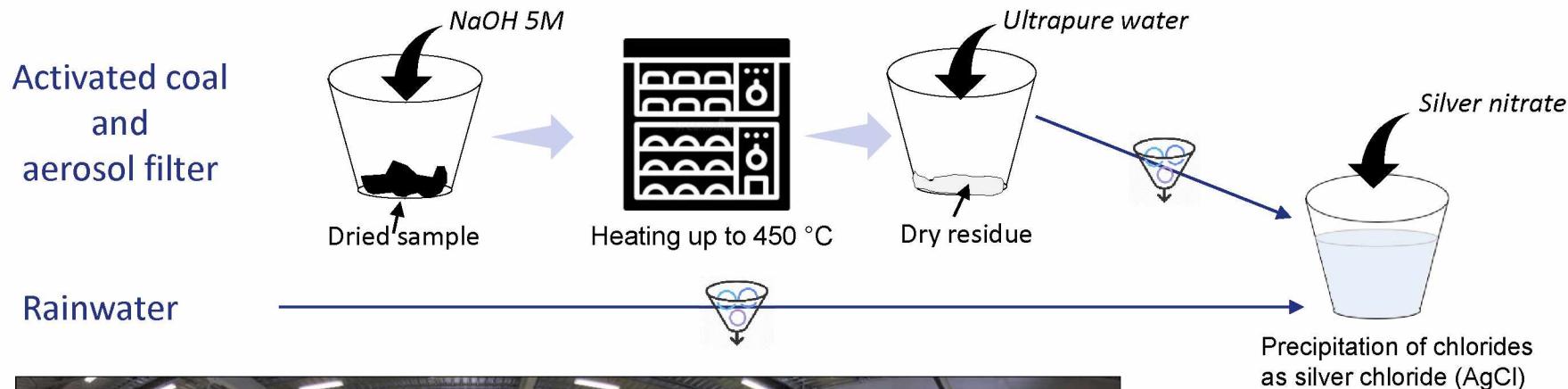
Two-weeks sampling of atmospheric (gaseous and particles) and rainwater ^{36}Cl



