Analysis of air discharges from a radiopharmaceutical production center based on a cyclotron

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Abstract. The control of air contamination in a Nuclear Medicine Center (NMC) provided with a cyclotron for the production of radiopharmaceuticals is based on an automatic systems for air sampling and measurements. An important control for the assessment of dose to the population is aimed at the measurement of air concentration of stack release and inside the cyclotron vault during irradiation.

The frequency of sampling can be setup as continuous with respect to the stack effluent release and cyclic for other work environments. The 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 allow us to have very numerous air concentration data and a software for the analysis is needed. In this work is presented the analysis of the main data recorded in "San Gaetano" NMC at Bagheria (Italy) and are highlighted some anomalous events that have been happened some years ago.

An evaluation with a common Gaussian Plume air dispersion modelling code allow us to verify the no-radiological significance of the stack effluent releases in terms of potential dose to population reference group, sited more than 100 m away from the plant. A proposal for optimization of procedures, with the provision of air compressing stations (ACS) for both hot cells and cyclotron vault releases besides a change in the position and height of the stack, is also described.

KEYWORDS: Medical cyclotron, PET, radioactive air effluent.

1. Introduction

The use of radioisotopes in nuclear medicine for diagnostic techniques, with particular reference to Positron Emission Tomography (PET), is taking over the years increasing attention. There are many radioactive isotopes used, and many of their applications, as is now well-known physical and biological principles that govern their use in medicine. The use of the cyclotron can produce a large activity of short-lived positron-emitting radioisotopes for in-house diagnostic uses and to provide radiopharmaceutical products to other centers. Over the years systems of protection for workers and the environment have all been improved. One of these systems regards the monitoring of the air coming from various environments, and of what is released into the atmosphere, to be assumed as basis of evaluations of potential dose for the population living around the plant. In the case of target malfunction or failure, a large activity release can occur as well as a significant fraction of volatile radioactive compound can be released during chemical syntesis of PET radiopaharmaceutical inside hot cells. Therefore, a suitable air monitoring system must be implemented to avoid or limit releases into the atmoshere. Controls can be made on cyclotron vault exhaust air during and after the irradiation, on exhaust air from the PET radiochemical hot cells, on air effluent from radioactive waste storage or on stack air release.

As a reference "San Gaetano" NMC located at Bagheria, a small town near Palermo, Italy, was considered. At present, ^{18}FDG (fluoro-deoxyglucose) is the PET radiopharmaceutical most used and the frequency of sampling is set –up as continuous with respect to the stack effluent release and cyclic for other work environments (e.g. 1 minute every 8 minutes). The gamma-ray spectrometric measurement are made on-line and for a short time (1 minute) by using a Marinelli beaker filled with sampled air and a shielded NaI(Tl) 2" x 2" scintillation detector. An automatic analysis based on counting values in specific ROIs (one Region Of Interest for the photopeak, two ROIs for background) allow to determine a concentration value to be compared with suitable limit values. The concentration values are very numerous and a software analysis is needed. The analysis of the main

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data has been highlighted some anomalous events happened some years ago. The aim of this work is to examine the operative conditions of the air monitoring system so to ensure the respect of a potential dose value to the population living near the facility. As a result, it was formulated a proposal for optimization of plant operations, with the provision of air compressing systems (ACS) to contain both hot cells and cyclotron vault release air volumes besides a change in the position and height of the stack. These conclusions, while referring to a specific plant, should be extended to all cyclotron plants worldwide in operation.

2. Sources of air contamination

Main air contamination sources can be identified in a possible leakage of ^{18}F inside the cyclotron vault, activation of air inside the shielding bunker and emissions from the hot cells during or after the radiopharmaceutical synthesis. Accidents, such as a fire or a target break must be taken into account also for the computation of potential dose to population. The methods used to evaluate the last value are taken from NCRP 123 Report [1], taking into account only the contribution due to inhalation because of orders of magnitude more significant than direct irradiation from the cloud and submersion. Since these evaluations are no valid for short-term or unintentional releases, HOTSPOT 2.07.2 program [2] was used to obtain evaluations correlated to accidental releases following fire and target break events. A distance conservatively low between the stack output and any individuals of the population, equal to 100 m, was assumed keeping in mind also a real configuration of a PET center usually surrounded by gardens and parking. This assumption will be verified later.

A first evaluation regards the concentration value at the point of release able to do not exceed a potential dose constraint value of 10 μSv y^{-1} (D_p) for an individual of the reference group. A ground-level centerline Gaussian plume dispersion transport model proposed in NCRP 123[1],with a conservatively high value of the diffusion factor P, equal to 10^{-3}, was used. In this way, a concentration limit value (C_L) at the outlet of the stack of the order of 20 Bq l^{-1} was determined with reference to ^{18}F. Therefore it is sufficient to check that air concentration values at output of the 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. This is the aim of the air monitoring system while the goal of this work can be identified in achieving, following ALARA optimization principle, a more consistent reduction of the air concentration values in releases from the stack.

