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Stack Monitoring at ANSTO and Comparison with International Guidelines

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EXECUTIVE SUMMARY

This report reviews the sampling systems at ANSTO in terms of current international guidelines for the design and operation of stack monitoring at nuclear facilities. A review of the literature and enquiries overseas have revealed the principal international guidelines for stack sampling and on-line monitoring are:

1. Monitoring of Radioactive Releases to Atmosphere from Nuclear Facilities, **Technical Guidance Note M11**, UK Environment Agency 1999. This Technical Guidance Note provides guidance on the monitoring of airborne releases from nuclear facilities regulated under the UK Radioactive Substances Act (**RSA 1993**). For sites in England and Wales, the authorisations are issued by the Environment Agency and in Scotland by the Scottish Environment Protection Agency.

2. Sampling and Monitoring Releases of Airborne Radioactive Substances from Stacks and Ducts of Nuclear Facilities. American National Standard **ANS/HPS N13.1-1999**.

A detailed review of the two guidelines has been conducted by Mr Fred May, an international expert in monitoring of radio-iodine. This is presented in Appendices I and II. As a result of this review, ANSTO plans to follow the UK M11 Technical Guidance Note because :

- It adopts the important philosophy of recognition that different sampling techniques are required for the different physical forms of radionuclides i.e. particulate, vapour or gas. Thus separate sampling systems and analytical methods may be required for the different radioactive species within the same effluent stream.
- It adopts methods for sampling of radio-iodine stack effluents that conform most closely to best practice recommended by Mr May. In particular it recommends that the distance between the effluent stream and sample collector is minimized to avoid plate-out and unrepresentative sampling.

In general, sampling at Lucas Heights complies with all the mandatory requirements of M11 and where it differs from the advice in the UK M11 standard, the techniques adopted are almost always more stringent and prudent than the techniques advocated in M11. An audit of all stack sampling at ANSTO facilities will allow any gaps in the current sampling practices to be identified and action plans for upgrades to be formulated.

CONTENTS

1 INTRODUCTION

Research Reactors and Medical Cyclotrons are nuclear devices primarily used for the production of various neutron-rich and neutron-deficient radioisotopes respectively. These radioisotopes are vital for a myriad of medical, health and industrial applications. Australian Nuclear Science and Technology Organisation (ANSTO) operates a 10 MW_{th} Research Reactor and a 30 MeV H^- ion Cyclotron for the production of various industrial and medical radioisotopes on a commercial basis. During routine operations, these facilities release small amounts of airborne radioactivity from stacks, which must be monitored to show regulatory compliance. It is also important to cover potential accidental releases of airborne radioactivity, which, while of low probability, could be of greater environmental impact.

This report reviews the sampling systems at ANSTO in terms of current international guidelines for the design and operation of the stack monitoring. The principal international guidelines for stack sampling and on-line monitoring are:

I. Monitoring of Radioactive Releases to Atmosphere from Nuclear Facilities, **Technical Guidance Note M11**, UK Environment Agency 1999. This Technical Guidance Note provides guidance on the monitoring of airborne releases from nuclear facilities regulated under the UK Radioactive Substances Act (**RSA 1993**). For sites in England and Wales, the authorisations are issued by the Environment Agency and in Scotland by the Scottish Environment Protection Agency.

II. Sampling and Monitoring Releases of Airborne Radioactive Substances from Stacks and Ducts of Nuclear Facilities. American National Standard **ANS/HPS N13.1-1999**.

2 BACKGROUND

The stack discharges may be vapour, gas, particulate or suspended droplets and all present their own individual characteristics in sampling the discharges. This often means that separate samplers may need to be used for each type of airborne emission.

Iodine-131 is routinely produced for medical and industrial radioisotopes by the irradiation of tellurium targets. Depending on the plant processes, it is frequently present in the stack effluent. Assessment is complicated by the number of forms in which radio-iodine may be airborne. As discussed in the ANS/HPS N13.1-1999 standard, they are:

a) Elemental iodine vapour, which is very reactive, especially when carrier-free. It is consequently important that this component is properly sampled and is not deflected away from a long entrance nozzle type sampling device.

b) An unstable intermediate inorganic compound, known as Compound X, which may be hypo-iodous acid [HOI].

c) Organic iodide; mainly methyl iodide, with decreasing amounts of the higher homologues, ethyl iodide, propyl iodide, butyl iodide, pentyl iodide, etc. (Atkins et al. 1963). In activated charcoal sampling devices, methyl iodide is the most penetrating form of radio-iodine.

d) Droplets of an alkaline solution containing iodine; not usually found in the stack effluent but not impossible to produce in some laboratory conditions. If the droplets dry they become solid particles.

When the radio-iodine is carrier-free, approximately equal amounts of a), b) and c) are initially created upon its release.

Other radioactive gases in the stack effluent may arise from activation products, e.g. argon-41, xenon and krypton. Radioactive particles are rarely present in the stack discharges owing to the presence of High Efficiency Particulate Air (HEPA) Filters. Only in abnormal circumstances might they be detectable.

Since the Australian reactor HIFAR is moderated and cooled by heavy water, tritium is an activation product to be monitored in several of the stacks. The interest here is in the amount of tritiated water vapour released but because of its low radio-toxicity it is unlikely to be a serious safety problem.

3 PAST AND CURRENT PRACTICE IN STACK SAMPLING AT LUCAS HEIGHTS

3.1 Isokinetic Sampling for Large Particles and Droplets

3.1.1 Background

Where unfiltered stacks are used to disperse dusts, (i.e., aerosols produced by mechanical abrasion), most of the mass is carried by particles in the 50 to 100 micron size range. To accurately assess these particles isokinetic sampling is required. Isokinetic sampling is even more important when the aerosol is composed of large droplets. However, when the stack aerosols are smoke or fume, produced by condensation from vapours and coagulation from sub-micron particles, the mass median diameter is much smaller, with the maximum size usually below 10 microns. At this size isokinetic sampling, while desirable, is not essential.

Isokinetic means equal velocity; but velocity has two components, speed and direction of which the latter is the more important. The so-called 'isokinetic' samplers described in both the guidelines match the airspeed in the stack but the designers appear to have neglected the directional component. In Figure 1a in the ANS/HPS N13.1-1999 Standard, the sample of stack gases is made to turn through a 90^0 bend, along several metres of horizontal pipework, followed by a second 90^0 bend, just for operator convenience. Failure to match airspeed usually leads to over-sampling of the largest particles, which the guidelines find acceptable. However, attempts to deflect the sampled gases out of the stack leads to the largest particles or droplets impacting the walls on the bends and the horizontal

section. Such samplers should be known as ***saltation*** not isokinetic samplers, since only those particles which can bounce off the walls will reach the filter.

3.1.2 Past Experience

True isokinetic sampling is possible provided that the sampler is placed within the stack. Prior to 1966, stack sampling at Lucas Heights was by the sampler shown in Figure 1. The stack samplers were modified in 1966 to make them truly isokinetic as shown in the inset. The modified samplers were capable of sampling all particles up to 100 microns and beyond with entry losses less than 10%. They operated for a period of about six years and clearly demonstrated in that time that there were no radioactive particles, of any size, in the stack effluent from Lucas Heights (see monitoring reports form that era). The isokinetic samplers were eventually removed because :

1. Abnormal operations causing damage to the HEPA filters is an unlikely event. There have been no cases of it in the history of the Lucas Heights laboratories.