Rain collector
Eigenbrodt
*with a detector
of rain*

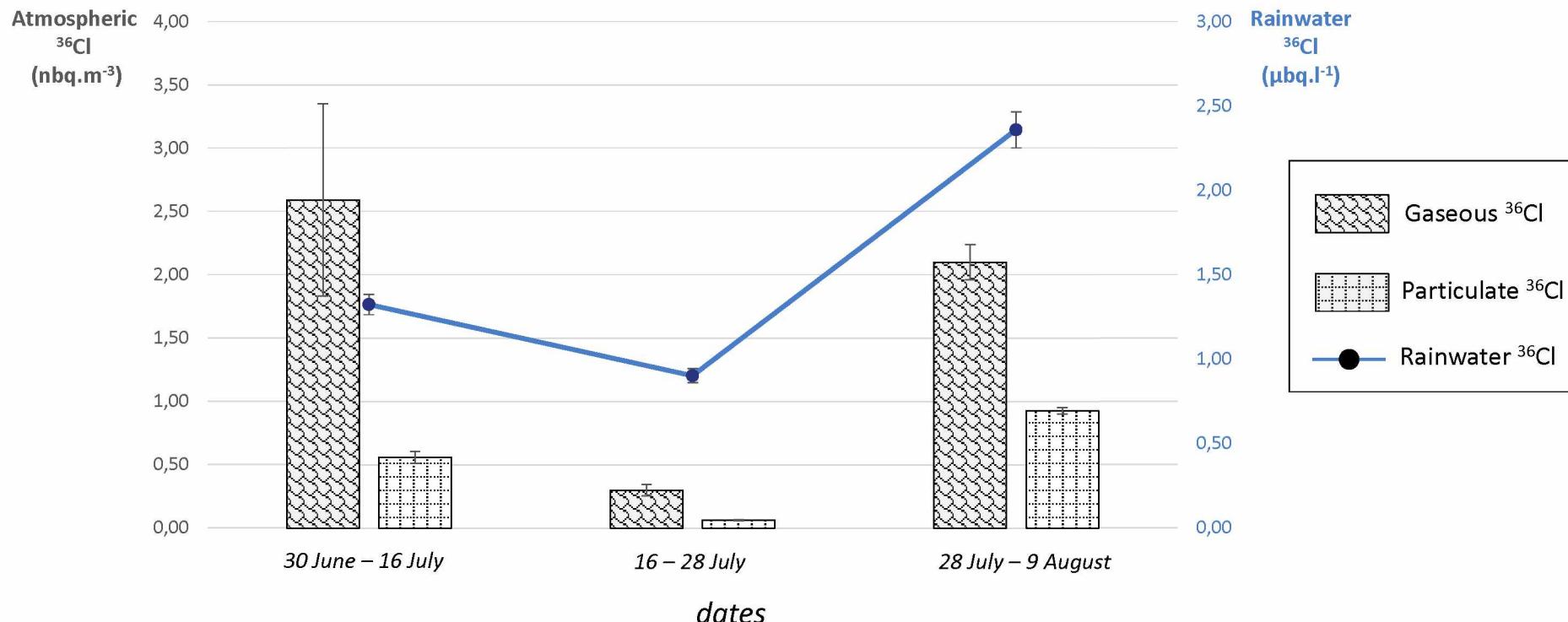
Atmospheric sampler
SDEC AS3000

Samples treatment and analysis

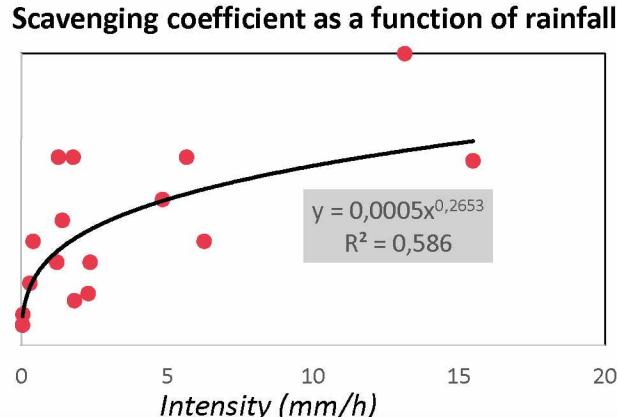
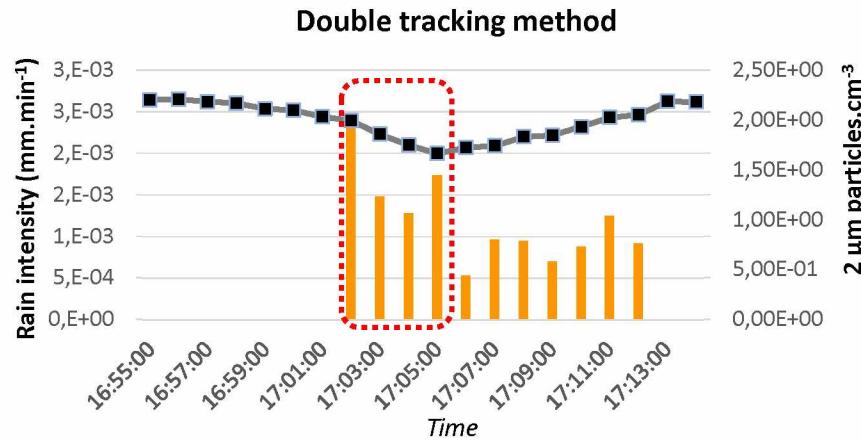
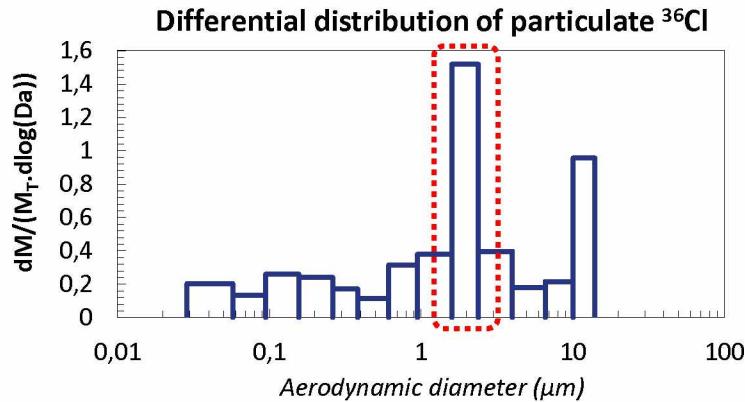


Accelerated Mass spectrometer (AMS) - Accelerator for Environmental, Earth and Risk Sciences (ASTER)

^{36}Cl content in atmosphere and rainwater



Particulate ^{36}Cl scavenging coefficients



Amount of particulate ^{36}Cl scavenged

$$\Lambda = A \times P_o^B \quad (\text{Andronache, 2002})$$

$$C_{(t)} = C_0 \times e^{-\Lambda(t)} \quad (\text{Seinfeld and John H., 1997})$$

$$C_{(t)} - C_0 = \text{particulate } ^{36}\text{Cl scavenged}$$

- P_o = rain intensity (mm.h^{-1})
- Λ = scavenging coefficient (s^{-1})
- C_0 = average concentration in the atmosphere (nbq.m^{-3})
- $C_{(t)}$ = atmospheric concentration at t time (nbq.m^{-3})
- t = rain duration (s)