2.1 Fire and target break events

Fire and target break are considered as reference accidents for the planning of air containment system in order to limit the potential dose (D_p) to reference group of population.

For the fire event dose is considered dependent only on the distance *d* from the point of the release. The conical-type plume has a diameter at the point of measurement equal to 0.4*d* while the volume is equal to 0.00419 d^3. The plume moves only in one direction and all the activity is ejected in one hour, equal to the residence time of the individual. The concentration in the plume is assumed uniform as well as the whole activity of produced ^{18}F (185 GBq, 5 Ci) is considered vaporized, mixing with the air inside the bunker and suddenly ejected. In Table 1 are reported the committed dose value computed with these assumptions as a function of distance from the fire point. A data examination confirms that, to achieve the objective mentioned above ($D_p < 10$ μSv), population do not occupy any placement within a radius of 100 m from the plant, regardless of the height of release.

As regards target break accident, keeping in mind the blocking of ventilation during irradiation, it must evaluate the effects of a release through the stack. For this purpose HOTSPOT code, version 2.07.2 [2], was used. Results in terms of Total Effective Dose Equivalent (TEDE) are summarized in Figs. 1 and 2 (related to release height H equal to 10 and 20 m). Maximum dose values at 100 m distance are lower than the value assumed as dose constraint, independently from meteorological conditions (Pasquill categories A,..,F).

Table 1: Committed effective dose due to a release of 185 GBq of ^{18}F.

Age (population group)	Distance (m)						
	10	*20*	*30*	*50*	*100*	*120*	*150*
	Committed effective dose (μSv)						
>17	868	217	96.5	34.7	8.68	6.03	3.86
<1	1,030	258	114	41.2	10.3	7.15	4.58

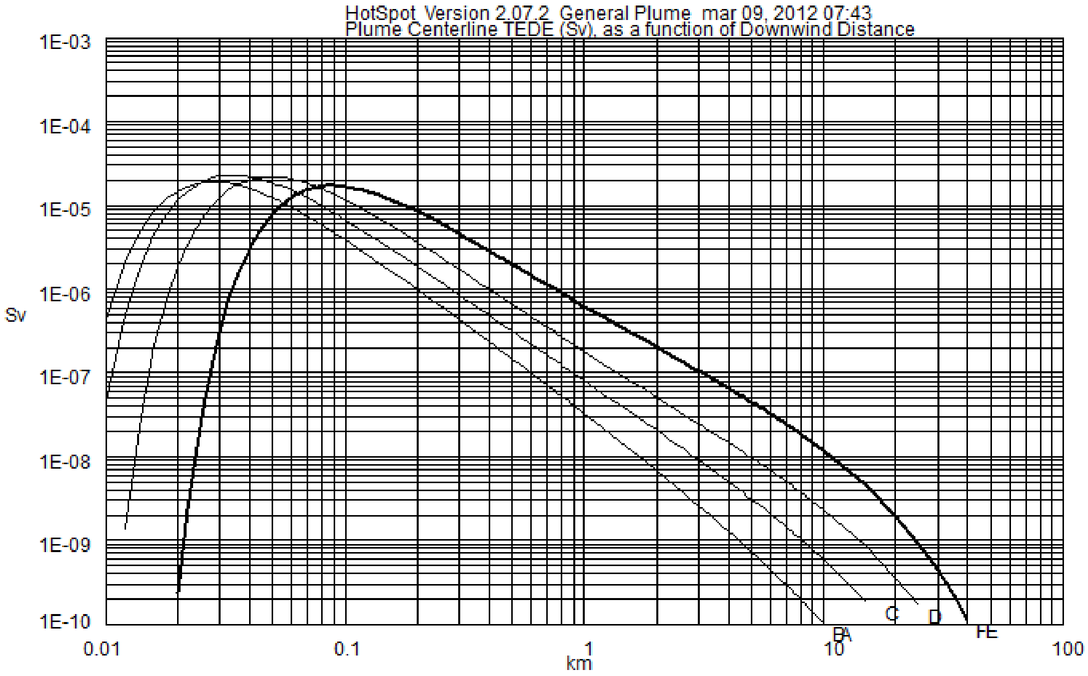


Figure 1: Plume centerline TEDE vs downwind distance from the release point. Stack height H=10 m.

2.2 Air activation inside the bunker

The activation of air inside the cyclotron vault is due to the neutrons produced by charged particles nuclear interactions with the target. In order to estimate the level of induced activity in the air the reactions reported in Table 2 can be considered [3]. The model for the evaluation of the dose to the population due to the activated air discharged outside assumes that, after the release, the radioactive gas is dispersed in the air within a plume along the direction in which the wind blows and that the receiving point is always at the center of the plume. The contributions of the individual dose to the population (reference group) is related to the following exposure modes: submersion in the cloud (gamma and beta radiation), and inhalation of radioactive gases.

To evaluate this last contribution can be conservatively assumed that ventilation should begin immediately after the end of irradiation, using the calculation of the equation.

