The Efficiency for Gas Capture Systems for PET Cyclotrons and Hot Cells.

Modern cyclotrons can produce large amounts (>500 GBq) of PET isotopes (usually F-18) per production run for radiosynthesis of various radiopharmaceuticals, mainly 2-[18F]-fluoro-2-deoxy-D-glucose (FDG). Operating cyclotrons inherently come with radiation protection issues

- neutron activated of the bunker,

- cyclotron targets, and

- consumables such as extraction and target foils, air and gaseous emissions, particularly during delivery of product from the cyclotron and during radiosynthesis.

- A stack to vent gases is a feature of a cyclotron and radiopharmaceutical facility as is radiation equipment to monitor stack emissions.

State	Hospital	Commercial	Research	Total
QLD	1	1	1	3
NSW	2	2	1	5
VIC	1	1		2
WA	1			1
SA	1			1
TAS				0
NT				0
ACT				0
			Total	12

Hot Cells.

Two decades ago cyclotrons had much lower yields (<20 GBq) and it was found that F-18 effluent releases air discharges ranged from 0 to 2.56 GBq per production run in the USA.

- There are various international references available, particularly from the IAEA on the guidelines and technical requirements for establishing and operating a cyclotron, along with a FDG production facility.

- Radiosynthesis of radiolabelled compounds is performed in hot-cells with significant thicknesses of lead shielding (up to 8 cm). Commercially available hot cells are designed to be airtight with inflatable door gaskets. The cell air pressure is depressed (>-30 Pa) both to prevent any gaseous activity release entering the workplace and for cell microbiology control.
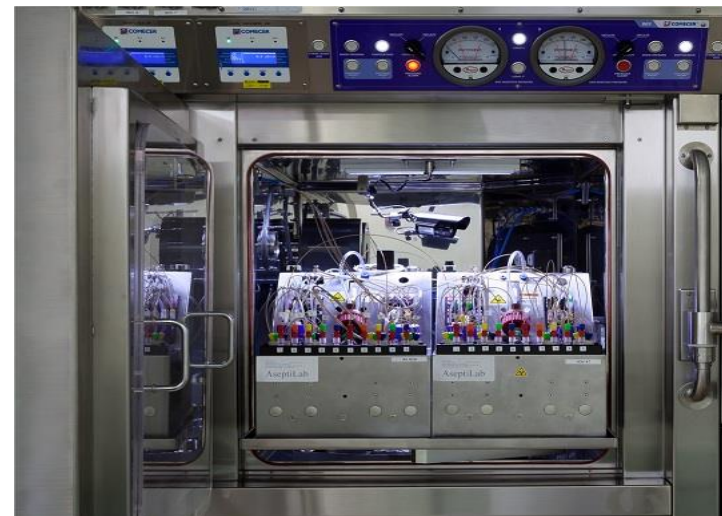
Exhaust Gases

Hot Cells – Exhaust Gases

The techniques applied to manage gaseous emissions from hot cells include in-line activated carbon filters, delay lines or collection tanks. On some newer hot cells, a radiation detector (GM type) in the air outlet will be linked to a control which opens and closes a baffle to prevent gases from further released from the hot-cell. The baffle is not reopened until the gas radioactivity falls to an acceptable level pre-set by the manufacturer or configured by the cyclotron operator. These radioactive gas trapping systems vary between hot cells but often significant releases have been known to occur before these engineering controls are activated.

Treatment of Exhaust Gases

The in-line activated carbon filter design is a one-pass exhaust system that allows for a combination of HEPA filters and activated charcoal gas capture banks. The charcoal used is nuclear grade carbon that meets ASTM standards and is very efficient at capturing radioactive iodine, fission products and gas such as radon. However, as with the carbon filters in hot cells, there is a real concern that with some positron-emitting gases the effectiveness of such filters are suspect. Depending on the type of activated carbon, its condition and the type of positron-emitting gases produced, such filters have been shown to be sometimes wanting in their effectiveness in trapping these gases

# Gas Capture Systems for PET Cyclotrons and Hot Cells.

A more elegant engineering solution is to capture of exhaust gas using a gas compression system that collect all the gases from the hot-cells during synthesis and stores them in a series of gas cylinders for later release. A typical compressed gas system can uses 2 groups of 4 gas bottles of 50 litres each that can collect 40,000 litres of waste gas from the hot-cells. Gaseous releases from a cyclotron itself are infrequent and generally not problematic. With a rupture of a target foil most of the gas ends up in the chamber of the cyclotron, thus contaminating the high vacuum system. Neutron activation of natural argon (Ar-40, with a natural abundance of 0.94% in the atmosphere) in the cyclotron vault occurs but at very low concentrations.

