

Air Sampling Programmes for Managing Internal Exposures: Review of Key Practical Issues

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ABSTRACT

Air sampling programmes are widely used within the nuclear industry to monitor workplaces. These programmes may also be used to provide quantified estimates of individual worker intakes and doses, especially where the expected exposures are low; or to provide reassurance that potential doses to groups of workers are below predefined reference levels. These programmes, if properly implemented and managed according to an effective Quality Management system, can provide a relatively simple and cost-effective means for demonstrating the radiological protection of workers; and off-set the need for more extensive and expensive individual bioassay monitoring programmes. However, there are various practical issues which need to be considered if air sampling programmes are able to achieve these aims reliably and with effective assurance. This paper provides an overview of some of the key issues which should be considered; and encompasses issues associated with sampling, monitoring, measurement, and interpretation of results. The purpose of this paper is simply to highlight these issues for information and awareness, rather than to provide detailed analyses.

INTRODUCTION

Inhalation of radioactive aerosols is the primary pathway for incorporation of radionuclides by workers, particularly in the nuclear industry. Therefore sampling and monitoring of the air in the working environment has for a long time been used as a suitable means for assessing and controlling the risk of intakes. However, despite widespread and long established use, there are still widely different approaches to the detailed techniques, methods and quality assurance by which air sampling and monitoring programmes are applied in practice. This paper provides a review of some of the practical issues which might need to be considered when establishing and operating an air sampling and/or monitoring programme: including sampling and monitoring in the workplace, laboratory methods, interpretation of data and quality management issues. The purpose is to promote awareness of these issues, rather than exhaustive detailed analyses, such that air sampling and monitoring programmes might be better optimised in terms of quality assurance and cost-beneficial operation.

SCOPE

This paper only considers sampling and monitoring of airborne particulates; it does not consider gaseous air activity. The term 'sampling' refers to the process of collecting airborne dust samples on a filter, which is then sent to a remote laboratory for radiometric assay. The term 'monitoring' refers to the collection of airborne dust samples on a filter which are subject to radiometric assay in near real time within the sample collection unit.

SAMPLING AND MONITORING: SYSTEMS AND METHODS

Personal Air Sampler (PAS)

PAS are portable, battery-powered devices worn by a worker and used to collect a sample representative of the activity concentration in the air inhaled by the worker; as such, PAS are most commonly used as a means to provide estimates of intakes by the individual worker. They are also used as a means for detecting localised acute exposures, which then trigger further investigation: e.g. involving bioassay measurements. A sampling head containing a filter is worn on the upper torso within the breathing zone, which is normally assumed to be within 30cm of nose and mouth. Ideally sampling rates should be the same as typical breathing rates for a worker ($\sim 1.2 \text{ m}^3/\text{h}$), but current devices often provide only about one tenth of this value. PAS devices with higher flow rates are available; however, this needs to

be balanced against the ability of the device to maintain stable flowrates for continuous operation over the whole of the wear period, which is typically one working shift.

PAS might also be fitted with particle size-selective sampling heads, such that particles of different aerodynamic diameter are deposited on discrete regions of the collection filter and allow for some degree of particle size analysis ^[1].

Experiments with a dense monodisperse aerosol and an array of low flow-rate samplers ^[2] have shown that differences in sampled mass can vary by factors of around 3, even over distances less than 30 cm. This is likely to be the result of spatial variations in the bulk transport of aerosol arising from turbulent air flow. In addition, at the low particle densities typical of many nuclear aerosols, sampling statistics can lead to wide variations in sampled activity ^[3].

Static Air Sampler (SAS) or Workplace Sampler

SAS are commonly used to monitor general workplace conditions, to provide assurance of effective control, and to detect any deterioration in conditions. If SAS are to be used to provide information on potential worker intakes, then ideally they should be located between potential release points and likely exposure points, or occupied areas. However, in practice this can be very difficult to accomplish due to often highly complex and variable patterns of air flows within an area ^[4]. These air flows can be significantly influenced by many factors: building ventilation, doors and windows, ongoing operations, worker occupancy. Various means have been employed to attempt to 'map' airflows in a facility, with smoke tests being the more typical, although more advanced methods have been attempted ^[5]. However, it is advised that none of these techniques can be considered sufficiently reliable, and in all but the simplest release-exposure scenarios, there is little option but to consider multiple sampling points. The number of sampling points required will be dependent on the assessed complexity of potential release points, occupied areas and potentials for variable flows between the two. The location and number of sampling points should be subject to regular routine reviews.

Partly as a result of the problems of siting SAS, it is known from experience that SAS measurements can significantly underestimate maximum airborne activity concentrations in the workplace; in extreme cases underestimations can sometimes be of several orders of magnitude ^{[4] [5] [6] [7]}. This highlights the importance of exercising caution when using SAS measurements to infer worker intakes.