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

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 reciprocations of air 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 volume depends on the neutron fluence rate Φ according to

$$C_S = \frac{\mu \, \Phi \, \lambda}{(\lambda + m_{on})} \tag{2}$$

and

$$\mu = \frac{\sigma \rho \ f \ N_A}{P_A} \tag{3}$$

with σ microscopic cross section for the specific reaction, ρ is the target density, f isotopic abundance, NA Avogadro number, P_A target atomic mass. Main parameters for the considered nuclear reactions are reported in Table 2.

In the case of the IBA (Ion beam Application) 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 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}. So the saturation activity for the reaction $^{40}Ar(n,\gamma)^{41}Ar$ is found to be $4.83 \cdot 10^5$ Bq m^{-3} and the induced activity after 2 hour of irradiation is about $1.5 \cdot 10^7$ Bq. This value is less than the activity first considered for the fire and can not originate a potential dose value exceeding specified limit. However, as pointed in [4], it can be considered mandatory to close the ventilation of cyclotron vault for a selected time period after the irradiation.

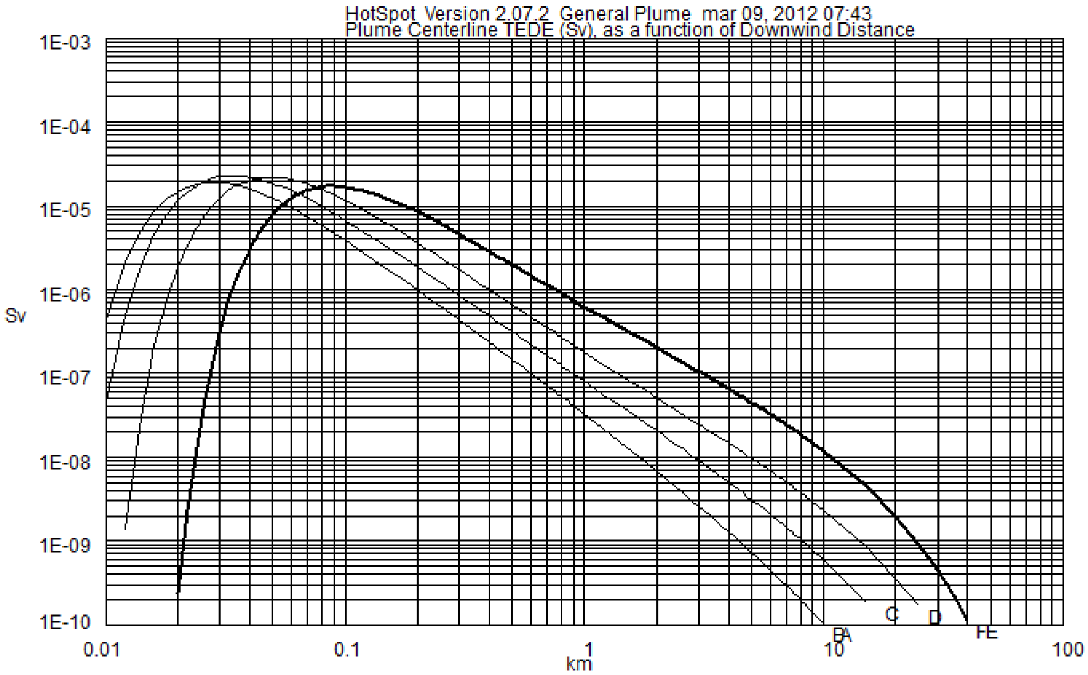


Figure 2: Plume centerline TEDE vs downwind distance from the release point. Stack height H=20 m.

Table 2: Neutron activation of air inside the cyclotron vault. Main reactions and nuclear data.

uclear reaction	T ½	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	3.90E-05
^{16}O (n, p) ^{16}N	7.13s	10.2	350	40	4.19E-05
^{40}Ar (n, α) ^{37}S	5 min	2.6	8.318	10	2.33E-07
^{40}Ar (n, p) ^{40}Cl	1.35 min	6.9	30.81	16	3.73E-07
^{40}Ar (n, γ) ^{41}Ar	1.83 h	thermal n	0.3788	630	1.61E-05

3. Air Monitoring System

Figure 3 shows the schematic diagram of the real-time stack effluent monitoring system at the “San Gaetano” NMC radionuclide production facility. It includes two independent systems, each provided with a NaI(Tl) 2”×2” 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. 4). Gamma-ray spectrometric measurement are made on-line and for a short time (1 minute). The automatic analysis operates in ROI mode set at 511 keV photopeak produced by positron emitters while two ROIs are selected for background. The evaluation of net 511 keV-counting allow to determine a concentration value to be compared with suitable alarm thresholds or limit values. A Personal Computer (PC) oversees all operations and generates automatic alarms by means of a programmable PLC logic.

A first system allows the continuous monitoring of the effluent released to the stack in order to have real-time information on the quantities released into the atmosphere and finally to put an alarm in case of air concentration values higher than C_L limit.