2. Smaller particles of 10 microns or less would be sampled by the non-isokinetic Maypacks with entry losses of around 30%, which is acceptable according to both M11 and ANSI/HPS N13.1. It should be noted that the requirement to fit an "isokinetic" sampling probe is advisory not mandatory in UK M11 and it is difficult to determine whether it is mandatory under the ANSI standard.

3. Fire or explosions may also damage the fabric of the building, opening alternative routes for the escape of radioactive material.

3.2 Iodine sampling

3.2.1 Background

The knowledge that radioactive particles were not a problem in the stack effluent at Lucas Heights, simplified the task of sampling for radioiodine. Isokinetic sampling is unnecessary for sampling gases, vapours, or sub-micron aerosols, so that the sample may be drawn off at right angles to the flow up the stack, which makes the design far simpler.

The Maypack is inserted through the stack wall into the stack, so that it projects into the airstream to a point where the gases are well mixed and beyond any effect that the stack wall might create. The Maypack is protected from deposition of radioactivity from the main stack flow by a short conical nozzle. This is most important since direct contamination by turbulent diffusion from the stack flow can be ten times the amount in the sampled stream (May 2002b).

The conical nozzle is short so that possible deposition on the interior of the nozzle from the sampling stream is minimised. It is estimated that, if the surface of the nozzle were a perfect sink for carrier-free elemental radioiodine, as many metals are (May 2002b), the entry losses would be at least 40%. However, Teflon coating the aluminium nozzle ensures that any deposition of the iodine on the nozzle is short-lived and the iodine desorbs

within a few seconds, so that a true sample is obtained even with short duration sampling periods.

The proof of these statements was obtained as soon as the Maypacks were mounted alongside the isokinetic samplers at Lucas Heights in 1966, when it was determined that the amount of radioiodine being discharged from the stacks was greater than had been detected by the isokinetic samplers. However, the short length (20 cm) of stainless steel nozzle on the isokinetic samplers completely absorbed the carrier-free elemental iodine vapour and about 90% of the Compound X. All the methyl iodide reached the filter paper but the ACG/B filter contained insufficient charcoal and of the wrong type to retain it for a week, after which time it was routinely analysed.

3.2.2 Current Practice at ANSTO

The iodine sampler mentioned in M11 is the original brass-bodied Maypack designed 45 years ago to distinguish between the various forms of airborne iodine. This was replaced 20 years later by the much cheaper, but superior, plastic Maypack (May 1981) which was designed at Lucas Heights specifically for stack sampling in adverse conditions, such as might occur in the case of an abnormal accident. It contained four times the quantity of charcoal of the same type as the brass-bodied Maypack. This was done because it was felt that under humid conditions the performance of the original Maypack was marginal for stack sampling and did not have sufficient reserve capacity to cope with poisoning agents, such as acid and/or organic solvent vapours, which may be present in accident conditions. This opinion still applies, especially in Australia, where high concentrations of eucalyptus vapour may also be present in summer. Thus the current ANSTO Maypack is designed to account for conditions of volatile vapours and high humidity.

The slight increase in size of the sampler brought to a head the problem of the change in counting geometry when the radioiodine penetrates more deeply into the Maypack, due to high humidity or poisoning effects at the leading edge. These effects were observed to change the apparent count rate for a given amount of activity by a factor of three, possibly higher under extreme conditions. The traditional way of dealing with this effect is to extract and mix the charcoal thoroughly, before counting in a known geometry. This is quite time consuming and can lead to cross contamination between samples. It was determined that performing a second count on the inverted Maypack and using the geometric mean of the two counts produced a determination of the activity, checked by counting at one metre in the Whole Body Monitor, which varied by only 5% over the complete range of possible activity distributions.

The plastic Maypack is still in use at ANSTO. All Maypacks are run with Whitman filter papers which would sample any particles/aerosols below 10μm in diameter. These filters are assayed separately with Alpha and Beta counters to give the gross activity. Currently, in order to ease the burden of manually intensive preparation of the Lucas Heights Maypack, the latest designed, commercially available units are being run in a parallel monitoring trial to assess their performance. If successful, they will be gradually introduced across all ANSTO facilities.

3.3 Radioactive Gas Sampling

The argon-41 release from HIFAR was assessed for many years by a weekly or monthly grab sample. This is in accordance with M11 which only calls for an annual grab sample. Currently releases of argon-41 and other noble gases to the stacks of B54, B15A and B15M are continuously measured by drawing a sample of the exhaust gas from the stack through a shielded flask, which is continuously monitored by a NaI scintillation detector and multichannel analyser. Although it was independently designed, it is exactly the same method proposed in M11 and ANSI/HPS N13.1. The detector measures the noble gas activity decaying in the flask and the activity passing into the stack can be calculated from the flask volume, the duct flow rate and the counting efficiency for each nuclide in the flask geometry. The calculation is independent of the flow rate through the flask, provided that the flask operates close to atmospheric pressure and the delay in the sampling line is not excessive. These conditions are met with a flow rate of around 4 l/min.

3.4 Tritiated Water Sampling

This is simply done by drawing air from the duct at 1 l/min through a set of four water bubblers. This is in accordance with M11. The only improvement that can be made to the procedure would be to top up the first bubbler two or three times a week in hot weather to make up for evaporation losses. Sampling for tritium is only necessary on the two HIFAR stacks (15A and 15M) and the two Waste Management stacks, buildings 20 and 57.

4 REAL TIME MONITORING

As part of development of an Environmental Management Plan for stack discharges which is a condition of the Regulation of Airborne Radioactive Discharges from ANSTO sites, ANSTO is in the process of installing real-time monitoring of radionuclides on a number of stacks at ANSTO facilities. Radioactive xenon and krypton discharge will be monitored using a sodium iodide (NaI(Tl)) gamma-ray spectrometer system and interfaced to the internal computer network. The data will be collected and sent to the network server at 10-minute intervals using locally written software to process and database the information. The discharge data will be calculated, plotted and displayed in real time. The software is also capable of interrogating the database so that the data can be graphically displayed, or retrieved in a spreadsheet format, in any combination of time intervals. This option allows comparison of discharge patterns between process runs and reactor periods as well as on-line yesterday-today comparisons. In this way operators will be able to relate stack discharges to processes and implement changes leading to minimisation of stack discharges, a part of the continual improvement philosophy under ISO 14001. The prototype system has already lead to decreases ranging from 75 to 90% in some noble gas emissions from the Building 54 stack (Blagojevic 2001).

5 CONCLUSIONS

A detailed review of the two international guidelines for stack monitoring has been conducted by Mr Fred May, an international expert in monitoring of radio-iodine. This is presented in Appendices I and II.

His conclusion is that Technical Note M11 can form the basis of stack sampling at Lucas Heights for the following reasons:

- The guideline endorses the philosophy of the use of separate and different stack sampling techniques for the different radioactive species (particulate, vapour and gases) within the same effluent. This ensures that each sampler is optimized for sampling a particular species and ensures that inlet losses are minimized (typically only a few percent).

- The guideline accepts that large radioactive particles are rarely present in nuclear plants fitted with HEPA filtration and would only be present in the abnormal case of damage to the HEPA filters, in which case other leak paths would be present.

- The guideline makes it mandatory to take all stack samples from a position where the stack gases are well-mixed.

- The guideline recommends the use of calibrated flowmeters in both sampler and stacks. Present ANSTO procedures are to check the flow through the samplers each week, but the stack flow is only measured annually, or when a filter change has been made. This procedure could lead to over-estimates in radionuclide releases.

