Radioactive gaseous releases from Belgian subclass IIA facilities

Real measurements in the environment

14/09/2018

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Preamble

- Belgian 20/07/2001 RD prescribes the nuclear facilities classification :
  - Class I (NPP, waste treatment facilities, ...), Class III, IV (out of scope)
  - Class II :

«Class IIA»

- facilities producing or conditioning radionuclides from irradiated fissile substances ;
- particle accelerators ;
- facilities containing high activity sources (irradiators, ...) ;

- nuclear medicine ;
- X-rays generators with nominal peak voltage > 200 kV ;
- ...

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Preamble (2)

- Among these IIA facilities, only cyclotrons irradiating liquid and/or gaseous substances or producers/conditioners of such substances can produce significant amounts of radioactive gaseous effluents.
- Measurement of these releases in the environment is not trivial.
  - ↳ Measurement are performed in the shaft or in the chimney, before the effluents release into the atmosphere.

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1. Positron emitters for metabolic imaging (1)

- Positron Emission Tomography is a powerful nuclear medicine technique able to diagnose early and follow the treatment of many diseases, including cancers.
- Radio-pharmaceuticals are labelled with positron emitting radionuclides. Positron (β^+) annihilates by production of two 511 keV photons.
- Positron Emission Tomography allows to represent the 3D distribution of the positron emitters as a function of the metabolic activity.

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1. Positron emitters for metabolic imaging (3)

Two important positron emitters produced by cyclotron :

^{18}F ⇨ 2-[^{18}F]-fluoro-2-deoxy-D-glucose (FDG), …

$t_{1/2}$: 109,7 min.

$^{18}O(p, n)^{18}F$: irradiation of $H_2{}^{18}O$ by protons

^{11}C ⇨ [^{11}C]CO2, [^{11}C]-methionine, …

$t_{1/2}$: 20,5 min.

$^{14}N(p, \alpha)^{11}C$: irradiation of gaseous nitrogen by protons

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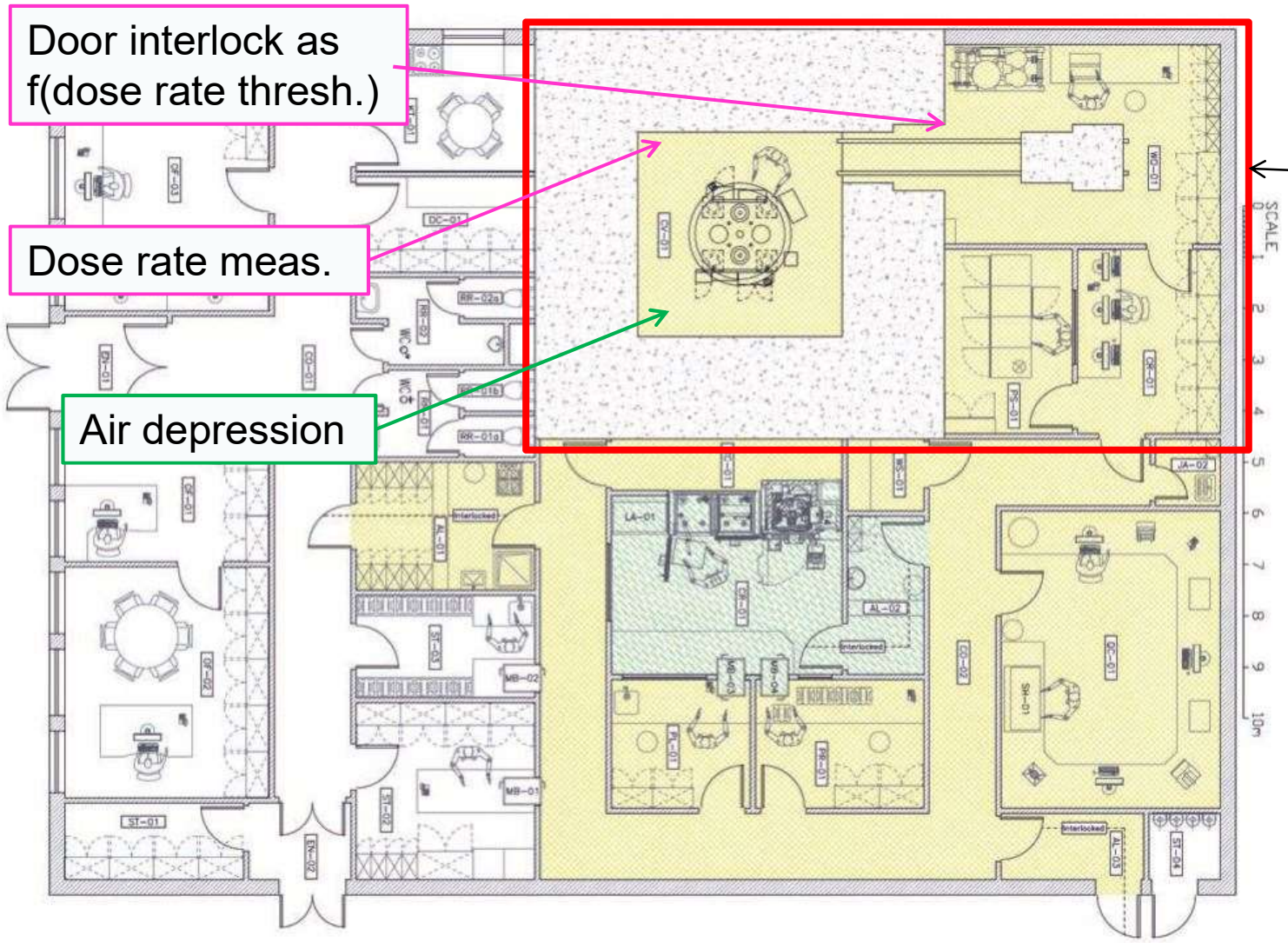
2a. Structure of a FDG facility : Particles accelerators (1)