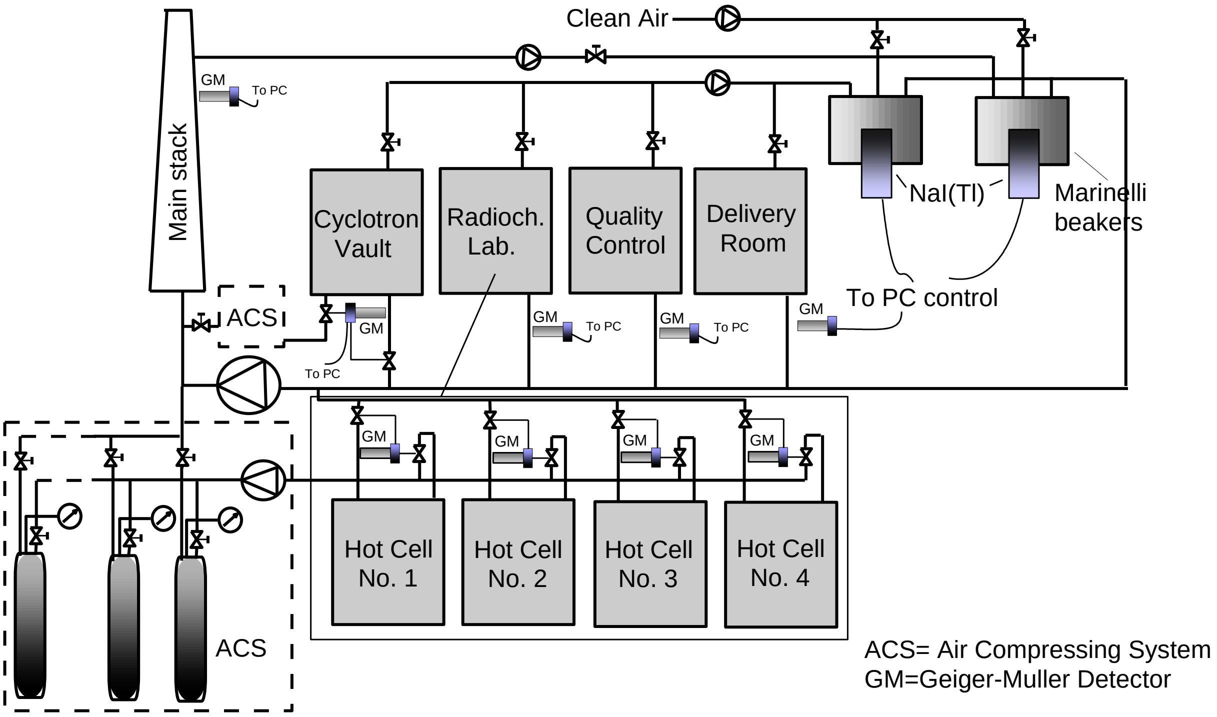


Figure 3: Schematic diagram of the real-time stack effluent monitoring system of the “San Gaetano” NMC. Valves and compressing systems as well as detector response are controlled with a PLC unit connected to a PC. Inside dashed lines the possible improvements of the system.

A second system, completely analogous to the previous one, is connected to 4 different environments:

1. Cyclotron vault;
2. Radiochemistry laboratory;
3. Quality Control Laboratory;
4. Delivery Laboratory.

The environment from which to draw the air is automatically selected and through a pump and appropriate valves the air shall circulate in the counting chamber. The system has the following operation: the air drawn in a continuous manner by the pump, passes through a filter which removes particulates, enters the sampling volume and, finally, is pushed towards the exit. The sequence of sampling and opening time of the valves are fixed. At this stage, a time of 1 minute is selected for sampling and measurement for each environment with a sequence of the type 1_2_3_4_1_2_3_4....

After a measurement, a 1 minute time period to clean the Marinelli beaker with air from a container is provided. So, for each laboratory, a 1-minute measurement every 8 minutes was performed.
The use of the systems above mentioned allow us to have almost 180 values per day for each environment while for the outlet of stack the data are about 720 per day.