In general, sampling at Lucas Heights complies with all the mandatory requirements of M11 and where it differs from the advice in the UK M11 Technical Note, the techniques adopted are almost always more stringent and prudent than those advocated in M11. An audit of all stack sampling at ANSTO facilities will allow any gaps in the current sampling practices to be identified and action plans for upgrades to be formulated.

APPENDIX I

Based on a Review of Technical Guidance Note M11

Monitoring of Radioactive Releases to Atmosphere from Nuclear Facilities

by F.G. May

This Technical Guidance Note is issued by the UK Environment Agency for guidance to the UK nuclear industry, among others, on radioactive discharges to the atmosphere. In particular, the principles of monitoring, system design and application, and the different types of radioactivity are discussed. Quality assurance procedures and inspection guidelines help maintain high standards of data collection.

This review will proceed sequentially through the Technical Guidance Note and highlight areas of interest to the ANSTO stack monitoring operations that require comments.

SECTION *4.2 GENERAL SAMPLING TECHNIQUES*

"Different sampling techniques are required for different physical forms of radioactive materials. The main forms are particulate, vapors and gases. Thus different radioactive species within the same effluent may require separate sampling collection and analytical methods."

This is a most important statement and fully endorses the stack sampling philosophy adopted at ANSTO.

Section 4.2.1 Sample extraction

"In order to obtain a representative sample for compliance monitoring purposes, several conditions must be fulfilled, namely:

(a) *"Sampling must take place downstream of any abatement plant."*

Agreed that this is a mandatory requirement.

(b) *"The sample **must** be taken at a position in the emission stack where all constituents are adequately mixed."*

Homogeneous mixing has not been checked at ANSTO and will be covered in the future action plan to ensure compliance with M11.

(c) *"The sample **must** be taken well away from any ductwork features such as dampers, bends and merged streams, which may have a detrimental effect on mixing and flow patterns, or a well mixed flow **must** be demonstrated at the plane where the sample is taken."*

All the sampling positions at ANSTO meet this requirement even though it is only strictly necessary for sampling particles and droplets larger than about 5 μm and should not be mandatory for sampling vapours, gases and sub-micron particles.

(d) *"The sample **should** ideally be taken via a probe and this **must** be pointing upstream."*

Note that the requirement for a probe is advisory only. If a probe is employed then it is mandatory that the probe be directed upstream. This advice should be for particulate sampling only. It is an essential requirement for sampling large particles (>50 μm) and is highly desirable for particles >10 μm. It is unnecessary when sampling gases, vapours or sub-micron aerosols. The trapping efficiency curves for the Casella Cascade Impactor are relevant in demonstrating the size of aerosol that requires isokinetic sampling and the sizes for which it is merely desirable.

"it may be noted that ventilation fans are effective mixing devices."

Good use of this has always been a part of stack sampling at Lucas Heights.

"In the case of particulate material, it is necessary to use a probe with a nozzle designed to ensure that isokinetic sampling is taking place and that the sample being taken is truly representative in respect of particle size distribution, i.e. no particles are rejected or preferentially drawn into the sampling probe. This requires the following conditions to be fulfilled:

*(a) The entry nozzle of the probe **must** point upstream."*

Agreed.

(b) *"The velocity at the nozzle inlet due to sample extraction* ***must*** *be equal to that in the duct or stack."*

This is an essential requirement for isokinetic sampling of large particles. However it should be pointed out that if the velocity is **less** than the stack velocity, the sampler will over-sample the largest particles which is an acceptable fault in most international guidelines on stack sampling.

(c) *"The nozzle must have a sharp (feathered) leading edge.*

Agreed.

(d) *"In dusty streams, the inside diameter of the nozzle at the inlet generally needs to be > or =10 mm. However, in nuclear plants the particulate loadings are usually very low, because of the use of HEPA filtration. In such cases smaller nozzles may be used to achieve isokinetic sampling conditions."*

The Lucas Heights isokinetic samplers of 1966 had nozzles > 10 mm in diameter. The results they gave for the succeeding six years confirmed this statement that radioactive particulate loadings were very low to not detectable.

It is suggested the list of **mandatory** conditions for isokinetic sampling is incomplete. In any application to ANSTO it should have the following:

(e) **The distance from the nozzle of the probe to the sampling filter must be as short as possible with a gradually expanding area of cross-section and without bends, which would cause the largest particles to impact the tube walls. This means that the sample must NOT be drawn out of the stack before collection of the particulate sample.**

The increasing area of cross-section is required to reduce the velocity of the sample stream to that which can be drawn through the filter in a controlled fashion (May 2002a). This requirement would mean the alteration of figure 3, figure 5 and figure 7. The sampler they describe is not an isokinetic sampler but a **saltation** sampler, since any large particles that reach the sample filter have bounced around the corners and along the horizontal pipework. Large droplets or deliquescent particles cannot do so and would be severely depleted. This violates the initial statement about particle rejection.

"It is common practice, however, to extract a single sample through a probe designed for isokinetic sampling for the purpose of measuring both particulate and vapor/gas forms of radioactive material in the effluent."

It is considered that this is not a good idea and contradicts the opening statement in section 4.2.

"In some circumstances, such as when sampling from the top of a high stack, a primary/secondary system is used. Here, a large-diameter (~150 mm) probe is used to extract a sample to a suitably accessible location where the primary sample flow is itself sampled by a conventional small-diameter probe. The primary flow is then returned

upstream of the discharge fan, so that it is driven by the fan pressure differential. The primary line is designed to minimise deposition and plate out effects and is normally trace-heated."

The Authors of this review disagree with this advice for the following reason:

> This suggestion, that it is possible to design the primary line to minimise plate out and deposition, is not plausible. Also the idea that trace heating of a transport line solves the problem of plate-out of carrier-free radioiodine vapour is a misconception, since heating the surface may promote the chemisorption effects one is trying to suppress. The only safe method of suppressing plate out is to Teflon coat the sampling nozzle and keep it as short as possible (<10 cm) . This means inserting it into the base of the stack.

Figure 4 Maypack insert

The illustration is of the highly expensive brass-bodied Maypack designed 45 years ago to distinguish between the four different forms of airborne radioiodine in laboratory experiments (Megaw and May, 1962). At Lucas Heights it was replaced by the superior plastic Maypack, designed at Lucas Heights specifically for stack sampling (May, 1981). It contains four times the amount of charcoal of the same type as the brass-bodied Maypack, because it was felt that the performance of the original Maypack was marginal for stack sampling in high humidity. In addition there may have been complications due to high concentrations of eucalyptus oil vapours in the air during Australian summers. The original Maypack did not have sufficient reserve capacity to cope with additional acidic or other organic vapours, which might be present in the some stacks. The current Maypack is designed to cope with all these conditions.

Figure 5 Stack sampler with Maypack insert

The illustration is of an over-engineered sampling arrangement suitable for removing a sample from a pressurised duct, always providing that one could ignore the inlet losses of the carrier-free elemental radioiodine, the Compound X [HOI] and the large particles. Since both the stack and the ducts leading to the base of the stack are at atmospheric pressure, such a construction is unnecessary and the inlet losses to the stack sample would be large. The Maypack should be inserted into the stack itself; shrouded from the stack gases by the outer holder and protected from direct deposition of radioiodine from the main stack flow by a short Teflon-coated conical nozzle. The fact that the sample is taken at right angles to the main flow is of no consequence with vapours or sub-micron aerosols and the inlet losses for particles up to 10 μm should be less than 30%, which is considered acceptable.