Sampling periods will need to be considered, which will be determined by various factors:

- expected magnitude of air activity
- expected risks for acute exposures or releases
- ease of access to the sampler to affect a filter change
- general environmental conditions: dusty workplaces would require high frequency filter changes to avoid accumulation of dust on the filter
- costs
- capacity of the analysis laboratory to process the required throughput

SAS devices can also provide useful information on radionuclide composition by spectroscopic analyses of the filter, and on particle size if used with a size analyser such as a cascade impactor ^[8].

Continuous Air Monitors (CAM)

CAMs are essentially an enhanced version of SAS which incorporate a detector facing the collection filter, connected to a real time activity monitor and alarm unit. The primary function is to provide a real time response to detect unexpected airborne releases, which would prompt evacuation of the area and remedial actions. These units might also be used like SAS for assessing chronic exposure levels within a workplace; however, it should be noted that alarm thresholds are typically set to detect acute

events, and that these thresholds might not be appropriate for monitoring chronic levels of air activity to sufficiently low levels, particularly for alpha activity. In this case it would be appropriate to treat CAMs as SAS and have filter samples periodically removed for more sensitive radiometric analysis.

The accumulation of radon daughter radionuclides on the filter might present a problem by causing false alarms. Detection systems will typically incorporate some form of compensation algorithm applied to the detected alpha energy spectra – radon daughters typically having higher alpha energies than, for example, actinides. However, the detected alpha energy spectra can be significantly influenced by a variety of variable factors – e.g. dust accumulation, humidity. Therefore, even radon-compensated devices cannot be considered to be completely reliable, nor are they immune to false alarms.

Some models of CAM are available with a moving filter strip, rather than single filters. These units can be set-up to automatically move the filter media incrementally after a pre-set period of time to minimise the dust accumulation of the sampling and monitored part of the filter media. The configuration of such devices will need to be customised to the specific environmental conditions in which they are installed.

The optimum siting of CAMs might be different from the optimum siting of SAS, depending on the designed purpose of the respective sampling and monitoring programmes: e.g. a CAM might be intended to provide the earliest feasible alert of a release, whereas a SAS might be intended to provide the best estimation of potential worker intakes ^[4].

Sample Collection and Filter Media

Various filter media are available ^{[4] [7] [9]}, the most commonly used for PAS, SAS and CAM being glass-fibre media. The primary considerations for choice of an appropriate medium are:

- Collection efficiency: this should be greater than 95% for the aerosols of interest, otherwise specific correction factors will be required to be evaluated and validated.
- Low pressure drop across the filter: if the pressure drop is too high then this could place excessive demands on the sampling pump, especially for battery-powered pumps, as are used for PAS. It could also incur excessive uncertainties for the sample collection flow-rates.
- General environmental conditions: e.g. high relative humidity could affect the performance and robustness of some filter media.

Typically, the exposed face of the sample collection medium should sample the air in the workplace, and in the vertical plane to avoid the effects of gravitational settling of larger aerosols. Where aerosol size-selective attachments (e.g. impactors) are employed then particle loss-rates within the sample head would need to be established.

SAMPLE MEASUREMENT & RECORDING

This section refers to the measurement systems and methods that are used for radiometric analyses of the sample collection filters separately from the sampling systems. These measurement systems should be located and operated within a laboratory designed for this purpose ^[10].

Measurement Systems

Various measurement systems are available; for the measurement of alpha and beta activity the most common systems use either proportional counter detectors or solid state (silicon) detectors. When only a low throughput of samples is required then simple single-detector manual counters might be employed; for higher levels of throughput then multiple-detector arrays and/or automated sample-changer counting systems are normally used.

System Performance & Quality Control

Measurement systems are required to be operated according to a clearly defined Quality Assurance programme to assure reliability of performance and output ^[10]. An effective quality assurance programme would include the following technical features ^[11].

Type Test: this defines the characteristics and expected performance of a system to enable the most appropriate choice of system for a particular application.

Test before First Use: this test provides assurance that the performance of a particular system conforms to the specified Type Test specification; this enables any defects or non-conformities to be identified and addressed before bringing the system into use.

Periodic Tests: this is a programme of routine periodic tests established to provide continuing assurance that the system is still performing according to the Type Test specification. These tests consider various factors as well as detector performance: e.g. mechanical reliability; software; data management; reliability of output reports.

Function check: this is a minimal check to provide assurance that a system is still functional; for air sample counters this check will normally comprise the measurement of a radioactive standard source and a background measurement.