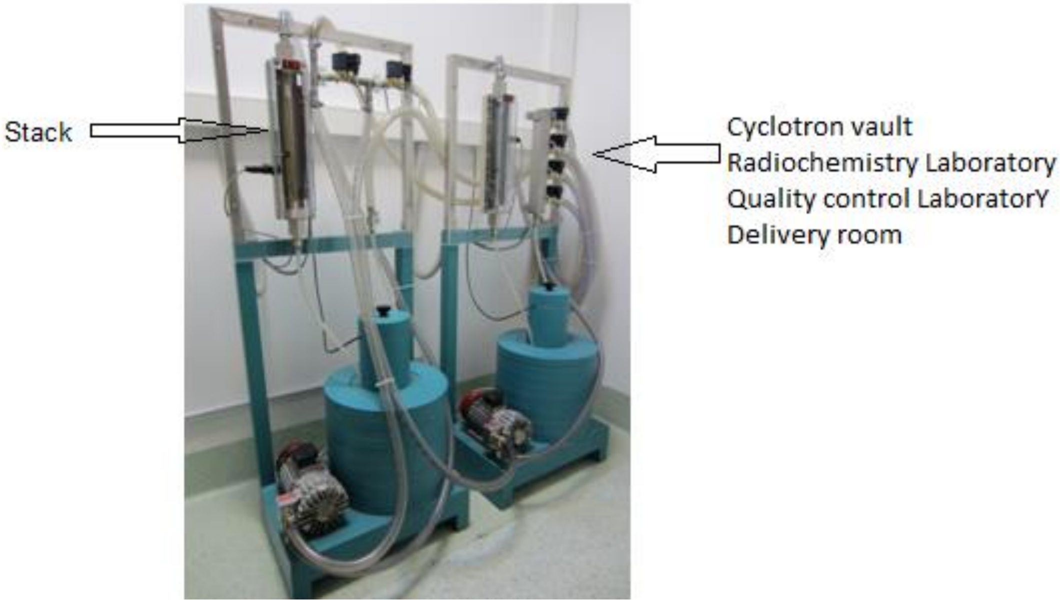
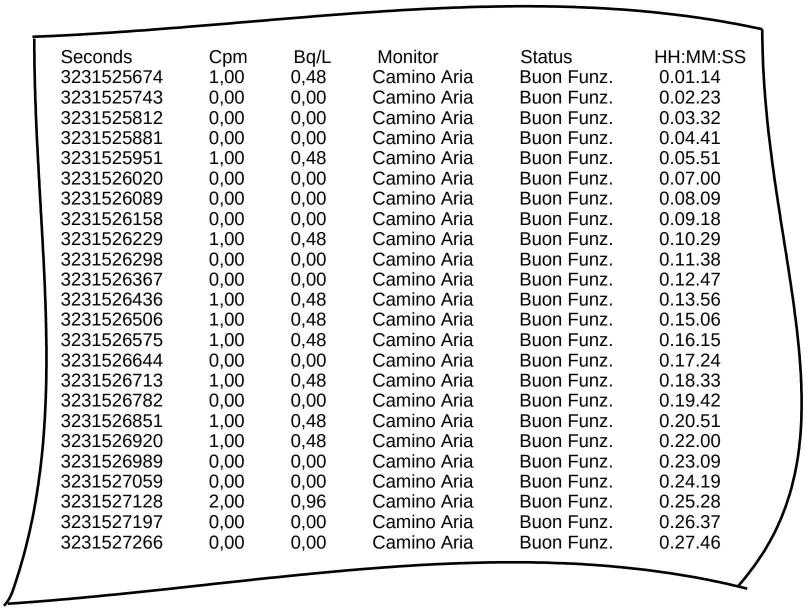


Figure 4: Air monitoring measurement station provided with two shielded Marinelli beakers and NaI(Tl) 2”×2” scintillators. The first (at left) refers to a connection with air passing in the stack (in continuous); the second (at right) is cyclically connected to one of the monitored environments.

4. Results and discussion

In Fig. 5 is reported an example of a typical file containing monitoring data. The counting values for a 1 min counting time are very low, referring to a stand-by time period. Counting data for each day are very numerous and a specific software analysis is needed. An home made software, written in Microsoft VISUAL BASIC language and operating in a Microsoft Excel (Office 2007) environment, allows to determine maximum or minimum values and performs the calculation of daily average concentrations. The latter values are stored in a database and ordered with reference to months and years.
A typical trend of air concentration values is reported in Figure 6. It can be noted an increase in values of air concentrations in the cyclotron vault, which is considered a normal event because of air activation inside the bunker, followed by an increase of the values of stack outlet. The two events can be directly related, such as for example in Figs. 6 and 7, but may also result from other events, as in Figs 6,8. In the latter, the high values of the stack output air concentration are undoubtedly related to a release from hot cells since it occurs a long time after the end of irradiation and are related to synthesis operations. The air of the other rooms are not affected, as shown in Figs. 7,8. Even with these anomalous events, air concentration values are lower than 20 Bq l^{-1}, as shown by the trend of average concentrations reported in Fig. 9. However, since it is easy to further reduce air concentration values with suitable containment devices, it can be proposed the adoption of ACS systems for both hot cells and cyclotron vault air confinement. Such devices are already adopted in other PET facilities provided of a cyclotron, with obviously good results. The adoption of ACS systems is a better solution with respect the adoption of a delay discharge line, which allows to enter the contaminated air in the environment after a given period of time so to reduce the stack air concentration. The operation of the ACS can be routinely performed, with storage of the air coming from the cyclotron vault or hot cells for one day and release to the atmosphere the next day, before a

new irradiation. Another solution could be represented by a use of the ACS with automatic start signal given by a detector (e.g. a Geiger-Muller, GM) positioned on the extracting line or by an alarm signal from the control PC (or by a PLC, Programmable Logic Control) of the monitoring system.