4.2.2.3 Gases

(a) ***Bubbler traps*** *are used to effect a reaction between a sample of the gaseous radioactive material and a liquid collection medium, resulting in the transfer of the radioactive material from the effluent stream to the liquid sampling medium by a chemical or physical reaction. In order to effect this reaction, the sample gas is*

passed into a bottle containing the liquid sampling medium through an inlet tube and is allowed to bubble through the liquid to an outlet tube. This ensures good contact between the sample gas and bubbler liquor to ensure a complete reaction. Bubbler traps are widely used for sampling tritium in both elemental and tritiated water (oxide) form.

This is a simple description of a bubbler trap, which is the preferred method for trapping tritiated water **vapour** not hydrogen gas. Elemental tritium is rarely a problem but, where it is, must be oxidised to water vapour before trapping in a bubbling train. One bubbler is rarely enough. A train of four or five bubblers each filled with 200 ml of water is normally required for a weekly sample at a flow rate of 1 l/min. Even then the first bubbler may require to be topped up to make good evaporation losses in hot weather. In very cold weather ethylene glycol may need to be added to prevent freezing. To ensure good contact it is usual to employ a sintered glass frit at the bottom of the inlet tube to produce tiny bubbles.

(b) ***Wet scrubber** is a means of transferring a sample of gaseous radioactive material from the effluent stream to a liquid collection medium. It is primarily used for ruthenium. It operates on the same principle as scrubbers used in the treatment of gas streams to remove radioactive material in particulate or vapor forms. It consists typically of a glass cylinder filled with glass beads/tubing or other inert material. A recirculating flow of liquor passes down through the cylinder over the inert material whilst the gas to be sampled passes upwards .....*

The use of a small counter-flow scrubbing tower is not sufficiently different from the use of a bubbling train to warrant a separate entry. It is disputed whether bubblers or scrubbing towers will trap or sample radioactive **gases.**

(c) ***Direct measurement** is used when the chemical form of the radioactive material is such that it is not practicable to remove it from the emission. This is particularly the case for radioisotopes of inert gases such as krypton and xenon. The emission is drawn into a 'measurement chamber', which is of fixed calibrated dimensions and controlled temperature. A detector and associated electronics are positioned to measure the activity in the chamber. This measurement can be related to the fixed volume of sample in the chamber, and the activity concentration can then be assessed making appropriate temperature corrections.*

It is impossible to tell from this description whether the sampling technique is a 'grab sample' or a 'continuous flow' gas sampler but, whichever technique it is, temperature correction is one of the least important criteria to be taken into account. Grab samples are only used where the rate of emission is fairly constant, e.g. for argon-41 produced in reactor shield cooling air by the activation of natural argon in the air. The rate of production is a function of neutron flux and volume of air within the shield.

A continuous flow sampler can be used for monitoring any radioactive gas, including noble gases, which may be released intermittently from radio-pharmaceutical production facilities. Air is drawn through a **shielded** 'measurement chamber', which is continuously monitored by a NaI scintillation counter and multichannel analyser. The activity decaying within the known volume of the chamber, together with the measured flow rate up the stack or duct, permits the total

activity of each radioactive gas in the stack effluent to be determined. It is most important that the pressure in the measurement chamber be close to atmospheric pressure and the time spent in traversing the transfer pipework be short compared with the half-life of the radioisotopes being measured. The shielding around the measurement chamber must be adequate to avoid picking up the radiation from the main duct. None of these points are mentioned.

System design

Figure 7 Typical system for extracting a sample

This schematic would be acceptable if the position of the sample collector was moved into the stack or duct for both isokinetic and radioiodine sampling.

5.1 Sample extraction

.... It is particularly important when sampling for particulates and condensable vapors that the length of pipework between the probe and the sample collector is ***as short as possible*** *in order to minimise condensation/plate-out and avoid unrepresentative sampling. Similarly* ***bends*** *in the pipework should be minimised so that any deposition of radioactive material by centripetal separation before collection is as small as possible. If bends are necessary,.....*

Bends can be eliminated if the sampler is placed in the stack or ductwork, which will also minimise the length of nozzle.

5.2 Deposition effects

Some volatile species such as isotopes of iodine and sulphur can 'plate-out' in sample lines. This is particularly enhanced due to thermophoresis deposition when sample lines are cold relative to the effluent.

Reactive vapours, such as carrier-free elemental radioiodine and HOI often plate-out in sample lines, especially if they are metallic or long. Thermophoresis only affects sub-micron aerosols and is caused by the difference in Brownian motion of the air molecules on the two sides of the particle driving the particle down a temperature gradient (Goldsmith and May, 1966). It has no effect on the movement of vapours.

In some circumstances, such as where the use of long sampling line is unavoidable, computer models can be used to access deposition effects and to optimise design of the sampling system.

This is not considered a sensible suggestion. Long sampling lines should be avoided under all circumstances. Only methyl iodide will pass down a long sampling line and computer programs are only as good as the original experimental data, which does not appear to have been done with carrier-free elemental radioiodine vapour. Trace heating the

long sampling line may cause more problems than it solves, since it may provoke breakdown of the methyl iodide.

Conclusions

While the stack sampling at ANSTO meets the mandatory requirements of the UK M11 Technical Guidance Note on stack sampling, there are a number of areas in which it is deficient and several pieces of advice contained in M11 may be misleading. Nevertheless it is felt that the M11 guideline is most appropriate to the Australian situation.

APPENDIX II

Based on a Review of ANSI/HPS N13.1-1999

Sampling and Monitoring Releases of Airborne Radioactive Substances from The Stacks and Ducts of Nuclear Facilities

by F.G. May

The American National Standard N13.1 establishes guidelines and performance criteria for sampling of airborne radioactive discharges at nuclear facilities. Sampling probes, transport lines, sample collectors, monitoring instruments are discussed. Like the UK Guidance Note, quality assurance issues and system optimisation are emphasized.

This review will proceed sequentially through the Technical Guidance Note and highlight areas of interest to the ANSTO stack monitoring operations that require comments.

Figure 1a – Generic sampling system, illustrating a flow schematic where components are identified.

This diagram illustrates the problem that runs through the entire standard; the sample of stack gas is shown being removed from the stack before collection. It is extracted from the centre of the stack via an isokinetic nozzle, traverses a 90^0 bend, several metres of horizontal pipe, another 90^0 bend and a vertical pipe before filtration. A simpler and, it is suggested, superior method of placing the collector **within** the stack is not considered anywhere in the standard. This in stack monitoring is the normal method for sampling radioiodine vapours and particulate activity at ANSTO (May, 1981).

Figure 1b – Generic sampling system, illustrating a component hierarchy of a sampling system.

Placing the stack sampler within the stack simplifies this hierarchy and makes much of it redundant.

3.1 Definitions

Isokinetic: *A condition that prevails when the velocity of air at the inlet plane of a nozzle is equal to the velocity of undisturbed air in a stack or duct at the point where the nozzle is located.*

There is no quarrel with this definition of isokinetic. However, there are several points to be made:

1. In the opinion of the reviewers, the need for isokinetic stack sampling has been overstated; isokinetic sampling is absolutely essential for particles >50 μm, highly desirable for particles >20 μm, but completely unnecessary for particles <5 μm. At 10 μm the need for isokinetic sampling is debatable (May 2002a).