Radiation Monitor Stack Discharges

Two different setups in the stack are possible, one with a pump to sample effluent for bypass measurements.

The other (most common) with radiation detectors in the stack. Mounting on the stack walls requires charged particle detection using large area proportional counters where positron radiations provide localised and low background sensitivity. "In stack" flow methods measure the positron annihilation radiation (511 keV photons) with gas filled Geiger Muller and NaI scintillation detectors, to provide sensitivity.
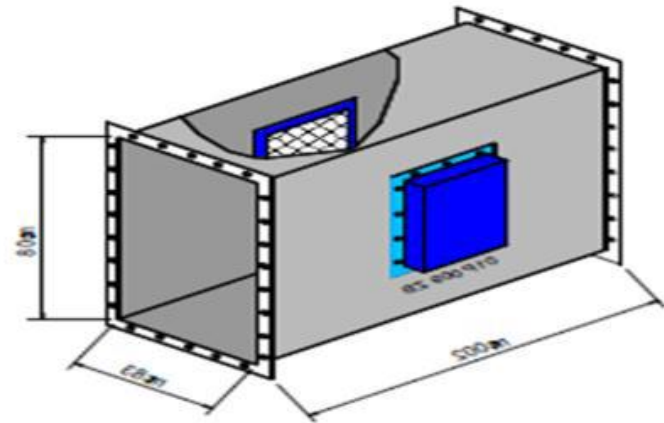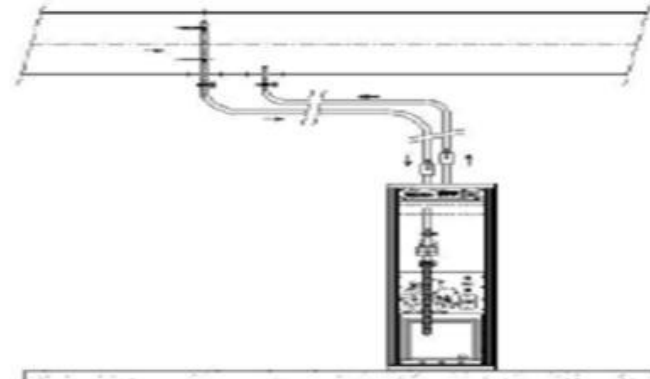
Radiation Monitor Stack Discharges