System Calibration & Characterisation

There is no ‘natural reference matrix’ available for performing direct calibration measurements: i.e. there is no calibration source which exactly mimics the physical nature of the sample, and which contains a known and traceable radioactive content. Therefore, all calibration measurements are, by nature, indirect approximations derived from the system’s response to an analogous ‘calibration standard’. This should be borne in mind, and can cause significant uncertainties in certain circumstances: e.g. a calibration standard which is constructed of a ⁹⁰Sr source plated onto an aluminium substrate will give rise to enhanced beta emission rates in the forward plane due to back-scattered beta particles from the substrate; this would not be replicated by ⁹⁰Sr activity collected onto a glass-fibre filter.

In practice the term ‘calibration’ is often presumed to mean subtly different processes and objectives: e.g. it might refer to the initial type testing, periodic tests and/or daily function checks. The authors suggest that the term ‘calibration’ should refer to the process for determining how the system performs according to its defined Type Test specification ^[10]. The result will be the calculation of a correction factor(s) which is to be applied to the measured quantity to calculate the required output quantity: e.g. for the conversion of measured counts to activity. In practice for operational purposes this ‘output quantity’ will need to be expressed as various ‘derived quantities’. Therefore the ‘derived quantities’ of interest – e.g. air activity concentration – will need to be ‘characterized’ by a process which considers all appropriate issues relating to measurement of the sample, and factors pertaining to the sampling programme – e.g. dilution factors. A system can be simply checked to see if it is still performing according to its calibration by periodic quality control checks, typically by measurement of a traceable reference standard ^[10] ^[11]. Systematic reviews of the system’s characterization are usually not so straightforward and, typically, are not performed on a routine basis. However, it would be advisable for the QA programme to consider the processes for how significant changes to general operating conditions might be detected: e.g. significant changes to patterns and levels of air activity concentration, exposures to different radionuclides. This could then initiate a review of the overall characterisation of the system.

Sample Measurement

A variety of factors can affect the reliability of the sample measurement:

Particulate radon-daughters collected on the sample: this can be mitigated by using radiometric compensation methods or by delaying measurement for at least five days to allow for radioactive decay.

Radon gas within the laboratory: specific environmental controls (e.g. effective ventilation) might be required.

Differential energy response by detector: this might be an issue where the sampled radionuclides have significantly different radiation emission energies to the radiation standard source as used for calibration; correction factors might need to be considered, or the calibration and characterisation processes might need to be reviewed.

Radiation emission characteristics: factors that will need to be considered are the number and/or probability of emission of particles, and also the presence of short-lived daughter nuclides: e.g. ^{90}Sr / ^{90}Y .

Differential media substrates: radioactive standard sources are typically produced onto a metal substrate, as opposed to the glass-fibre media typically used for sample collection; this can be a significant factor for calibrating beta response due to the back-scatter of beta particles from the metal substrate, which isn't replicated for beta sample measurements.

Calibration source construction: if the radioactive element of a radiation standard source is too deep within the source then there is a risk of self-collimation of emitted particles; this might provide a forward bias for particle emissions which won't be replicated for sample measurements, where emissions are more likely to be semi-(2- π)-isotropic.

Differential edge effects: radioactive standard sources are typically constructed to have a homogenous distribution of the activity over most of the surface area; sampled particulates will likely have discrete deposition patterns. This issue can be overcome by employing detectors which have a surface area greater than that of both sources and samples; otherwise this factor might need to be evaluated as part of uncertainty estimations.

Self-absorption: it is feasible that sampled aerosol particulates might penetrate into the filter media, or be obscured by later accumulations of particulates. This is not generally considered a significant issue in practice for environments with low dust loading in the workplace air; however, this will need to be monitored: e.g. by periodic inter-comparisons involving full-destructive assay of samples by radio-chemistry techniques. A study was conducted at the Harwell and Dounreay sites as part of the Nuvia Dosimetry Services QA programme. This included 15 air samples (14 SAS and 1 PAS) which were exposed as part of routine sampling programmes in different facilities, at different times. In all cases the primary radionuclides were expected to be plutonium and americium isotopes. The samples were subject to the standard radiometric measurement and then sent for radiochemical analysis. The mean ratio for radiochemical to radiometric results was 1.22 ± 0.69 (1 std dev.)^[12], which provided an indication that self-absorption is not a significant problem.

Background corrections: all detector systems will be subject to 'background'. In practice it can be difficult to ascertain the source of this 'background' and it is normally just considered to be undefined 'noise'.

Sampling handling: sample measurements for alpha-emitting radionuclides are especially sensitive to careful handling, due to the low detection levels which are typically required. In addition to normal sampling handling requirements the laboratory should also be aware that samples collected on glass-fibre filters might be sensitive to risks of exposure to static electricity (e.g. from use of polythene bags).

NORM within filter media constructs: Some treatments of cards, particularly glazes, can include traces of naturally occurring radioactive material.