2. Velocity consists of two components; airspeed and direction. While entry to the nozzle is isokinetic, the flow must also not change direction in the transport line to the collector. If it does, it is **not** isokinetic sampling. The airspeed of the sample flow must be reduced before it can be filtered. This is done by incorporating a smoothly expanding tapered section after the nozzle, but no bends can be permitted. This usually means that the isokinetic sampler must be totally within the stack. The particulate samplers described in N13.1 are not isokinetic but **saltation** samplers, since most of the large particles reaching the filter have bounced around the bends and along the horizontal pipework.

4.3.2 Sampling for aerosol particles

3rd paragraph.

Although the particle size most likely to penetrate HEPA (high efficiency particulate air) filter media is in the 0.1 – 0.3 μm diameter range, it is erroneous to assume that the sampling system need only be designed for sub-micrometer particles. Larger-size particles may be transmitted through HEPA filter banks due to small openings in HEPA frames, gasket seals, and filter media defects developing especially after extended periods of use.

Current Quality Assurance on HEPA filters demands that a penetration test, usually 'Cold DOP', is performed after a filter change or installation and any defects made good with silicone sealant. The performance of HEPA filters actually improves with increasing dust load. Even the coarse filters that are used to extend the working life of the HEPA filters will remove the particles for which isokinetic sampling is necessary.

3rd paragraph

***When accidental or off-normal conditions occur,** a wide range of particle sizes may be present in the effluent, and this **shall** be a factor in the design. Particles well into the **inertial** size range (sizes larger than about 2 μm aerodynamic diameter, AD) should be considered to be present.*

Particles of 2 μm have so little inertia that they easily penetrate commercial cyclones. It is true that HEPA filters trap particles down to 0.3 μm by their inertia, but this is only because Brownian motion is ineffective above 0.3 μm. It does not mean that isokinetic sampling is called for at these sizes.

4th paragraph

***But the accident case** is much harder to characterize in advance because changes in effluent discharge rate, added dust, smoke, and debris may be expected. However, it should be noted **that particles greater than 100 μm would not be expected to occur in large numbers** in a stack effluent due to gravitational settling and inertial impaction removal effects during transport in the accident environment, and in the ductwork leading to the stack. **Therefore, there are upper bounds to particle sizes that need to be evaluated.** For typical stack conditions, evaluation with 10 μm AD particles should suffice; for the accident case, consideration of transport of somewhat larger particles is prudent.*

The mass median aerodynamic diameter (MMAD) for mechanically abraded dusts is usually in the range 50 – 100 μm, for which true isokinetic sampling is essential. This aspect of the standard seems to suggest that these large particles can be ignored even if present under accident conditions. So-called "saltation" samplers cannot cope with this size of particle. To test the assertion that test particles of 10 μm size are appropriate, it would better to look at the trapping efficiency curves for the Casella Cascade Impactor which demonstrate the size of aerosol that requires isokinetic sampling and the sizes for which it is merely desirable.

4.3.3 Concerns for large particles

last paragraph

*As previously noted use of HEPA filtration upstream of the sample withdrawal location does not eliminate all concerns for large particles due to possible transmission through leaks in frames and seals. However, even nonfilterable radioactive gases and vapors may become associated with large particles by adsorption effects. **For example, radioactive vapors or gases penetrating filtration can deposit on downstream stack or duct walls and become incorporated into surface layers, corrosion or rust.** Subsequent flaking or shedding of these layers may lead to the release of large radioactive particles that may go undetected by sampling systems that are optimized for collection of small particles.*

This argument is not plausible. First; radioactive gases do not deposit on stack surfaces, even though their short-lived daughters may do so. Second, even if the walls become contaminated with radioactive vapours, such as iodine-131 or sulpur-35, the hazard arising from any rust particles spalling from the walls would be orders of magnitude smaller than that arising from the release of the contaminating vapour, which normally has already been sampled.

4.4 Determining action levels

8th paragraph

***For example, effluents containing highly reactive constituents such as radioiodine may be particularly difficult to extract and transport without significant sampling bias,** leading to higher estimated sampling accuracy uncertainties than 20%.*

Taking the sample of stack gas through a completely unnecessary probe and long sampling line can be guaranteed to lose all the carrier-free elemental radioiodine and HOI. The only **prudent** place to place the Maypack iodine sampler is within the stack and fitted with the shortest possible Teflon coated conical inlet nozzle.

Last paragraph

...if the sampling nozzle and transport line are not properly designed and properly placed, uncertainty will be created in sampling accuracy, and no amount of attention to measurement accuracy in the system will prevent the system from generating poor, biased data, and faulty alarm responses.

This is strongly agreed.

5.2 Selection of sampling sites

5.2.1 General considerations

1st paragraph

Generally, the sample extraction location should be situated between the discharge plane of a fan and the stack exit plane, with caution that the location should not be so close to the stack exit plane that wind effects can significantly influence the velocity profile at the sampling location. In addition, the sampling location should be in a region where the contaminant profile is well mixed and stable, it should be readily and safely accessible, it should not present a problem for sampling services and maintenance activities, and it should be able to accommodate analysis or collection equipment ***that does not compromise the quality of the sample.***

Agreed.

6.2 Bulk stream volumetric flow measurement

1st paragraph

Accurate measurements of the flow in stacks and ducts must be provided because the accuracy of any emissions estimate is directly related to the accuracy of flow measurements. The flowrate of air exhausted through a stack or duct ***shall be periodically measured and may need to be continuously monitored if there is a potential for significant emissions.***

The stack flow rate is measured annually and again immediately after a HEPA filter change, so that the stack flow is always equal or less than the latest measured flow. This ensures that the quoted stack discharges are always a slight over-estimate. This is permitted under the regulations but continuous measurement of stack flow by a built-in meter with provision for recording the previous 48 hours might be a worthwhile improvement, especially to cover potential accident conditions.

6.3 Nozzle design

6.3.1 Basic considerations

1st paragraph

However, the use of nozzles that have constant internal cross sections and a 90^0 bend of the same cross section is ***no longer considered good practice*** *due to substantial aerosol particle losses in both the straight entrance region and the bend, and due to errors associated with off-design operational conditions (Fan et al. 1992; McFarland and Rodgers 1993).*

Also, ANSI N113.1-1969 recommended the deployment of multiple nozzles in circular ducts greater than 6-in. (152 mm) diameter or in rectangular ducts with cross-sectional areas greater than 0.5 square foot (0.046 sq. m). For larger ducts as many as 20 nozzles were recommended, with rakes (several nozzles on a common manifold) of such nozzles spanning the stack or duct. ***The use of these rakes is no longer considered good practice*** *because for a given flowrate, smaller nozzle inlet diameters are used as the number of nozzles is increased to accommodate the ANSI N13.1-1969 recommendation of isokinetic (air velocity at the inlet plane matching that of the free stream air velocity) sampling. The use of large numbers of small nozzles exacerbates sampling deficiencies inherent in the ANSI N13.1-1969 nozzle design.*

The reviewers wonder whether the American standards committee made a mistake when writing this section of the Standard in 1969. The difficulty of tuning each nozzle to be isokinetic in each locality, especially with varying lengths of transport line, was never an appealing proposition and was surely unnecessary provided that the mandatory condition of good mixing at the sampling position was met.

2nd paragraph

Studies have shown that isokinetic operation is not a prerequisite for obtaining representative samples (McFarland and Rodgers 1993).

This comment only applies for particles at a MMAD of 10 μm, but not to the 50–100 μm particles which may be released in the damaged HEPA filter condition.