Coincidence detectors of 511keV radiations potentially offer an improved level of sensitivity and are now becoming more widely available. Until recently the calibration of the stack monitoring detectors required the release of radioactive gas. Eberline market (FHT53511) a coincidence unit with signal-to-background ratio 1 to 2 orders of magnitude better than non-coincidence units with a detection limit below 1 $kBq.m^{-3}$. The manufacturers report that the system is self-calibrating using a point Ge-68 source on the outer surface of the stack.
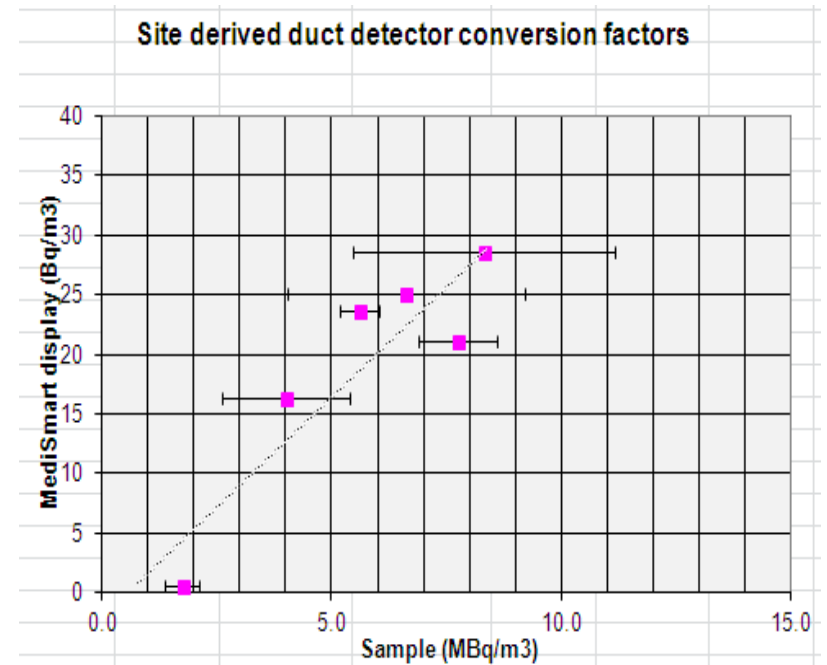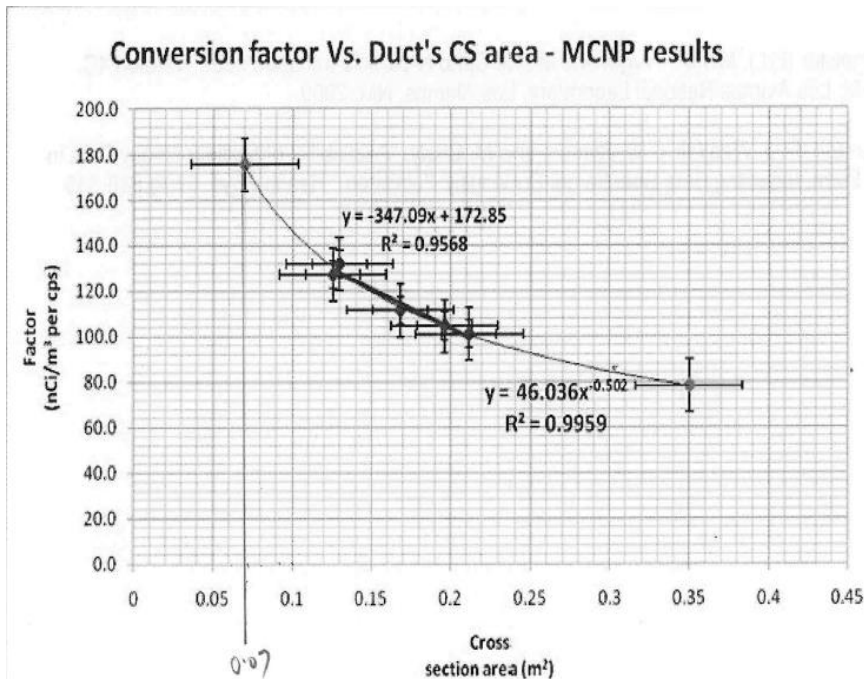
PET Waste Gas Release Standards

Internationally the positron-emitting (PE) gas release levels from these facilities vary dramatically as shown in Table below.

Country	Isotope	Release to air discharge limit Bq.m⁻³	Derived limit GBq				Investigation GBq		
			Daily	Month	Quarter	Annual	Month	Quarter	Annual
Scotland	PE		74			500			
ARPANSA (NSW)[1]	F-18 & C-11			250	625	1250	50	125	250
	Note: Based on annual member of public limit to be less than 10 μSv								
USA[28]	F-18	3700							
	N-13	640							
Italy[8]	PE	129							
	Note: Annual member of public limit to be less than 10 μSv								
QLD	F-18	320							
	Note: From Schedule 3 Queensland Radiation Safety Regulation 2010								

Radiation Monitoring of Stack Gases

The **MediSmarts** stack monitoring manufactured by Rotem Industries Ltd is a commonly installed system in Australia and allows real time monitoring and logging of data. The stack monitoring system uses a low range NaI detector that records in counts per second (cps) and a high range Geiger Muller detector that records in µSv/hr. Both detectors can be calibrated in software using known releases of C-11 in the form of CO_2 gas to record Bq m^{-3} with a calibrated flow sensor.

Conversion factor Vs. Duct's CS area - MCNP results

$y = -347.09x + 172.85$
$R^2 = 0.9568$

$y = 46.036x^{-0.502}$
$R^2 = 0.9959$

Site derived duct detector conversion factors

Radiation Monitoring of Stack Gases

As an example of a stack monitoring system, MediSmarts provides the following:

- Quantitative activity release measurements.
- Automatic activity release reports and cumulative release reports.
- Integrated on-line air flow data for activity release report.
- Measuring wide range concentrations concentration levels from 1 MBq m^{-3} to 150 MBqm^{-3}
- Software calibration routine for activity released in ducts that converts raw data in cps from NaI and GM detector into activity concentration data in Bq m-3 or pCi m^{-3} .
- Local alarms and relay output for triggering switches for engineering control.