IAEA RRS n° 3

2a. Structure FDG facility : Particles accelerators (2)

- Nuclear safety issues :
  - High dose rate during irradiation and several hours after :
    - ↳ shielding
    - ↳ interlock on the vault door as a f(doserate)
- Radiation protection issues :
  - Presence of gaseous radionuclides in the vault atmosphere :
    - ↳ air depression in vault
      - Activation of air by neutrons:

        $^{14}N(n, 2n)^{13}N$: β^+ emitter (t1/2 : 9,97 min)

        $^{16}O(n, p)^{16}N$: β^- emitter (t1/2 : 7,13 sec)
      - If leakage from target, then dispersion of aerosols or gas containing ^{18}F and other impurities coming from the target.

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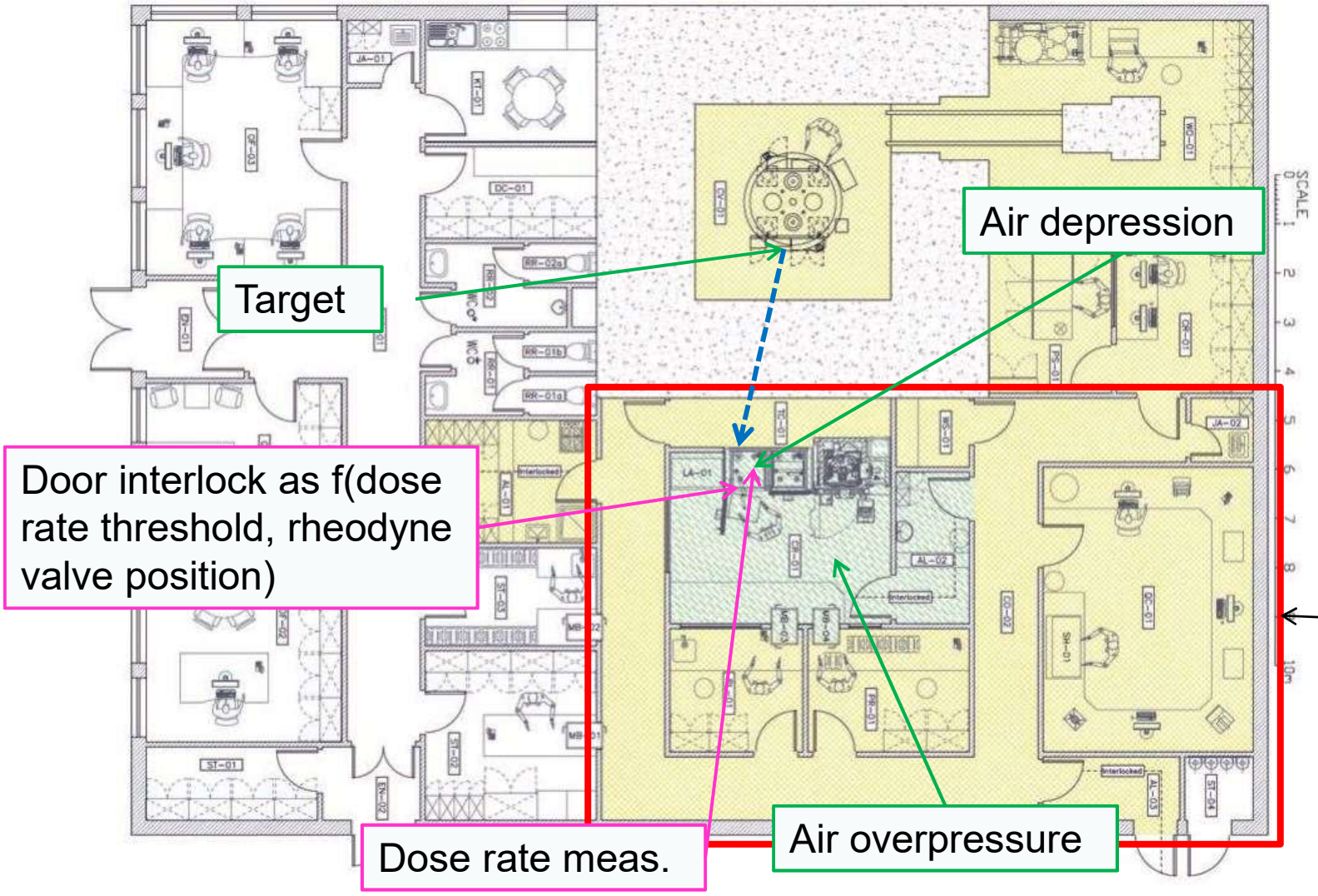
2b. Radionuclides producers – conditioners (1)



IAEA RRS 3

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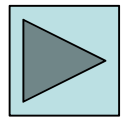
2b. Radionuclides producers – conditioners (2)

- Nuclear safety & radiation protection issues :
  - High dose rate in shielded cells during target transfer, synthesys and dispending, and during following hours :
    - ↳ shielding
    - ↳ interlock on the cell door as a f(doserate)



- Production of radioactive gaseous effluents in routine :
  - in the target : $^{16}O(p, \alpha)^{13}N$: β^+ emitter (t1/2 : 9,97 min)
    - ↳ Evacuation of ^{13}N at start of ^{18}F solution transfer to hot cell
  - in hot cell : HF, Fluor-compounds during FDG synthesis
    - ↳ Dispersion of volatile ^{18}F-compounds in hot cell and in ventilation.