Gaseous ^{36}Cl scavenging coefficients

Generic expression for the scavenging coefficient of solubles gases

$$\Lambda = \frac{6 \times 10^{-3} P_0 K_c}{U_r \times D_r} \quad (\text{Sportisse, 2007; Hewitt and Jackson, 2008; Seinfeld and Pandis, 2016})$$

$$K_c = \frac{D_g}{D_r} \left[2 + 0,6 \left(\frac{D_r U_r}{\nu} \right)^{1/2} \left(\frac{\nu}{D_g} \right)^{1/3} \right] \quad \text{Hewitt and Jackson, 2008}$$

with -

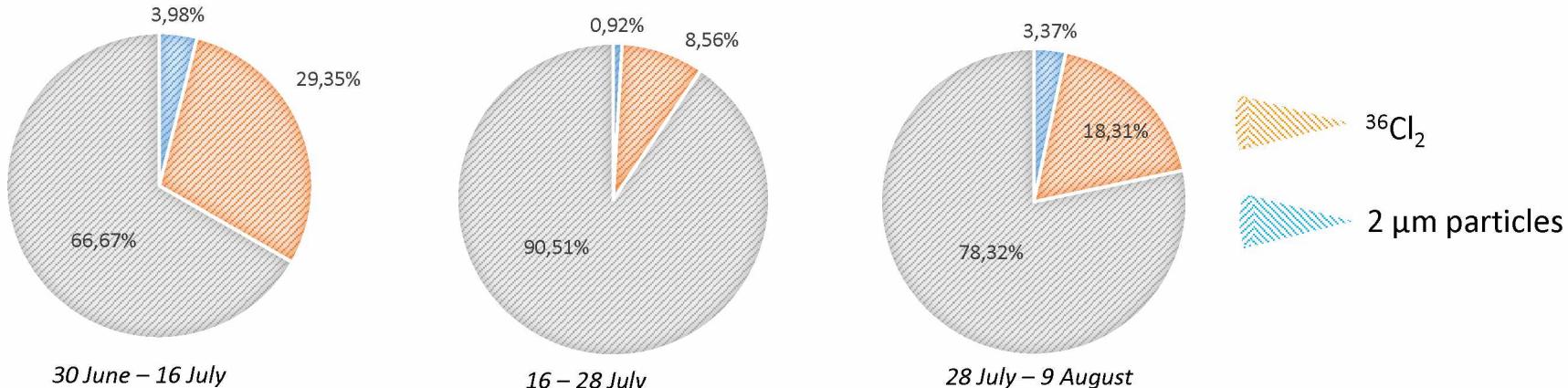
$$D_r = 0,243 P_0^{0,214} \quad (\text{Andronache, 2004})$$

$$U_r = 130\sqrt{Dr} \quad (\text{Kessler, 1995})$$

- P_o = rain intensity (mm.h^{-1})
- Λ = scavenging coefficient (h^{-1})
- K_c = Mass tranfer coefficient (m.s^{-1})
- D_g = diffusivity of the gas ($\text{m}^2.\text{s}^{-1}$)
- ν = Cinematic viscosity of air ($\text{m}^2.\text{s}^{-1}$)
- U_r = Terminal raindrop velocity (m.s^{-1})
- D_r = Raindrop diameter (m)

- Considering that ^{36}Cl is only bonded on 2 μm particle diameter and all scavenged particles were dissolved
- Considering that gaseous ^{36}Cl is only on Cl_2 form and neglecting other forms
- Considering a below-cloud height (or chlorine 36 plume height) of 300 m
- With a ^{36}Cl gaseous scavenging coefficient of around $5 \cdot 10^{-4} \text{ s}^{-1}$

Theoretical amount of chlorine 36 scavenged by rain vs. measured amount



Conclusions

Gaseous and particulate ^{36}Cl scavenging coefficients are similar to some in litterature

(Andronache, 2004; Sportisse, 2007; Duhanyan and Roustan, 2011)

The scavenging of particles is significantly lower than gaseous ones: “greenfield gap” effect??

Theoretical rainwater ^{36}Cl < measured rainwater ^{36}Cl : *influence of others factors?*

What's next??

Improvement of modelled rainwater ^{36}Cl

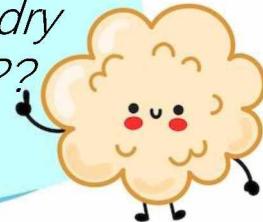
Validation of double tracking method for particles with particulate ^{210}Pb

Integration of a relative below-cloud height (or ^{36}Cl plume height) → *more accuracy*

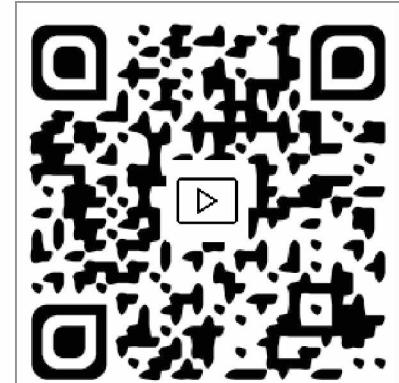
Thanks
for your
attention



*Curious about
chlorine 36 dry
deposition ??*



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