An example of a recurrent production opposite This shows between the period 21 to 26 /11/2011 a total of 155 MBq (Ave168Bqm^{-3}).
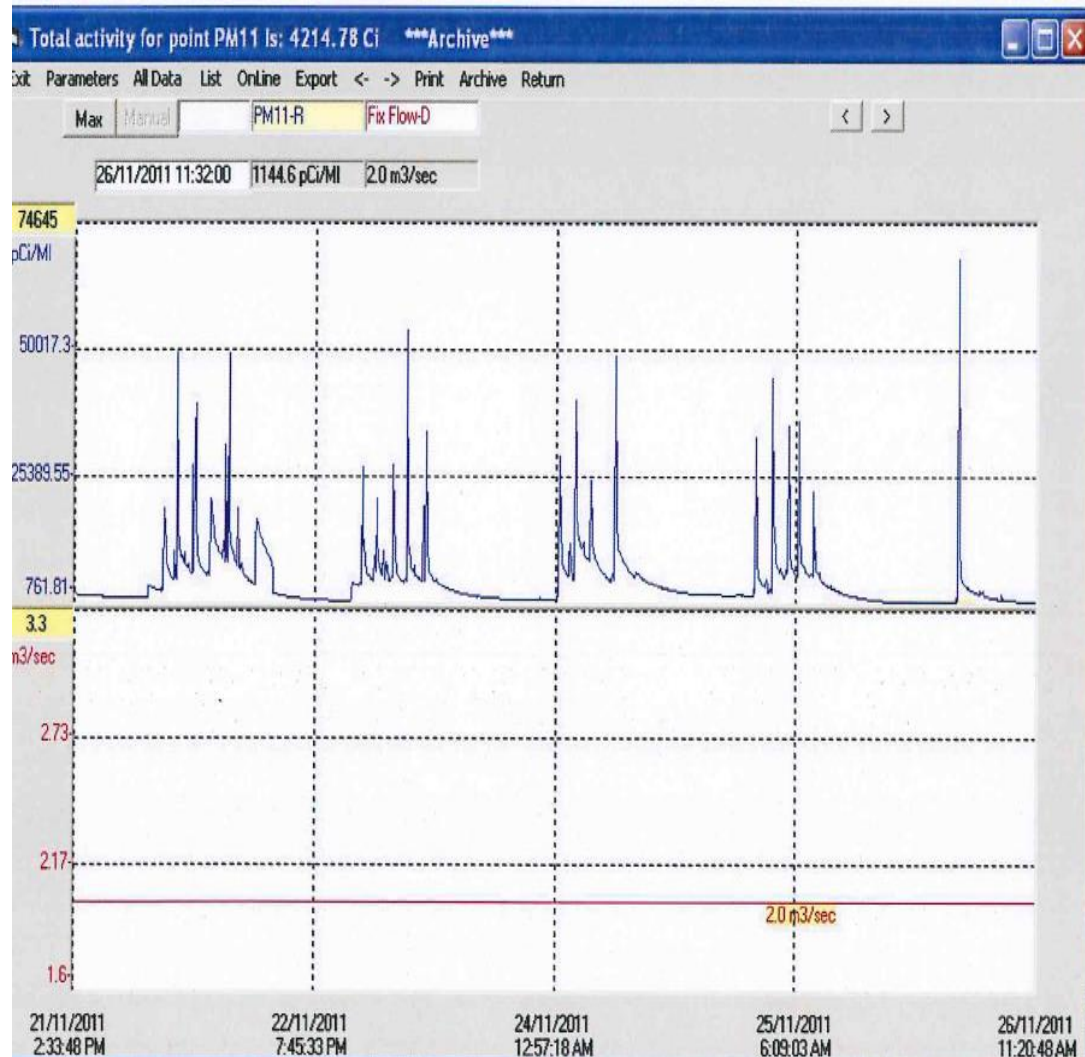
Radiation Monitoring of Stack Gases

Radiation measurements show that gaseous compounds that plate-out and contaminate the inside of exhaust ducting give rise local contamination near radiation detectors. This plate-out in the stack makes it more difficult for operators to assess release and cumulative activity compliance from stack monitoring systems and as most systems integrate the total release on a daily basis this activity due to contamination is added to the releases. Therefore, these systems overestimate the daily releases by about 5 to 10% depending on the positron-emitting gases released. This local contamination is more pronounced for F-18 gaseous releases compared to C-11 or N-13 releases as shown in Figures 6 & 7.