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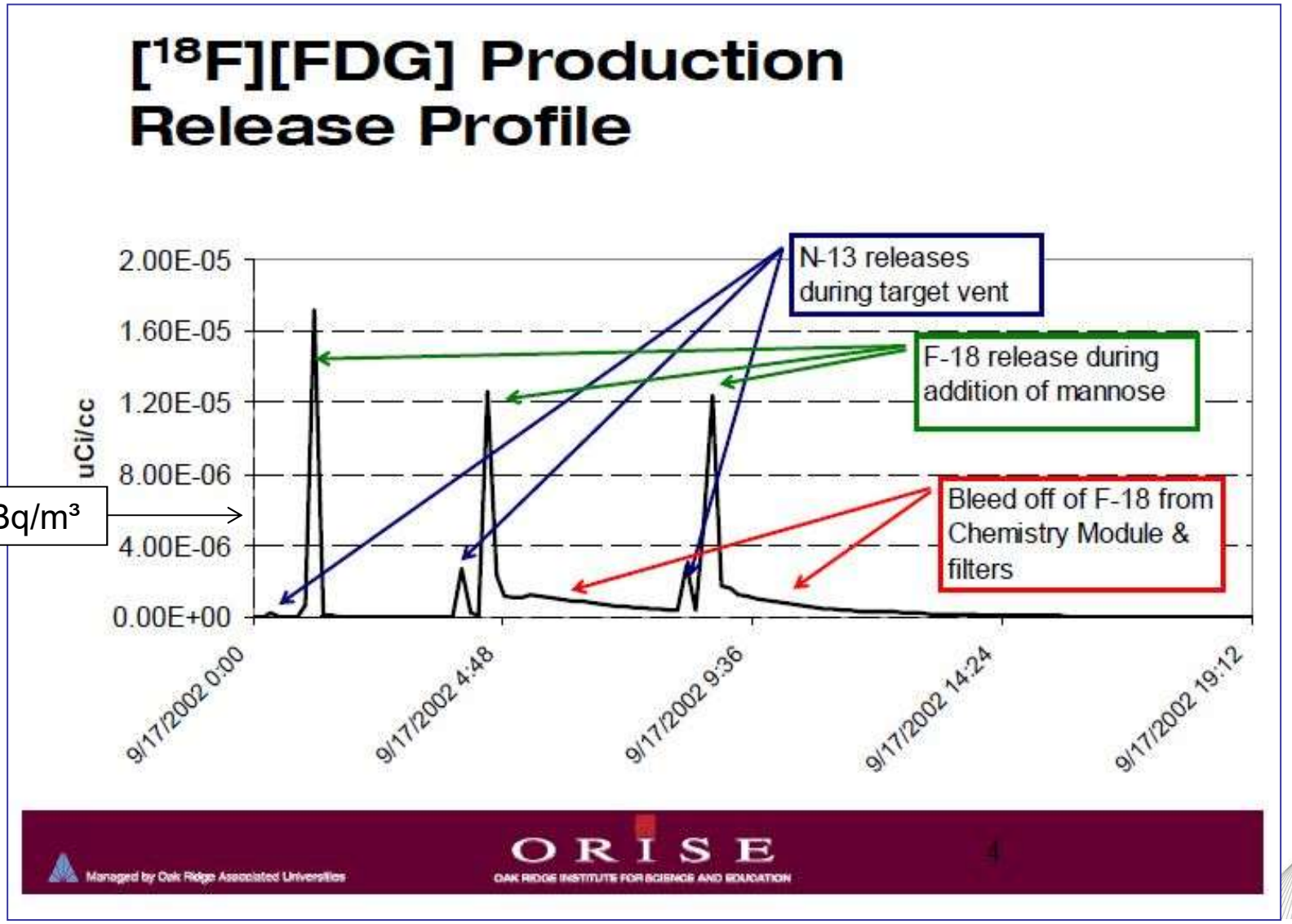
2b. Radionuclides producers – conditioners (3)



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2b. Radionuclides producers – conditioners (4)

To limit the radioactive effluents dispersion in the hot cells during the synthesis :

- Collecting the β^+ volatile compounds and wait significant decay before to bleed to the chimney :
  - ↳Connecting plastic bags on the synthesis module (or compressing the effluent in specific tank)
- Filter on the hot cell air extraction.