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Seconds         Cpm     Bq/L    Monitor         Status          HH:MM:SS
3231525674      1,00    0,48    Camino Aria     Buon Funz.      0.01.14
3231525743      0,00    0,00    Camino Aria     Buon Funz.      0.02.23
3231525812      0,00    0,00    Camino Aria     Buon Funz.      0.03.32
3231525881      0,00    0,00    Camino Aria     Buon Funz.      0.04.41
3231525951      1,00    0,48    Camino Aria     Buon Funz.      0.05.51
3231526020      0,00    0,00    Camino Aria     Buon Funz.      0.07.00
3231526089      0,00    0,00    Camino Aria     Buon Funz.      0.08.09
3231526158      0,00    0,00    Camino Aria     Buon Funz.      0.09.18
3231526229      1,00    0,48    Camino Aria     Buon Funz.      0.10.29
3231526298      0,00    0,00    Camino Aria     Buon Funz.      0.11.38
3231526367      0,00    0,00    Camino Aria     Buon Funz.      0.12.47
3231526436      1,00    0,48    Camino Aria     Buon Funz.      0.13.56
3231526506      1,00    0,48    Camino Aria     Buon Funz.      0.15.06
3231526575      1,00    0,48    Camino Aria     Buon Funz.      0.16.15
3231526644      0,00    0,00    Camino Aria     Buon Funz.      0.17.24
3231526713      1,00    0,48    Camino Aria     Buon Funz.      0.18.33
3231526782      0,00    0,00    Camino Aria     Buon Funz.      0.19.42
3231526851      1,00    0,48    Camino Aria     Buon Funz.      0.20.51
3231526920      1,00    0,48    Camino Aria     Buon Funz.      0.22.00
3231526989      0,00    0,00    Camino Aria     Buon Funz.      0.23.09
3231527059      0,00    0,00    Camino Aria     Buon Funz.      0.24.19
3231527128      2,00    0,96    Camino Aria     Buon Funz.      0.25.28
3231527197      0,00    0,00    Camino Aria     Buon Funz.      0.26.37
3231527266      0,00    0,00    Camino Aria     Buon Funz.      0.27.46
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Figure 5: Example of a typical file containing monitoring data for the outlet of the stack.

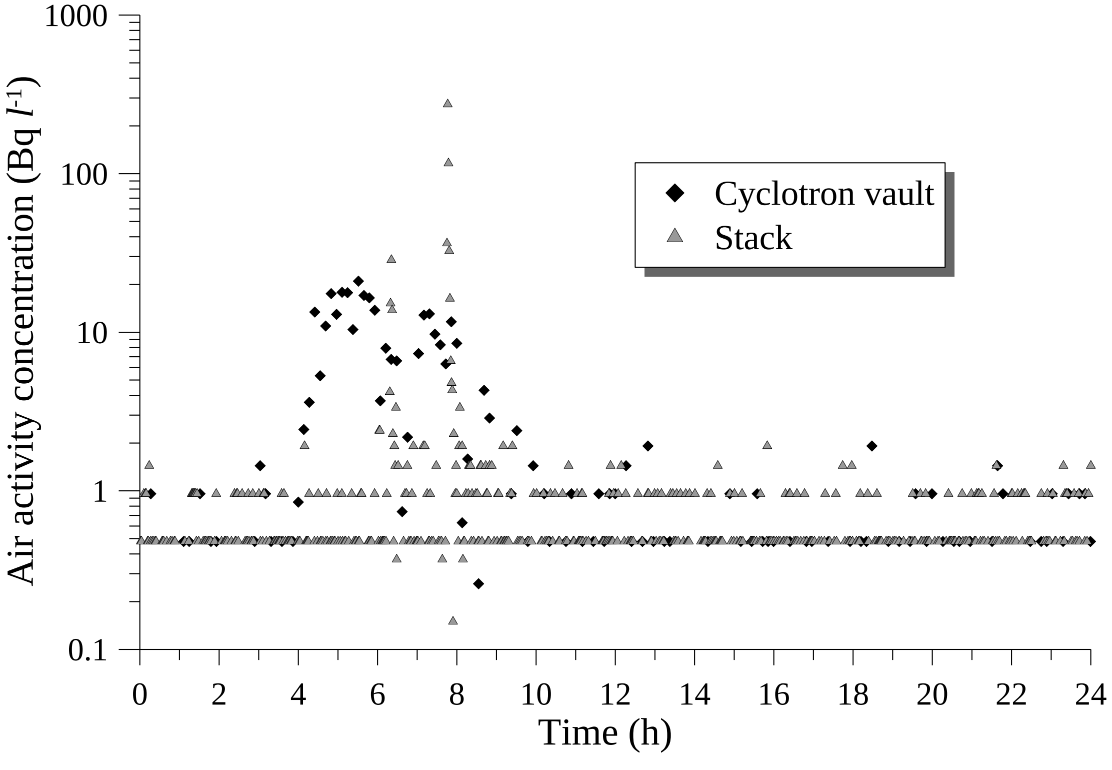

Figure 6: Daily trend of air concentration values. An increase of concentration values of cyclotron vault exhaust air during and immediately after the irradiation and higher values of stack effluent concentration are highlighted.