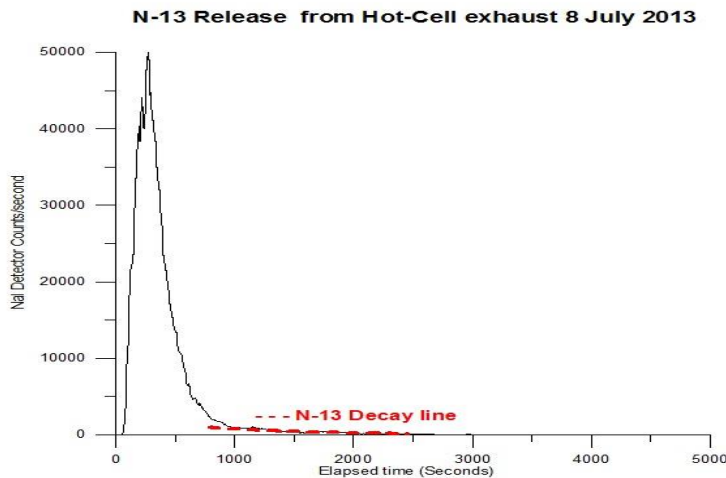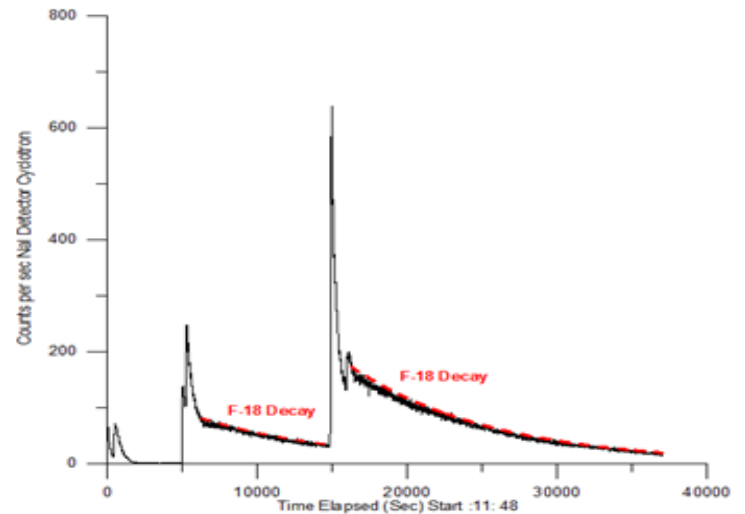**Figure 6 Stack plate o of N-13**
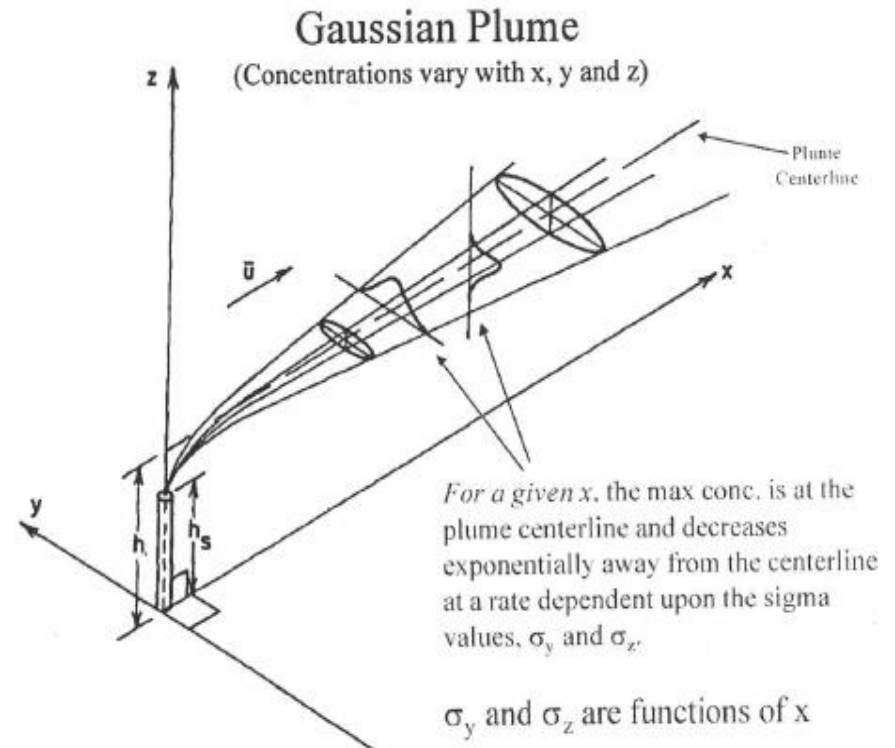
Figure 7 Stack plate out F-18