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3. Regulation about β^+ gaseous releases (1)

- Belgian RD 20/07/2001 prescribes the radioactive gaseous release limits which are authorised by the FANC for each licensee.
- See in table H2, appendix III of the RD 20/07/2001

  Release limits leading to 1 mSv/y (conservative scenario) :

  ^{18}F : 4500 Bq/m³ (F)

  2200 Bq/m³ (M)

  2100 Bq/m³ (S)

- Issues :

  - How to understand these values ? To be verified for each m³ of air evacuated from FDG production center ?
  - Is the radiation monitoring at the chimney able to measure so low activity concentration ?

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3. Regulation about β^+ gaseous releases (2)

Presentation of the FANC approach during stakeholders meeting on 9/11/2009 :

- FANC analysed the situation in the different facilities producing ^{18}F and ^{11}C radiopharmaceuticals about their installations and their productions.
- FANC presented to the stakeholders some key points in order to correctly manage the radioactive gaseous effluents released from the positron emitting radionuclides producing facilities.

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3. Regulation about β^+ gaseous releases (3)

Key points :

- Identify the **critical steps** in the production processes for the correct management of the gaseous releases
- Install **physical barriers** or **chemical traps** to confine radiaoctive gaseous
- Inform and **sensibilise** the operators to these issues
- Need for **preventive maintenance** of these barriers in order to avoid defaults leading to release
- **Monitoring** of the releases in order to evaluate the integrity of the barriers, the correct application of the work procedures and the respect of the releases limits

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3. Regulation about β^+ gaseous releases (4)

- In accordance with the RD 20/7/2001, FANC is able to allow a **specific release authorisation during normal operation** of the facility.

- This authorisation identifies the **nature of the radioactive substances** which could be released, and the **release conditions** :

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3. Regulation about β^+ gaseous releases (5)

- Annual dose to the public due to the releases < **200 µSv**
- **12** consecutive **months** total β^+ release (^{15}O,^{13}N,^{11}C,^{18}F) < **1,85 TBq** (50 Ci)
- Decreasing release fraction as a function of the production (ALARA process) :
  - 3 % for the production range 0 - 18,5 TBq
  - 1 % for the production range 18,5 – 111 TBq
  - 0,5 % for the production range 111– 185 TBq.
- **Daily** release < **18,5 GBq**
- Monthly report to the FANC / Bel V of the released activity.
- Annual declaration and evaluation of the radiological impact to the public.

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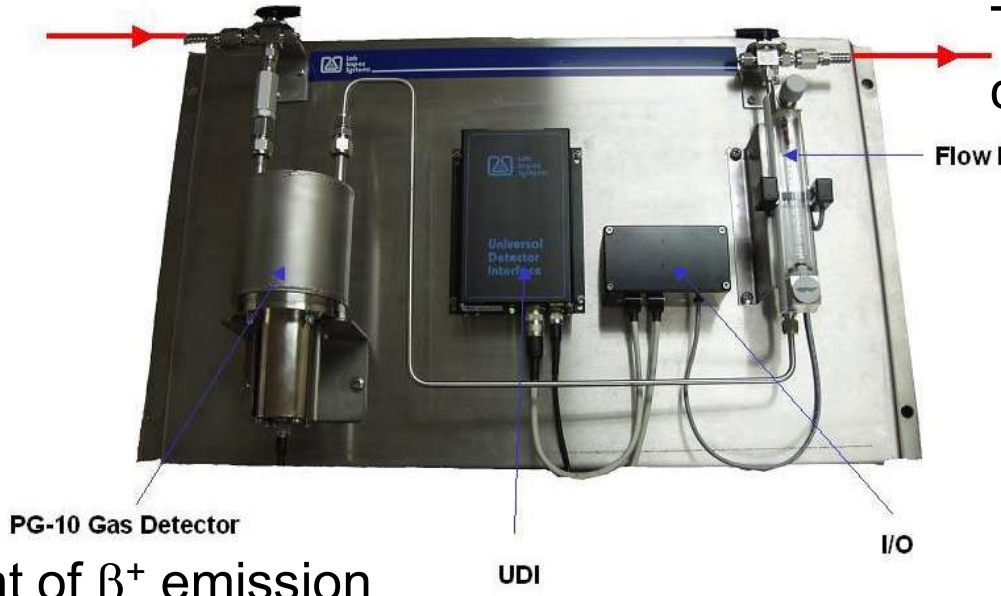
4. Monitoring of radioactive gaseous effluents (1)

- In order to verify the compliance to the releases limits.
- Different designs are possible :

Sampling from the chimney or shaft.



Then back to the chimney or shaft.

Measurement of β^+ emission by scintillator in a chamber.

From Lab Impex

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4. Monitoring of radioactive gaseous effluents (2)

- Detectors placed against the chimney or shaft wall :
  - γ detector or gas proportional counter
  - plastic scintillators

    (detection 511 keV annihilation photons)