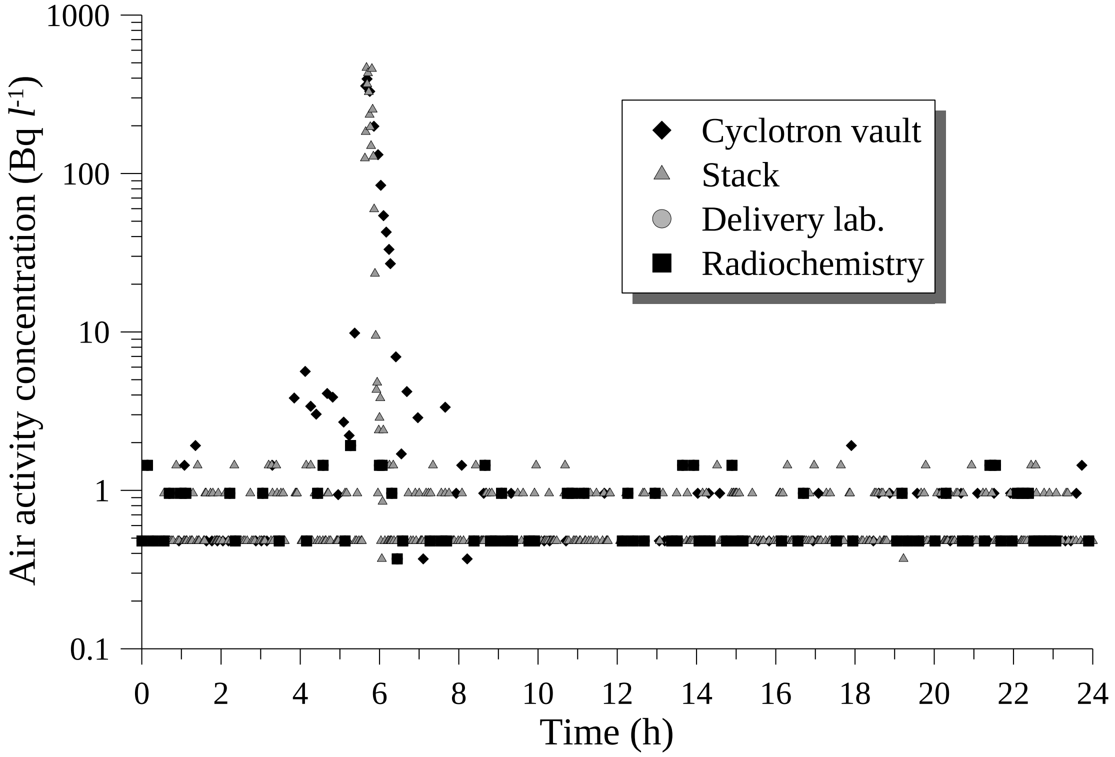

Figure 7: Example of daily trend of air concentration values.

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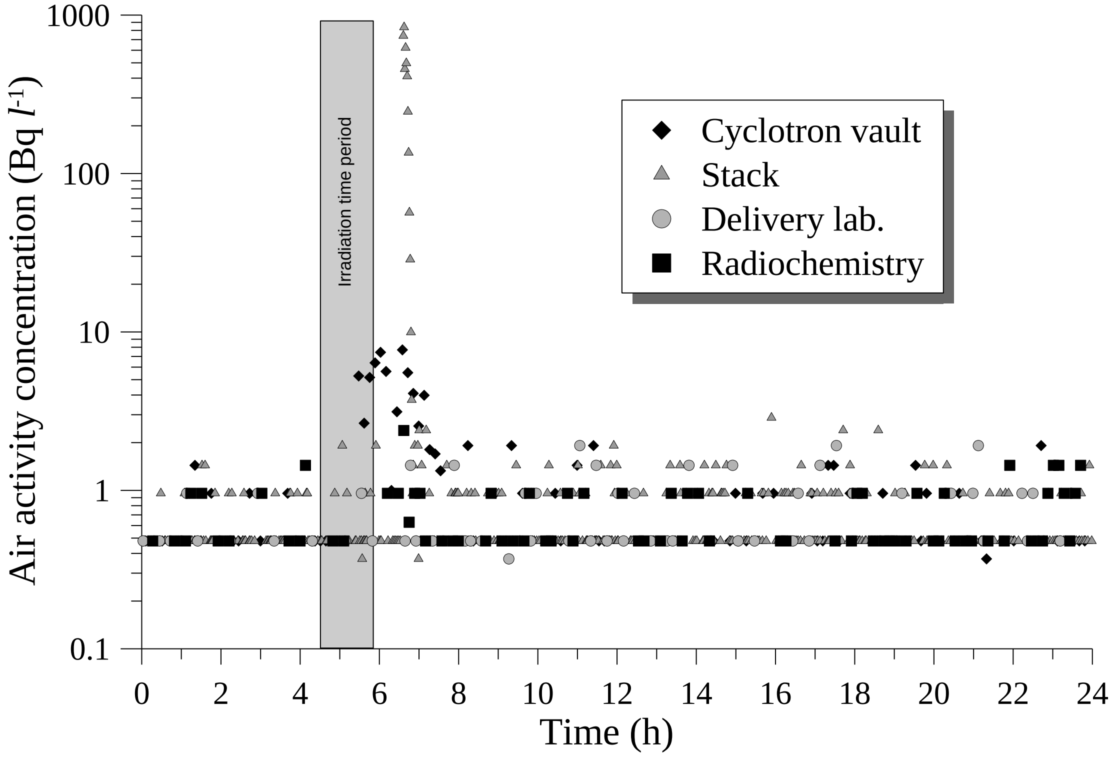

Figure 8: Time dependence of air activity concentration values during a day. A release from the hot cells during a synthesis procedure is highlighted.