Common radioisotopes produced from liquid, gas and solid targets by cyclotrons

Radio-isotope	Nuclear Reaction	Target	Chemical Form	Physical Form	Half Life $T_{1/2}$
Fluoride-18	$^{18}O(p, n)^{18}F$	Liquid	$^{18}F^-$	Liquid	110 min
Nitrogen-13	$^{13}C(p, \alpha)^{13}N$	Liquid	$^{13}NH_3$	Liquid	10 min
Carbon-11	$^{14}N(p, \alpha)^{11}C$	Gas	$^{11}CO_2$	Gas	20 min
		Gas	$^{11}CH_4$	Gas	20 min
Oxygen-15	$^{15}N(p, n)^{15}O$	Gas	$^{15}O_2$	Gas	2 min
Copper-64	$^{64}Ni(p,n)^{64}Cu$	Metal plated	$CuCl_2$	Liquid	12.8 hrs
Iodine-124	$^{124}Te(p,n)^{124}I$	TeO_2 (oxide powder melted)	^{124}I as iodide in alkaline NaOH solution	Liquid	4.2 days

Dose Constraint Methodology in Setting Airborne Releases
Atmospheric Modelling

The most popular atmospheric model is the Gaussian plume dispersion model as shown below . In the absence of detailed local meteorology and detailed dispersion the application of Gaussian models are considered to be acceptable. This example shows the ICRP Publication 101 methodology for estimating dose constraint principles for the setting of release limits.

Gaussian Plume
(Concentrations vary with x, y and z)

For a given x, the max conc. is at the plume centerline and decreases exponentially away from the centerline at a rate dependent upon the sigma values, σ_y and σ_z.

σ_y and σ_z are functions of x

Atmospheric Modelling

The lateral spread of the plume or crosswind dispersion and vertical dispersion are dependent on the amount of turbulence in the atmosphere or atmospheric stability classification. The most widely used classification scheme was developed by Pasquill (1961) and subsequently modified by Turner (1967) as discussed in Hanna et.al[25]. The classification or categorization of the atmosphere into six turbulence categories (A,B,C,D,E,&F) was devised, as shown in Table below.

| Turbulence description	Pasquill category	Comment
Very unstable	A	Hot summer day with good vertical mixing generated by very warm ground heating and strong cross-wind turbulence.
Moderately unstable	B	
Slightly unstable	C	
Neutral	D	Calm conditions at sunset and sunrise. Air cools at the adiabatic rate of 1^oC per 100m for dry air.
Slightly stable	E	
Moderately unstable	F	Calm clear sky nights with ground radiative cooling.
Very Stable		Not applicable

Dose Conversion Factors

The current effluent release rates to atmosphere are based on the ICRP and are also reproduced by the US Nuclear Regulatory Commission. Table below summarises some of the dose conversion factors used in this radiation dose modelling. The Derived Air Concentrations (DAC's) for submersion are based on a 1 mSv annual limit while the Inhalation DAC's are based on a 0.5 mSv annual limit that allows a factor of 2 for younger age groups present in members of the public.