From Victor Leach (2013) and Thermo Scientific

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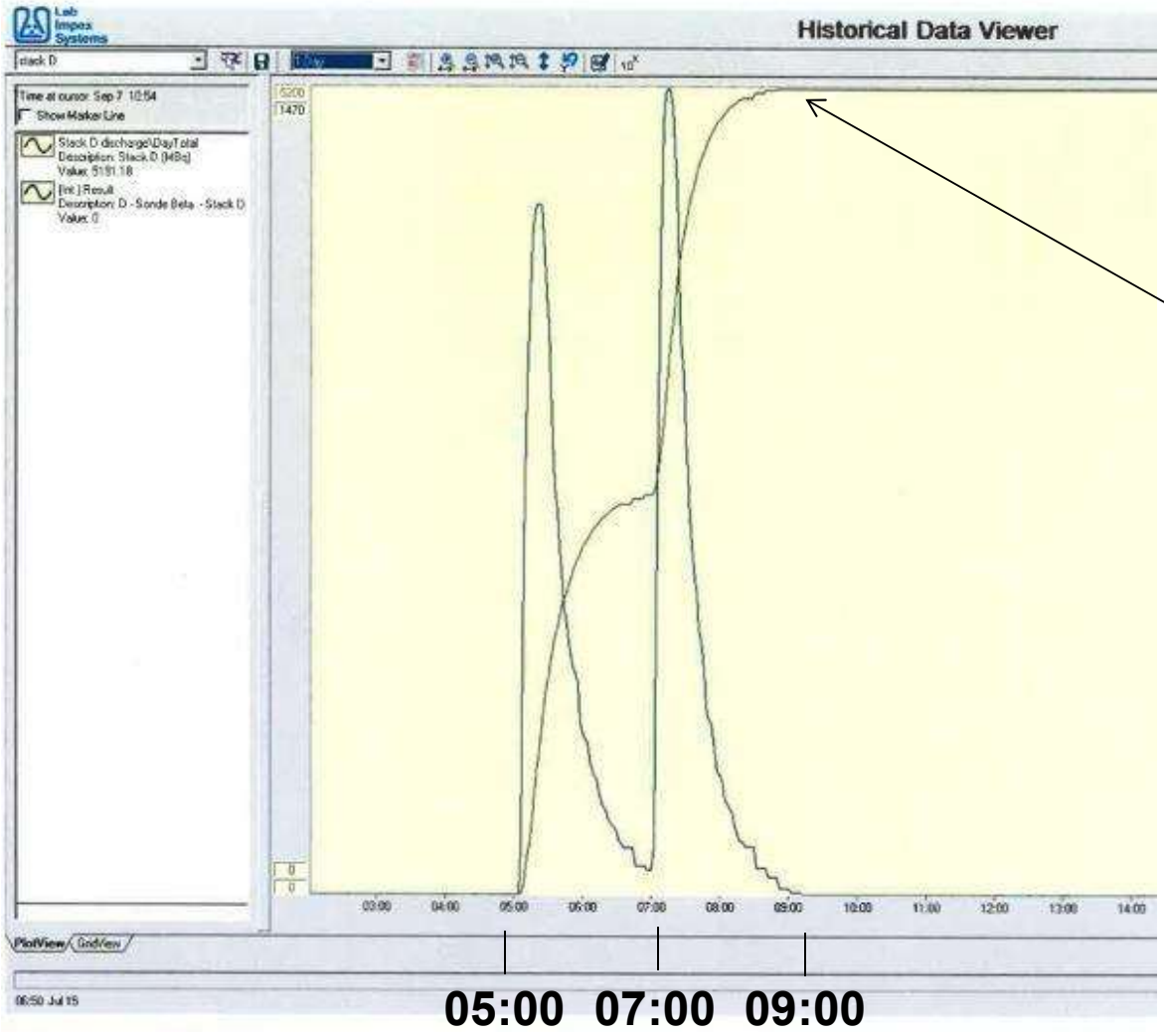
4. Monitoring of radioactive gaseous effluents (3)



Graph showing β^+ releases during two FDG productions :

- Total released activity : 5190 MBq

From Beta+ Pharma (WSL)

4. Monitoring of radioactive gaseous effluents (4)

- Operating experience feedback of the monitoring :
  - Fluor-compounds behaviour : adsorption on the inner surface of the chimney ⇨ overestimation of the activity
  - Software analysing the signal. Issue with the dead time of the system ⇨ overestimation of the activity
- So need for a correct calibration of the system :
  - FANC recommends test of release of known ^{11}C activity in the hot cell in order to verify the correct response of the system.

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DOSSIER D'INFORMATION

Rejets d'effluents radioactifs des établissements industriels de classe IIA en 2017

Mai 2018

AFCN
agence fédérale de contrôle nucléaire

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5. Information file (1)

FANC presented this file during the stakeholders meeting IIA on 22/11/2012 :

- Objectives : to inform the public on the annual releases and on the radiological impact
- FANC Values :
  - Transparency
  - Openness
  - Objectivity
  - Exhaustivity.

5. Information file - Context (2)

- Application of the law 5 of August 2006 about the access of the public to the information related to the environment (MB 28.08.2006);
  - **Article 12: «** *Chaque instance (…) prend les mesures nécessaires pour organiser les informations environnementales dont elle dispose et qui sont en rapport avec ses fonctions, en vue de permettre leur mise à disposition active et systématique auprès du public, notamment en utilisant les moyens de communication électronique* **».**

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Information file - Context (2')

- Application of the law 5 of August 2006 about the access of the public to the information related to the environment (SB 28.08.2006);
  - **Artikel 12: «** *Elke milieu-instantie (…) neemt de nodige maatregelen om de milieu-informatie, die relevant is voor haar taak en waarover ze beschikt, te orderen met de bedoeling ze actief en systematisch onder het publiek ter beschikking te stellen, met name door de elektronische communicatie* ».