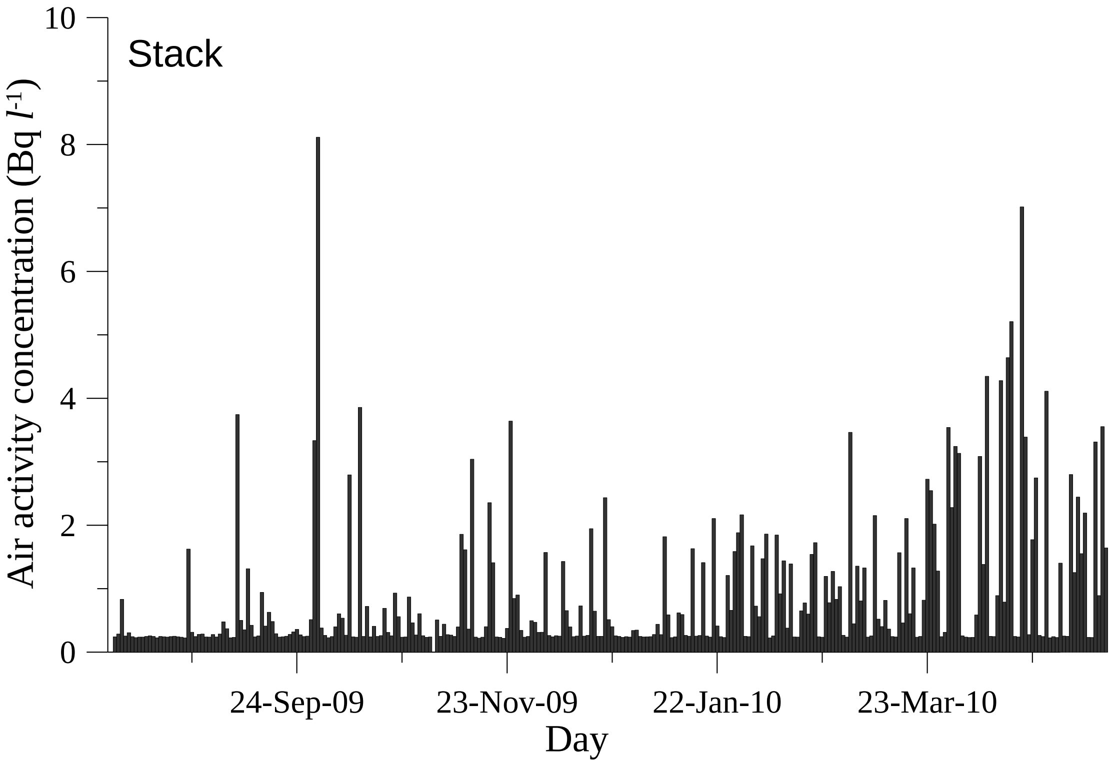

Figure 9: Daily average air concentration values of stack release for a time period of about 10 months.

5. Conclusions

The analysis of the air concentration data allowed us to obtain the behaviour of activity concentration values for stack air releases, without finding average values that lead to exceed the dose constraint of 10 μSv y^{-1} for inhalation of the population. However, some anomalous events were noted in the daily trends of concentration values. Most of the critical values are probably due to defects in mounting of target components or a no sufficient containment of synthesis modules used within the Hot Cells, the latter causing a ^{13}N release during the synthesis reaction.

The adoption of an ACS for the discharge of the hot cells exhaust air and, ultimately, for the cyclotron vault air, may reduce the occurrence of these events and/or mitigate their effects. Therefore, possible improvements on air monitoring system can be:

- A link with ACS of hot cells air releases. The use of ACS can be automatically controlled by PLC after an overload signal from GM placed on the outlet duct.
- A second ACS connected with cyclotron vault to contain activated air produced during irradiation and, as shown in [4] for a few hours after the end of irradiation.
- Alternatively, a delay pipeline (long enough to delay the release of contaminated air from the stack of at least 4-6 h).
- Besides the above items, the forecast of increasing the height of the stack and place it at greater distances from the first settlement outside the plant.

The adoption of proposed devices could reduce the concentration values during the operations even 1000 times, and therefore dose to the population can become indistinguishable from what is due to the environmental background.

Acknowledgements

The author wish to thank heads of “San Gaetano” NMC and, in particular, Dr. Daniele GRECO to have provided some air concentration data and useful information.

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