Positron-emitting gas	Exposure pathway	Dose conversion factor μSv^- per Bq m^{-3}	Comments
N-13	Submersion	1.35	10 min half-life
F-18	Inhalation	0.135	Reference only
C-11	Inhalation	0.015	Reference only
I-124	Inhalation	33.7	Reference only
O-15	Submersion	0.67	Reference only

Radiation Dose Modelling

The results of the radiation dose modelling for a daily release of 8 GBq of N-13 gas over a year will result in the whole body radiation dose of less than 1.2 µSv and well below the annual dose constraint of 300 µSv. The annual radiation dose of 1.2 µSv can be compared with the radiation dose (cosmic ray exposure) of 16 µSv received when travelling by plane from Darwin to Perth. This modelling uses a number of very conservative assumptions with a view to estimating the maximum credible radiation dose to an individual who is a member of the critical group.

Annual Radiation Dose for 8 GBq Daily N-13 Release
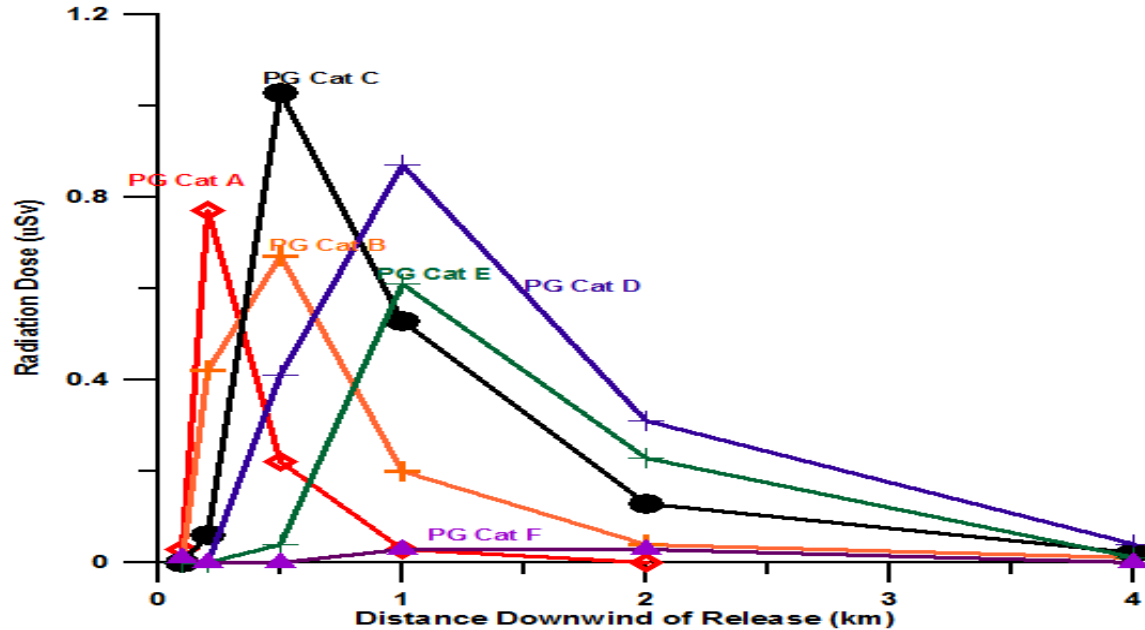
Limitations and Validation of Dose Modelling

There are significant limitations to modelling of plume dispersion due to building obstacles, stack configuration, topography and the micrometeorology of eddy and wind shear effects on mixing, etc. Results should be viewed as indicative and perhaps most useful in alerting a cyclotron operator to locations that require further investigation.

Conclusions – What have we learnt ?

 We propose that greater consideration of building ventilation must occur in order to incorporate the most effective design for gas containment for positron-emitting gases. Our evaluation highlights that no single system on its own will allow centres using multiple radioisotopes to meet the target of zero emissions during routine operations. For example, the charcoal filters provide an excellent mechanism to capture radioiodides and radiolabelled organic compounds but are less suitable for nitrogen-13 gases. Nitrogen-13 (typically formed as a by-product during the formation of F-18 fluoride) can be better contained by the use of gas capture bags which are connected to vents and exhaust associated with the synthesis modules.

Containment systems should form a critical part of the development of PET facilities and it is important to include a variety of experts (radiochemists, engineers, health physicist etc) in this process. New PET centres should work closely with the local environmental agencies and regulatory organisation to ensure the design is both efficient and practical.

While there are many problems with atmospheric modelling it's the only way to apply the dose constraint criteria and should be validated at least with external dosimetry measurement and air sampling where viable.