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5. Information file - Content (3)

- The file is organised in 2 parts :
  - **Generic part**, common to all the Belgian subclass IIA facilities ;
  - **Specific** part for each facility:
    - Description of the activities
    - Radioactive gaseous effluents releases during 2017
    - Results assessment and radiological impact to the public in 2017.

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5. Information file - Content (4)

IIA facilities with specific release authorisation

- Bêta Plus Pharma (site des cliniques universitaires St Luc)
- Cliniques de l'Université Libre de Bruxelles (Hôpital Erasme)
- Universitair Ziekenhuis van Vrije Universiteit Brussel (VUB)
- Universitair Ziekenhuis van Leuven
- Universitair Ziekenhuis van Antwerpen
- Universitair Ziekenhuis van Gent
- IRE ELiT (Fleurus)
- Université de Liège

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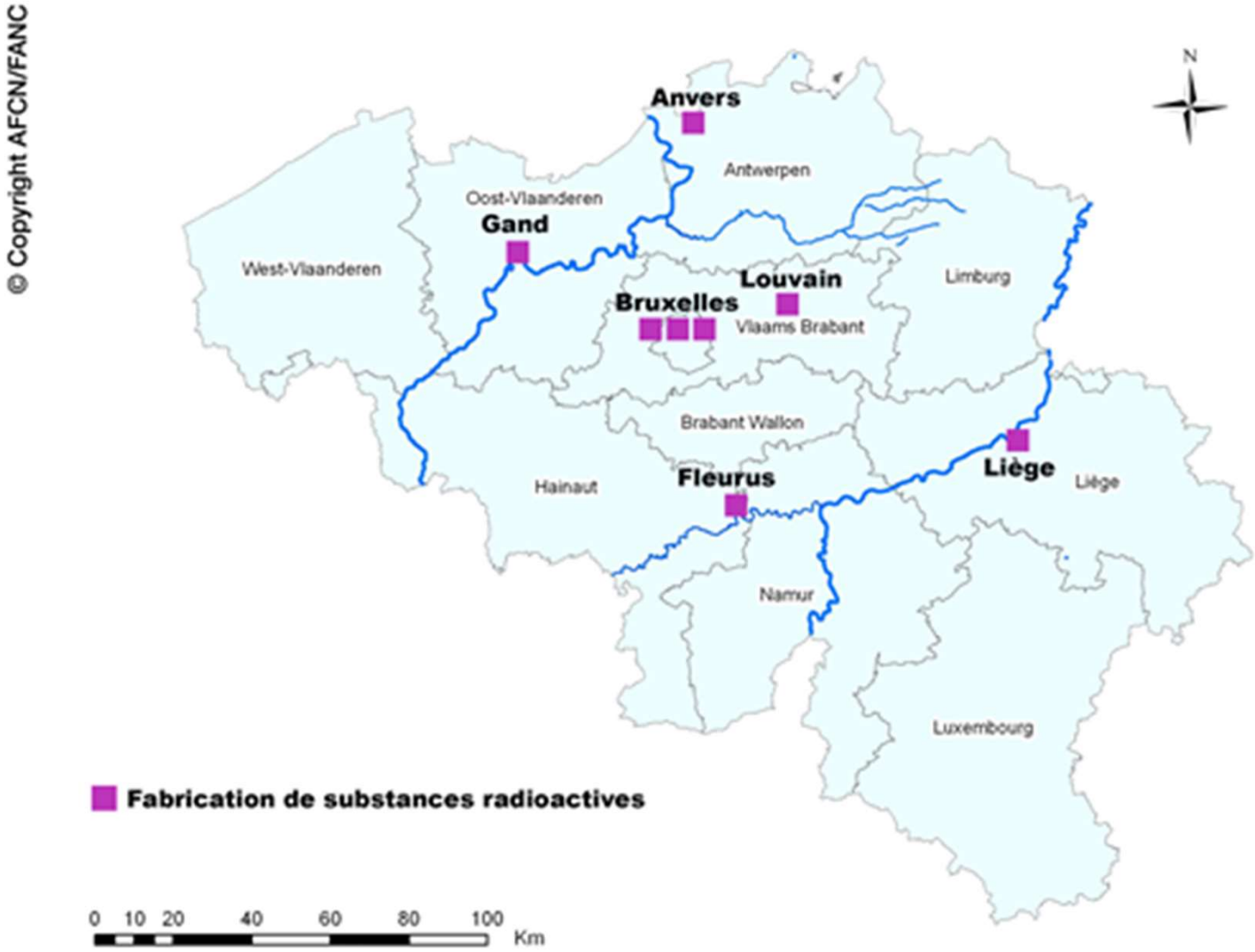
IIA facilities with specific release authorisation



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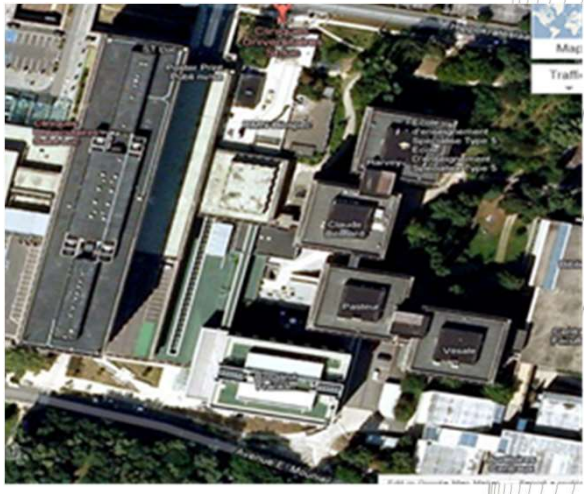
Bruxelles — Bêta Plus Pharma

