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Analysis of radionuclide concentration in air released through the stack of a radiopharmaceutical production facility based on a medical cyclotron

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HIGHLIGHTS

- Monitoring of air released through the stack of a Nuclear Medicine Center.
- Identification of main contamination sources and operative procedures optimization.
- Improvements in the protection devices to reduce the air concentration at stack.
- Analysis of concentration data to evaluate dose to population living near the plant.

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ABSTRACT

Positron emitting radionuclides are increasingly used in medical diagnostics and the number of radiopharmaceutical production facilities have been estimated to be growing worldwide. During the process of production and/or patient administration of radiopharmaceuticals, an amount of these radionuclides might become airborne and escape into the environment. Therefore, the analysis of radionuclide concentration in the air released to the stack is a very important issue to evaluate the dose to the population living around the plant. To this end, sampling and measurement of radionuclide concentration in air released through the stack of a Nuclear Medicine Center (NMC), provided with a cyclotron for radiopharmaceuticals production, must be routinely carried out with an automatic measurement system.

In this work is presented the air monitoring system realized at "San Gaetano" NMC at Bagheria (Italy) besides the analysis of the recorded stack relesead air concentration data. Sampling of air was carried out continuously and gamma-ray spectrometric measurement are made on-line and for a short time by using a shielded Marinelli beaker filled with sampled air and a gamma detector. The use of this system allows to have 1440 values of air concentration per day from 2002, year of the start of operation with the cyclotron. Therefore, the concentration values are very many and an analysis software is needed to determine the dose to the population. A comparison with the results of a simulation code based on a Gaussian Plume air dispersion modelling allow us to confirm the no-radiological significance of the stack effluent releases in terms of dose to population and to evaluate possible improvements in the plant devices to reduce the air concentration at stack.

1. Introduction

Positron Emission Tomography (PET) is taking over the years increasing importance as diagnostic tool due to the chance to produce a large activity of short-lived positron-emitting radionuclides by a medical cyclotron located inside the same hospital structure. Workers and environment are protected during cyclotron irradiation and radiopharmaceutical synthesis activities through safety systems based on technologically enhanced facilities. From the point of view of protecting environment and public, a significant issue is the control of activity of volatile radionuclides released during irradiation or chemical synthesis of PET radiopharmaceuticals inside hot cells, or in case of target breaks or failure (Mukherjee, 2002; Pascali et al., 1996 Tomarchio, 2012). Therefore, the health physics surveillance program must include

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an analysis and activity estimation of radioactive gaseous species released through the stack by means of a suitable air monitoring system implemented to limit releases into the atmosphere (Pascali et al., 1996).

As a reference production of ^{18}FDG (fluoro-deoxyglucose) PET radiopharmaceuticals inside "San Gaetano" Nuclear Medicine Center (NMC) located at Bagheria, a small town near Palermo, Italy, was considered. The aim of this work is to examine the layout of the "San Gaetano" NMC stack air monitoring system and the operative conditions to ensure the respect of dose limits and correlate the air concentration values at the stack with monitored environment measurements. Data analysis was necessary to verify radiation levels at stack related to the increase from 2008 of production due to a more intense commercial activity to supply nearby PET centres. Major source of contamination of the air was identified in a release inside the hot cells during the preparation of radiopharmaceuticals and in activated air inside the cyclotron vault. As a result, it is possible to perform a proposal for optimization of plant operations, with the provision of ventilation blocking of cyclotron vault until a few hours after the irradiation, the planning of buffer tanks to contain both hot cells and cyclotron vault release air volumes or long pipes for delaying the discharge of radioactive-labelled gases. Furthermore, a change in the position and height of the stack is able to reduce the radiation concentrations to levels comparable with the environmental background. These conclusions, while referring to a specific NMC, should be extended to all cyclotron plants worldwide in operation.

2. Materials and methods

2.1. Air contamination sources

Main contamination sources of air released at stack can be identified in air activation during the irradiation and production of gases in radiopharmaceutical preparation as well as possible target breaks and other accidents.