6.3.3 Nozzle designs

1st paragraph

Fan et al (1992) wind tunnel tested an elbowed nozzle with a nearly constant internal diameter designed in accordance with ANSI N13.1-1969. They found the wall loss ratio was approximately 75% for 10 μm *AD aerosol particles at isokinetic conditions over a range of stack velocities.*

This comment shows the importance of removing bends in an isokinetic sampler.

2nd paragraph

Modern nozzle designs have better performance than the original ANSI nozzles. As an example, McFarland et al (1989) and Chandra and McFarland (1995) have developed shrouded nozzle designs that considerably reduce wall losses.

The physics of this system should be investigated/discussed.

Cont 2nd paragraph

Figure 2a shows a typical shrouded nozzle that was designed for use in stack sampling in the nuclear industry. The transmission of 10 μm AD aerosol particles through the nozzle as a function of free stream velocity is shown in figure 2b. In general terms the wall losses are about one-fifth of those of the ANSI/HPS N13.1-1969 nozzles tested by Fan et al. (1992)

The shroud is close fitting around the tapered knife-edge nozzle and it is difficult to see from the laws of physics why it should have the beneficial effects claimed. It does not appear that tests were carried out with aerosols of a different size.

3rd paragraph

*The shrouded nozzles are designed to be compatible with single point sampling, where the sampler is operated at a constant flowrate with the **flow velocity in the shroud being about 30% that of the nominal stack free stream velocity.***

This does not seem to be compatible with the concept of isokinetic sampling.

4th paragraph

The transmission ratio for 10 μm Ad aerosol particles is relatively unaffected by variations in stack velocity, flowrate, free stream turbulence and angle between the free stream and nozzle entrance.

This is a peculiar statement to make in view of most of the other statements about the need for isokinetic sampling and the 75% wall losses found by Fan et al. (1992).

4th paragraph

*Williamson et al. (1987) and Chandra (1992) designed sharp-edged **unshrouded** nozzles that have wall losses of 10 μm AD particles that are approximately half of those of the ANSI N113.1-1969 nozzles.*

This seems to add to the confusion. Perhaps if a truly isokinetic sampler (without bends) was designed and tested with a wider range of particle size the confusion would be resolved. Wind-tunnel tests of such a sampler with 30 •m particles showed wall losses of less than 2%, May (2002a).

6.3.4.5 Nozzle configuration

The leading edge of the nozzle should have a sharp edge with external cone angle not to exceed 30 degrees. Other configurations may be used if experimental data show either

equivalent or superior performance to the sharp edged nozzle. ***If the sampling nozzle is shrouded, the shroud should not have a sharp leading edge.***

There are good reasons for the sharp leading edge from the laws of physics. The only justification for the shrouds appears to be some experiments with a single size of particle. It is suggested that these experiments would benefit from being repeated with a true isokinetic sampler and a wider range of particles, especially those around 50 μm.

6.3.4.9 New concepts

When new approaches are developed for design and operation of nozzles, such designs may be used in ducts and stacks if it can be demonstrated experimentally that the designs meet, or exceed, the performance specifications given in clause 6.3.2. The test conditions should include experiments to determine the wall losses and aerosol transmission at conditions of 1) particle sizes of 3, 10, and 20 μm *AD at the nominal free stream velocity and nominal flowrate; 2) maximum and minimum operational or anticipated free stream velocities for a particle size of 100* μm *AD at the nominal sampling flowrate; and 3) maximum and minimum anticipated sampling flowrates for a particle size of 10* μm *AD at the nominal free stream velocity.*

Test conditions 2 & 3 would only differ from condition 1 if the flow in both the stack and sampler vary markedly from the nominal rate. It is debatable that new approaches are required, since true isokinetic sampling was in use both in Britain and Australia over 35 years ago.

6.4.1 Depositional losses

2nd paragraph

The straight sections of transport tubes, particularly horizontal tubing sections, should be kept as short as possible, and the number of bends should be minimized within the geometrical constraints of the application. There should be no inward facing steps at the tubing connections that cause more than a 1% reduction in tube diameter. The tubing ends shall be free from burrs and crimping. For record samples, it is recommended that not more than one 90^0 *bend be used, although there may be applications where more than one bend may be necessary.....*

In general terms, there will be some losses of aerosol particles in transport lines, *and any design will entail compromises.*

The biggest compromise is to remove the air sample from the stack before collection of the aerosols and vapours.

6.5 Gas and vapor sample extraction and transport

1st paragraph

..... however, consideration shall be given to extracting and transporting vapors and gases to determine where special system design may be required. ***There are instances where gases may be monitored in the stack using an in-stack radiation detector. In these instances, sample extraction and transport are not considerations.***

Stack sampling for radioactive gases is one of the few occasions when it is quite permissible to draw the sample down a fairly long transport line without undesirable repercussions. Whereas to place the radiation monitor within the stack means trying to measure the activity in a high and variable background radiation field. It is difficult to provide sufficient shielding within the stack and the long and tortuous path that the inlet pipe must take through the shielding means severe inlet losses for both radioiodine and large particles.

Last paragraph

..... The minimum transport efficiency for vapor or gas samples from the free stream to the collector/ analyser should be 50%.

This figure of 50% seems to be too liberal in view of the fact that radioiodine is usually the critical nuclide in any release of fresh fission products to the atmosphere. An under-estimate of the iodine-131 release by a factor of two should not be tolerable. There should be no problem in meeting a transport efficiency of over 90% for gases since there are no deposition problems. Reactive vapours, such as elemental iodine and HOI, are much more difficult, especially when carrier-free. The only satisfactory way of dealing with this problem is to mount the iodine sampler, Maypack, so that it projects into the stack or duct at a point where good mixing obtains, with only a short conical Teflon-coated inlet nozzle to protect the sampler from direct deposition of iodine vapours from out of the main stream. When this is done, transport efficiency is over 98% (May, 1981). This is the current situation with the ANSTO Maypack samplers.

6.6.2 Filter media

1st paragraph page 42

..... As shown in figure 3, filters typically have a minimum collection efficiency for particles that are approximately 0.2 – 0.5 μm diameter. Above about 0.3 μm diameter, filtration efficiency increases due to inertial impaction and below this size efficiency increases due to Brownian diffusion.

Page 45

Figure 3 – Schematic of filtration efficiency vs. particle size illustrating the different filtration regimes (Lee and Ramamurthi 1993)

This graph is appropriate for a coarse filter and is only suitable for protecting and extending the working life of HEPA filters. It is certainly not appropriate in stack sampling for radioactive particles, which only employ HEPA filters. The fact that that part of the graph above 0.3 μm is labelled "Inertial, Impaction and Interception Regime" is confusing when considering the question of the need for isokinetic sampling. Of more use would be the trapping efficiency curves for the Casella Cascade Impactor which would be much more relevant in demonstrating the size of aerosol that requires isokinetic sampling and the sizes for which it is merely desirable.

6.10 4 Upgrading and retrofit of existing stacks

Many existing nuclear facilities have stack sampling systems built during the 1960's through 1980's, which have isokinetic sampling with multiple small-diameter nozzles. In addition, many facilities have flow systems that are fitted with flow straighteners, which serve the purpose of making the velocity profile uniform, but which impede mixing of contaminant mass in the flow stream. ***It is now known that if an aerosol contaminant is not well mixed, and a multitude of poorly designed, small diameter nozzles are deployed, seriously nonrepresentative samples may result.***

This may have resulted from the compliance of these establishments with ANSI/HPS N113.1-1969.

7.6.1 Calibration of sampling system flowmeters

5th paragraph

Rotameters shall be calibrated at flowrate conditions that correspond to the average anticipated flowrate during sampling, and at 75% and 125% of the anticipated sampling flowrate.