Implantation :	Woluwe-Saint-Lambert (1200), Bruxelles
Activité(s) principale(s) :	Fabrication de substances radioactives pour la médecine nucléaire, recherche médicale
Type(s) d'installation(s) :	Cyclotron et installations annexes
Première mise en service :	2004
Phase de vie actuelle :	En exploitation
Contrôle des rejets potentiels :	Liquides : • pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)
	Atmosphériques : • *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)



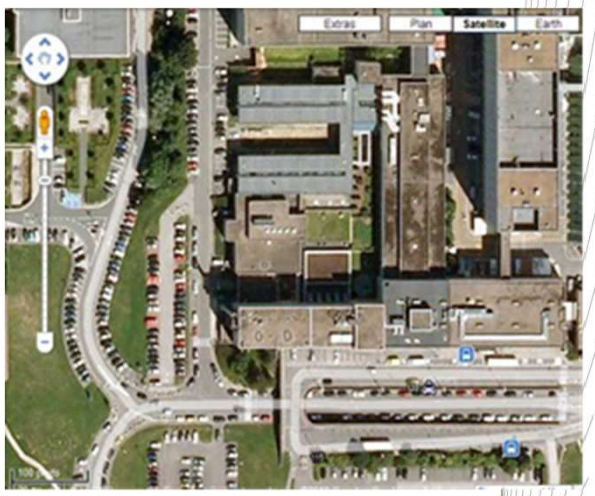
Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	1421	1497	1771	1438	1488

The released activity represents **0,62 %** of the produced activity in 2017.
The maximal radiological impact to the public, calculated for these releases, is equal to **0,010 mSv**.

Bruxelles — Vrije Universiteit Brussel (VUB)

Implantation :	Jette (1090), Bruxelles
Activité(s) principale(s) :	Fabrication de substances radioactives pour la médecine nucléaire, recherche en physique
Type(s) d'installation(s) :	Cyclotron et installations annexes
Première mise en service :	1985
Phase de vie actuelle :	En exploitation
Contrôle des rejets potentiels :	Liquides : • pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)
	Atmosphériques : • *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)



Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	42,5	24,45	18	14	11

The released activity represents **0,15 %** of the produced activity in 2017.
The maximal radiological impact to the public, calculated for these releases, is equal to **0,001 mSv**.

Bruxelles — Hôpital Erasme

Implantation : Anderlecht (1070), Bruxelles

Activité(s) principale(s) : Fabrication de substances radioactives pour la médecine nucléaire, recherche médicale et formation

Type(s) d'installation(s) : Cyclotron et installations annexes

Première mise en service : 1989

Phase de vie actuelle : En exploitation

Contrôle des rejets potentiels :

Liquides :
- pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)

Atmosphériques :
- *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)

Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	198	630	599	320	520

The released activity represents **1,07 %** of the produced activity in 2017.
The maximal radiological impact to the public, calculated for these releases, is equal to **0,005 mSv**.

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Louvain — Hôpital universitaire de Louvain

Implantation : Louvain (3000), Brabant flamand, Flandre

Activité(s) principale(s) : Fabrication de substances radioactives pour la médecine nucléaire, recherche médicale

Type(s) d'installation(s) : Cyclotron et installations annexes

Première mise en service : 2004

Phase de vie actuelle : En exploitation

Contrôle des rejets potentiels :

Liquides :
- pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)

Atmosphériques :
- *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)

Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	394	332	354	479	465

The released activity represents **0,29 %** of the produced activity in 2017.
The maximal radiological impact to the public, calculated for these releases, is equal to **0,001 mSv**.

Anvers — Hôpital universitaire d'Anvers

Implantation : Edegem (2650), Anvers, Flandre

Activité(s) principale(s) : Fabrication de substances radioactives pour la médecine nucléaire, recherche médicale

Type(s) d'installation(s) : Cyclotron et installations annexes

Première mise en service : 2011

Phase de vie actuelle : En exploitation

Contrôle des rejets potentiels :

Liquides :
- pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)

Atmosphériques :
- *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)

Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	106	125,3	150	125	153

The released activity represents **0,30 %** of the produced activity in 2017.
The maximal radiological impact to the public, calculated for these releases, is equal to **0,011 mSv**.

Gand

Hôpital Universitaire de Gand (ex Bêta Plus Pharma)

Implantation : Gand (9000), Flandre orientale, Flandre

Activité(s) principale(s) : Fabrication de substances radioactives pour la médecine nucléaire, recherche médicale

Type(s) d'installation(s) : Cyclotron et installations annexes

Première mise en service : 2006

Phase de vie actuelle : En exploitation

Contrôle des rejets potentiels :

Liquides :
- pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)

Atmosphériques :
- *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)

Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	215	219	199	342	359

The released activity represents **0,66 %** of the produced activity in 2017.
The maximal radiological impact to the public, calculated for these releases, is equal to **0,002 mSv**.

Fleurus — IBA s.a./IRE ELiT s.a.