The activation of air inside the cyclotron vault is due to the interactions of neutrons produced by charged particles with the target. In order to estimate the level of induced activity in the air, reactions reported in Table 1 can be considered. To evaluate population dose due to the released activated air was conservatively assumed that ventilation is blocked during irradiation and should begin immediately after the end of irradiation, and that the radioactive gas after the release is dispersed in the air within a plume along the wind direction and receiving point always at the center of the plume. In this case, it can be used the equation (Birattari et al., 1986).

$$A(t) = m_{\text{off}} C_s V \frac{1-e^{-(\lambda+m_{\text{on}})t}}{(\lambda + m_{\text{off}})} + m_{\text{on}} C_s V \frac{1-e^{-(\lambda+m_{\text{on}})t}}{(\lambda + m_{\text{on}})} \quad (1)$$

where $A(t)$ is the total activity extracted from the cyclotron vault by the ventilation system after a period t of irradiation, m_{on} and m_{off} are the number of changes of air volume per hour in beam-on and beam-off operative conditions of the cyclotron, C_s is the saturation activity per unit volume, V the volume of the bunker, λ the decay constant of the radioisotope. The saturation activity per unit of volume depends on the neutron fluence rate Φ according to

$$C_s = \frac{\mu \Phi \lambda}{(\lambda + m_{\text{on}})} \quad (2)$$

and

$$\mu = \frac{\sigma \rho f N_A}{P_A} \quad (3)$$

with σ microscopic cross section for the specific reaction, ρ is the target density, f is the isotopic abundance, N_A is the Avogadro number, P_A is the target atomic mass. Main parameters for the considered nuclear reactions are reported in Table 1 (Birattari et al., 1986).

As can be seen, production of ^{41}Ar inside the cyclotron vault is likely. In the case of the Ion beam Application (IBA) CYCLONE 18/9 cyclotron of "San Gaetano" NMC, the yield of the reaction ^{18}O(p,n)^{18}F is approximately 250 mCi/μA which, for a maximum beam current of 80 μA, m_{on} and m_{off} equal to 0.05 h^{-1} and 10 h^{-1}, cyclotron vault volume assumed equal to 50 m^3, results in a neutron fluence rate $\Phi = 3.4 \cdot 10^{10}\ s^{-1}\ m^{-2}$. The saturation activity for reaction ^{40}Ar(n,γ)^{41}Ar is $4.83 \cdot 10^5$ Bq m^{-3} and the induced activity after 2 h of irradiation is about $1.5 \cdot 10^7$ Bq, with a concentration value at stack less than 1 Bq L^{-1} Assumed a dose constraint value of 10 μSv y^{-1} (D_p) for an individual of the population placed 100 m away from the plant (reference group), it can be derived a concentration limit value (C_L) out of the stack using a ground-level centerline Gaussian plume dispersion transport model proposed in NCRP 123 (NCRP, 1996). In this way, a value of C_L of the order of 5 Bq L^{-1} was determined with reference to ^{41}Ar and 20 Bq L^{-1} for ^{18}F, with a conservatively value of the diffusion factor P, equal to 10^{-3}. Therefore, it is sufficient to check that air concentration values at stack should never exceed C_L on average. This may mean that, although higher values can be detected, the average daily values must be significantly lower than C_L.

2.2. Air monitoring system

Fig. 1 shows the schematic diagram of the real-time effluent monitoring system at the "San Gaetano" NMC radionuclide production facility. It includes two independent systems, each provided with a Scionix Holland NaI(Tl) 2 in. × 2 in. scintillation detector, resolution 7% to ^{137}Cs, placed inside a lead-shielded Marinelli beaker provided with connecting valves to select air from different laboratories (Fig. 2). The first system allows the continuous monitoring of the effluent air released to the stack in order to have real-time information on the quantities released into the atmosphere. The second, similar to the previous one, is connected sequentially to cyclotron vault, radiochemistry, quality control and delivery laboratories. The air is drawn automatically from one selected environment through a pump and appropriate valves

Table 1
Neutron activation of air inside the cyclotron vault. Main reactions and nuclear data (Birattari et al., 1986).

Nuclear reaction	$T_{1/2}$	Activation energy (MeV)	Decay constant λ (h^{-1})	Microscopic cross section σ (mbarn)	Macroscopic cross section μ (m^{-1})
^{14}N (n, 2n) ^{13}N	9.96 min	11.3	4.176	10	3.90×10^{-5}
^{16}O (n, p) ^{16}N	7.13 s	10.2	350	40	4.19×10^{-5}
^{40}Ar (n, α) ^{37}S	5 min	2.6	8.318	10	2.33×10^{-7}
^{40}Ar (n, p) ^{40}Cl	1.35 min	6.9	30.81	16	3.73×10^{-7}
^{40}Ar (n, γ) ^{41}Ar	1.83 h	Thermal n	0.3788	630	1.61×10^{-7}