There should be a warning here that filter blockages could cause pressure changes at the rotameter causing marked changes in calibration.

Annex C Special considerations for the extraction, transport, and sampling of radioiodine.

C.1 General considerations

Obtaining samples of airborne radioiodine is complicated because it is present in air effluents in several forms, viz: particulate matter, elemental iodine (I_2), hypoiodous acid (HOI), and in organic form, principally methyl iodide (CH_3I). The existence of the HOI form is not universally accepted, but it is postulated to be that otherwise indeterminate form with a deposition velocity lower than that of elemental iodine and that it will penetrate a cadmium iodide bed but will be collected by an iodophenol bed in a species sampler.

These chemical forms of radioiodine, particularly the elemental form, may be expected to initially deposit in ducts and in sampling lines and then subsequently be resuspended and emitted as the same or another form (Cline 1991).

The deposition of elemental iodine in sampling lines is especially high for carrier-free radioiodine (May, 2002b, Chamberlain, 1963). It cannot be relied upon to desorb readily from long sampling lines even when coated with special materials, such as Teflon, and use of such lines can lead to significant under-estimates of the iodine release.

Cont 2nd paragraph

The organic form is the least depositing and only a small fraction is collected by some of the solid adsorbents that are used to limit radioiodine emissions (Kovach 1980).

This statement seems appropriate to the USA. Elsewhere the solid absorbents used to trap radioiodine were impregnated charcoal specially developed in England for the capture of methyl iodide at high humidity. They also trapped the other forms of iodine vapour even more readily. (Collins 1962)

Cont 2nd paragraph

However, it cannot be ruled out that during off-normal events the major form of radioiodine will be elemental. Therefore, evaluations of sample transmission under off-normal conditions should assume that form. Glissmeyer and Sehmel (1991)) summarized the more recent studies on radioiodine sampling and transport, and many of the following considerations are based on that summary.

It is always more prudent to assume that there will be a mixture of radioiodine forms released under both normal and off-normal conditions. It is true that it is usual these days to pass any effluent likely to contain appreciable amounts of iodine-131 through an impregnated charcoal trap, which will remove most of the methyl iodide and all the other forms of iodine vapour, however minor accidents can lead to some of the effluent bypassing the charcoal beds. This means that any form may be present in the off-normal situation. When only methyl iodide is present in the normal effluent, transport losses will be small even when the sampling lines are long. This may lead to a lack of realisation of the importance of short sampling lines in off-normal situations.

C.2 Extraction and transport

The considerations for the extraction of gases and vapors set forth in clause 4.3 of this standard are applicable to radioiodine. ***In view of the likelihood that at least some of the radioiodine in the air effluent will be attached to particulates, all of considerations applicable to them as set forth in the main body of this standard and its annexes also apply to the extraction and transport of radioiodine.***

This is not considered correct by the reviewers. Particulate bound iodine is hardly ever present in stack effluent as evidenced by the post 1966 isokinetic sampling. It was thought in the early 60's that elemental iodine might adsorb on the surface of aerosols in a similar fashion to the behaviour demonstrated by radon and thoron daughter products. However this was rapidly shown not to be the case, since the only aerosol that radioiodine vapour showed any affinity for was one of silver (Clough et al. 1965) and silver aerosols are not particularly common in stack effluents even in off-normal situations. This is not to say that particulate radioiodine cannot be produced, since droplets containing radioiodine are produced as a spray in some iodination techniques and submicron satellite drops are produced when bubbles burst during electrolysis of radioiodine solutions. They would normally be removed from the effluent by the HEPA filters. Should they bypass the filtration system, they would be caught on the HEPA filter in the Maypack. The possible presence of particulate iodine cannot be used to justify the sampling probe and long transport line that are called for by this standard.

2nd paragraph

Laboratory studies have shown that in the extraction and transport of radioiodine, materials that contact the radionuclide may interact with it (e.g. , copper, PVC, Buna-N). These materials should be avoided. Studies by Kabat (1983) indicate that the preferred materials are Teflon, polyethylene, aluminium, carbon steel, and stainless steel.

Earlier studies (May 2002b, Chamberlain 1963) had shown that carrier-free elemental radioiodine vapour reacted with aluminium surfaces as if they were perfect sinks but, when carrier iodine was present, the aluminium surface saturated at a small fraction of a monolayer. Similar effects were observed on steel and magnox surfaces, but that freshly etched copper continued to act as a perfect sink even though many monolayers of copper iodide had formed on the surface. Other methods of cleaning the copper surface caused saturation of the surface at a monolayer. Obviously the oxide layer on some of the surfaces was interfering with the deposition of the iodine, but it would be imprudent to rely on the oxide layer on aluminium or steel to prevent any losses down a transport line composed of these materials. Glass surfaces were not perfect sinks even when the iodine was carrier-free but neither were they completely inert. Paraffin wax showed the least deposition of carrier-free radioiodine with Teflon a close second. The only prudent conclusion was that sampling nozzles should be coated with either paraffin wax or Teflon and kept as short as possible, since these surfaces could acquire a patina of dust in use which would change their deposition characteristics.

3rd paragraph up on page 69

In a summary of sampling systems for reactors, Glissmeyer and Sehmel (1991) indicate that a typical sampling system consists of a 15-mm diameter (5/8 in.) stainless steel transport line that is about 52 m (170 ft) in length with a flowrate of about 57 liter/min (2 cfm). Simulated sample transport lines with a range of similar designs were tested by Unrein et al. (1985) who measured short-term (on the order of two hours) penetration factors of 0.62 for injected ^{131}I (as I_2) through 19 mm (3/4 in.) diameter by 48 m (1400 ft) long tubes. Glissmeyer and Sehmel (1991) predicted the equilibrium penetration factor to be about 0.75 with an approximate time of two weeks to reach equilibrium.

Long sampling lines are not advisable for many reasons discussed above. The only place for a rapid and accurate measurement of the radioiodine discharge is to place the Maypack in the stack, with only a short Teflon coated nozzle for the sample to traverse.

C.3 Collection media for radioiodine

While carbon is an efficient collector of I_2, it is much less efficient for the low deposition velocity organic iodides. Kovach (1992)) has indicated that the removal mechanism of elemental iodine on carbon adsorbents is primarily by physical absorption, the removal of hydrogen iodide (HOI) by physical absorption, chemical reaction and isotopic exchange, and that the removal of organic iodides such as CH_3I is by isotopic exchange. To improve the latter process, carbon filter media treated with potassium iodide (KI) or triethylene-diamine (TEDA) should be utilized. Packages with different types of adsorbents in series are available for the collection of radioiodine with separation by chemical form.

There are many substances that can form the basis of an elemental iodine removal plant, from caustic soda solutions, silver knitmesh, silver nitrate and charcoal, but only very special, impregnated charcoals can trap organic iodide at high humidity. The trapping of elemental iodine on unimpregnated coconut charcoal is largely by physical absorption with only a small amount of chemisorption taking place. This is why around 90% of the iodine can be driven off this type of charcoal if air at 100^0C is passed through the bed for a few hours (R.D. Collins, 1962). However, if the air temperature is raised to 200^0 C, chemisorption is promoted and little iodine is released (May, 1961). Moreover, with the impregnations of TEDA and KI developed by A.D. Collins (1967) the absorption, which may be physical at first, rapidly becomes chemisorption and the iodine is permanently absorbed on the charcoal. HOI (which is hypoiodous acid not hydrogen iodide, HI) is almost certainly chemisorbed also. Isotopic exchange is certainly not the mechanism for the trapping of methyl iodide. There is very little stable iodine in the TEDA impregnated charcoal with which to exchange and the behaviour of the iodine on the KI impregnated charcoal does not fit with isotopic exchange. In both cases the organic iodide is permanently chemisorbed on the charcoal (May and Polson, 1974).