Implantation : Fleurus (6220), Hainaut, Wallonie

Activité(s) principale(s) : Fabrication de substances radioactives pour la recherche médicale

Type(s) d'installation(s) : Cyclotron et installations annexes

Première mise en service : 2001

Phase de vie actuelle : En exploitation

Contrôle des rejets potentiels :

Liquides :
- pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)

Atmosphériques :
- *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)

Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	1,46	2,9	0	0	0

Interprétation des résultats de l'année 2017

En 2017, il n'y a pas eu d'activité pouvant engendrer des rejets gazeux radioactifs.

Liège — Université de Liège

Implantation : Liège (4000), Liège, Wallonie

Activité(s) principale(s) : Fabrication de substances radioactives pour la médecine nucléaire, recherche médicale

Type(s) d'installation(s) : Cyclotron et installations annexes

Première mise en service : 1999

Phase de vie actuelle : En exploitation

Contrôle des rejets potentiels :

Liquides :
- pas de rejets directs d'effluents radioactifs liquides (gérés en tant que déchets radioactifs)

Atmosphériques :
- *émetteurs* β^+ (^{11}C, ^{13}N, ^{15}O, ^{18}F)

Résultats des rejets des années 2013-2017

Rejets atmosphériques	Activité totale rejetée				
	2013	2014	2015	2016	2017
Emetteurs β^+ (GBq)	358,6	186,5	334	341	409

The released activity represents **1,57 %** of the produced activity in 2017.
The maximal radiological impact to the public, calculated for these releases, is equal to **0,005 mSv**.

5. Information file – Conclusions (14)

- Since 2013 :
  - FANC reports the annual activity declared by subclass IIA facilities.
  - Better management of the radioactive gaseous effluents releases.
  - Globally, the facilities respect the authorised releases limits.
  - Maximal annual radiol. impact to public < 10 µSv
  - Sometime, excess of the daily limit is declared to FANC / Bel V ⇨ see next point.

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β^+ gaseous releases
Operating experience feedback (1)

- 2010/2011 – 2017 : about 30 releases of β^+ emitters > daily limit of 18,5 GBq.

  Very few events comparing to the thousands of shooting and synthesis of FDG and of other radiopharmaceuticals.

↳ **2 releases of ^{18}F in the cyclotron and in vault due to:**

  - piercing of the cyclotron window target after shooting on empty target. Water in He cooling circuit. Particular event.
  - leakage in the transfer line between the target and the hot cell. Cause : replacement of transfer lines after target maintenance.

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IIA facilities radioactive gaseous releases Operating experience feedback (2)

↳ 28 releases of ^{18}F or ^{11}C in hot cells > daily limit.

- Causes (by decreasing order of occurence):
  - Leakage from the plastic bag collecting effluent from synthesis module ⇨ need for preventive replacement.
  - Leakage in the tubing between the module and the bag.
  - Problems during the chemical process (testing phase).
  - Inadequate design of the effluent collecting system or of the filtration at the outlet of the hot cell.
  - Procedure of checking the effluents collection system from synthesis module not correctly followed by the operator.

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Conclusions on β^+ gaseous releases management (1)

- FANC **identified issues** about radioactive effluents during the production of radiopharmaceuticals labelled with positron emiting radionuclides.
- FANC highlights **active participation** of each stakeholder in order to improve the installations (barriers) / procedures / radiomonitoring in order to limit / control the releases to the environment.
- **Harmonisation** of the FANC authorisations .
- Radiological impact to the population of the routine is **very low** (< 0,010 mSv).
- Globally the release limit is **respected** by the facilities.

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Conclusions (2)

- **Recommendations** for a correct radioactive gaseous effluents management in β^+ producing facilities :
  - Correct **design** of the effluent **collecting** system
  - **Preventive replacement** of the most fragile components (bag, connectors, tubing)
  - **Awareness of the operators** and correct application of the procedures
  - Correct **design** of the **monitoring** of the radioactive releases through the chimney
  - **Correct calibration** of this monitoring.

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Many Thanks to Pascal Carlier (AFCN) for the technical and regulatory work

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Do you have any question ?

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References

- 20/07/2001 Royal Decree : Arrêté royal du 21 juillet 2001 portant règlement général de la protection de la population, des travailleurs et de l'environnement contre le danger des radiations ionisantes (Moniteur belge du 30/08/2001).
- Dossier d'information sur les rejets radioactifs gazeux des établissements industriels IIA. Agence Fédérale de Contrôle Nucléaire : https://afcn.fgov.be/fr/professionnels/activites-industrielles/classe-iia/rejets
- Informatiedossier over gasvormige radioactieve lozingen. Federale Agentschap voor Nucleaire contrôle : https://fanc.fgov.be/nl/professionelen/industriele-activiteiten/klasse-iia/lozingen

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Appendix

Dose rate from ^{18}F sources as f(activity, geometry) (from D. Delacroix, 2002)

Contamination of the **skin** by 37 GBq ^{18}F (1 Ci) on 1cm² skin

↳ β dose rate to skin : **72.000 Sv/h ?!**

γ dose rate at **1 m of glass vial** containing 370 GBq ^{18}F (10 Ci) : **59 mSv/h**

γ dose rate at **0,3 m of « punctual source »** 370 GBq ^{18}F (10 Ci) : **670 mSv/h**

β dose rate at **0,3 m of « punctual source »** 370 GBq ^{18}F (10 Ci) : **44 Sv/h !?**

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