It is interesting that there is now another attempt to separate the chemical forms of airborne radioiodine by their behaviour in different types of adsorbents in series, since this is how the Maypack started (Megaw and May, 1962)).

F.3.2 Radioiodine sampling examples

2^{nd} paragraph

Curtis and Guest (1986) used stable methyl iodide and elemental iodine injected into the stack flow upstream of the fan. Collected iodine samples were analysed using neutron activation.

Unfortunately neutron activation cannot be used at the extremely low concentrations that represent carrier-free iodine-131, even when the possible presence of iodine-129 is allowed for. There is such a difference in deposition between carrier-free radioiodine and the usual low chemical quantities of elemental iodine on most metal surfaces that these tests cannot be applied to real conditions of stack sampling. No indication is given of the levels of stable 'carrier' iodine in the other experimental work quoted in the report, but it is known that most release methods involve the presence of carrier iodine. It is usually added by chemists to swamp the side reactions leading to the production of organic iodine and HOI, which misses the point completely, since it is the complicated mix of different behaviour of carrier-free airborne radioiodine that is reality.

Annex G Transuranic aerosol particulate characteristics

G.2 HEPA filtration effects

Nuclear facility stack emissions are typically controlled by multiple stages of HEPA filters. The HEPA filter is designed to remove particulates from a gas stream with an efficiency of at least 99.97%. Selective penetration of HEPA filters by submicrometer size particles (d_p = 0.1 – 0.4 μm) and negligible penetration of other particles is predicted by filtration theory (Scripsick 1994). Therefore it is sometimes concluded that it is not necessary to design sampling systems for HEPA filtered stacks that take into account inertial effects in the sampling nozzle inlet and transport line. ***However, this conclusion is invalid.***

HEPA filters were available long before 1994 and the experimental data on penetration was available long before filtration theory referenced here.

2nd paragraph

In studies at the Rocky Flats Plant of particulate emissions in stack effluents, Nininger and Osborne (1992) sampled for particles downstream of HEPA filters. A laser particle counter was employed to obtain number size distribution data over the size range from submicrometer to over 10 μm (optical diameter). There was observed to be only a small fractional percentage of particle counts corresponding to diameters > 5 μm, but when these data are converted to volume distribution (hence reflecting the actual distribution of particle mass, ***and possibly radioactivity,*** *in the samples rather than the number of particles) the volume percentages corresponding to particles with diameters greater than 5 μm were quite significant (greater than 30%). The authors observed the presence of white fibers in the vent discharge and speculated that the large particles may have been shed by the HEPA filters.*

This is very interesting but rather speculative. However, even if particles as large as 5 μm are being occasionally shed by the HEPA filters this does not by itself call for isokinetic sampling as they will be sampled with only minor losses by anisokinetic samplers. The real justification for true isokinetic sampling, not the "saltation" sampling favoured in this standard, is the possibility of complete breakdown of the HEPA filters in either fire or explosion (both of which have occurred in plutonium processing facilities). In this case the subsequent size distribution of the resulting aerosol cannot be predicted precisely and may be greater than 20 μm where true isokinetic sampling is highly desirable if not essential.

3rd paragraph

A different explanation for the presence of the larger particles downstream of HEPA filters comes from recent studies by Scripsick (1994) of leak phenomena in HEPA filters systems (pinhole leaks, frame seal leaks, etc.). Particle penetration through filter perimeter seals and the filter pack were determined separately and in combination. Penetration was observed only when the challenge aerosol was introduced into the system, ruling out the possibility that the observed particles were shed from the filter pack. He found that whereas filtration theory predicts a penetration fraction of 10^{-16} for 0.7 μm particles, the observed penetration was approximately 10^{-5}. Scripsick concluded that system leakage phenomena and the size distribution of the challenge aerosol can surpass filtration theory considerations in predicting the size distribution of particles penetrating the HEPA filter systems.

The figure of 0.7 μm for the MMAD of the challenge aerosol suggests that the aerosol was 'Cold DOP' which is not a monodisperse aerosol but contains satellite droplets down to a size of 0.1 μm. This may explain part of the penetration. However, the figure of 10^{-5} for the penetration is very close to the radioiodine penetration observed on newly installed SIAM beds at Lucas Heights (May 1975) whose expected decontamination performance was greater than 10^{9}. This may in fact be the limit of performance for foam rubber seals. It still means that 99.999% of all activity is trapped by the filtration system.

4th paragraph

This is consistent with studies of multiple HEPA filter banks by Ettinger et al. (1973), whose data are summarized in table G.1. Here the presence of a small but significant fraction of supra-micrometer sized particles beyond the second and third stage is indicated because the geometric standard deviations remain large. But the respective activity concentrations are very small due to the reduced challenge and narrowing spectrum of particle size at each successive stage. At the same time, failure of earlier stages would be expected to result in both higher concentrations and larger quantities of particles in the ***inertial size range (1 μm AD and larger).*** *Therefore, the design of extractive sampling systems in HEPA filtered stacks should reflect consideration of the presence of large particles, even under the presumption of normal operating conditions and HEPA filtration.*

This does not make out the case for isokinetic sampling, which is only required for particles >20 μm. The real and unanswerable case for true isokinetic sampling, with the sampler completely within the stack, is the off-normal case of damage to the HEPA filters. Unfortunately the "saltation" samplers described in this standard do not comply with what is required.

Conclusions

Some parts of this American standard on stack sampling seem both confused to the reviewer and confusing to those seeking advice. It is not helped by several statements in the new standard that some of the advice in the previous standard (1969) was faulty and should now be discarded.

This standard should not form the basis of stack sampling at Lucas Heights for the following reasons:

- The standard considers the needs of isokinetic sampling, even possibly overstating the case for isokinetic sampling, yet the samplers described and promoted in the standard are not strictly isokinetic samplers and would not satisfactorily sample the largest particulate aerosols (>50 μm), which may be present in abnormal cases.

- The standard promotes the use of long sampling lines, which are known to cause unacceptable inlet losses of some of the radioactive vapours in many circumstances (i.e. carrier-free elemental radioiodine vapour and higher concentrations of elemental iodine vapour in moist air).

- The standard suggests that materials such as aluminium and steel are suitable materials for the composition of long sampling lines (up to 40 m) for radioiodine vapour, despite much evidence to the contrary by other workers elsewhere that only short Teflon-coated inlet nozzles can be tolerated for reliable sampling under all circumstances.

- The standard promotes the use of trace-heating of sampling lines as a cure for the difficulties of plate-out of elemental radioiodine on the internal surface of the sampling line, despite other evidence that high temperatures can cause thermal decomposition of methyl iodide and the promotion of chemical reactions between carrier-free elemental iodine and the oxide surface on the metals.

- The standard does not investigate the prudent alternative to long sampling lines, which is to place the isokinetic sampler and the radiodine sampler within the confines of the stack itself. The only sampler that the standard suggests can be placed in the stack is a continuous sampler for noble gases, which is one of the few sampler types that is better placed outside the stack and which does not suffer from unacceptable inlet losses in long sampling lines.

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Figure 1 Lucas Heights Isokinetic Sampler pre 1966