Statistical Analyses of Samples with Very Low Count Rates

When the sampled activity is very low, the count rates measured from PAS or SAS filters approximate to background counts. These counts follow a Poisson distribution ^[13]. This can cause problems when a background count is subtracted from the sample count. In such a case the mean of a Poisson distribution is being subtracted from members of another distribution with nearly identical mean. Since the mean of a Poisson distribution does not coincide with the median, the effect of this can be that a preponderance of negative net counts can result. This can lead to the mistaken impression that a bias exists in the background subtraction. It becomes important to ensure that the background subtraction is truly representative and that allowance is made for subtle variations in background count throughout the year. Consideration is also needed as to whether it is the mean or median of the background distribution which should be subtracted; this will be influenced by the primary purpose to the sampling – to measure individual exposure events, or cumulative exposure conditions: this decision might have a significant impact on the reported numerical values of the results.

Quantities and Units

In practice there are a variety of quantities used, and sometimes for different purposes: this is illustrated in Table 1.

Table 1: quantities and units typically used in air sampling programmes

Quantity	Units	Applied correction factors	Typical Application(s)
Count rate (measured quantity)	Cps, cpm, dpm, dps	Background subtraction	Type test, periodic tests, function test
Activity (output quantity)	Bq	Calibration factor	Activity on sample; Routine Lab reports and records
Airborne concentration (derived quantity)	Bq.m ⁻³	Characterisation factors(s) related to sampling process (eg flow rates and filters)	Health physics reports; workplace reports
Time integrated airborne concentrations (derived quantity)	Bq.h.m ⁻³ Bq.s.m ⁻³	Characterisation factors(s) related to sampling programme (e.g. coverage, sample periods, dilution factors)	Potential worker intakes
Intake rate normalised by dose coefficients (derived quantity)	DAC	Worker breathing rates, Exposure time, dose coefficients	Internal dose <i>rate</i> estimates
Total intake normalised by dose coefficients (derived quantity)	DAC- hours; DACH	Worker breathing rates, dose coefficients, default working year (hours)	<i>Cumulative</i> internal dose estimates

Uncertainty, Detection Levels, Decision Thresholds and Censored Data

The method for the determination of uncertainties in measurements is well described ^[14]. A thorough determination would require the construction of a mathematical model that represents all of the physical process which contribute a source of uncertainty to the final measured outcome. This would need to account for all of the effects noted in the section on **Sample Measurement**; in practice this is never attempted. It is more common to limit the quantified estimation of uncertainty to a few parameters – typically the observed count-rates from the sample, background and calibration reference standard. It can be seen from the discussions presented in the preceding sections that this approach, albeit a necessary pragmatic shortcut, can only provide a partial and qualitative understanding of the overall uncertainty in the process.

The standardised definitions and uses of the terms ‘Detection Limit’ and ‘Decision Threshold’ are generally accepted ^[15]. For simplicity the ‘Detection Limit’ can be summarised as the expectation of the lowest value that can be measured with a defined degree of confidence: i.e. this determination is made *before* the measurement is made. This parameter is particularly useful in planning measurement equipment, methods and procedures: e.g. the identification of appropriate equipment, sample measurement periods, tolerance levels for background rates etc. The ‘Decision Threshold’ is the lowest value of a measurement that can be claimed to be a genuine (non-background) result with a defined level of confidence. As this value is intrinsically dependent on the actual value of a measurement it can only be determined after the measured value is known. It is typical that Detection Levels and Decision Thresholds are applied to the output quantity (sample activity) or to the measured quantity (e.g. count rates). Caution should be used when using these terms applied to any derived quantity, as their determination will then need to include uncertainty estimates from all of the corrections and assumptions used in the derivation process.

Typically, standard reports and outputs will use ‘censored’ data: e.g. this is data that, if the measured value is less than the Decision Threshold, then the value is reported as a ‘less than’ value. However, there are certain applications which will require the actual measured value to be reported, with the associated measurement uncertainties: i.e. ‘uncensored’ data. Such reports might be required for further data analysis, particularly on bulk data: e.g. to determine mean activity levels over a period of time or a large number of samples, in which case the use of censored data would lead to biased analyses.

INTERPRETATION OF DATA AND ESTIMATING INTAKES

It is sometimes considered appropriate to use SAS measurements to estimate intakes for workers when expected doses are either low, or for confirmation that workplace conditions do not require individual monitoring programmes ^[16]. This is appropriate when air activity characteristics are well described and are reasonably stable; and is most often applicable for large scale processes involving low-specific activity nuclides: e.g. U, NORM. The SAS data is usually either expressed as average air activity concentrations ($\text{Bq}\cdot\text{m}^{-3}$) which need to be modified by presumed default breathing rates and dose coefficients to derive intakes; or as time integrated exposures (DAC-hours) which need to be modified by a presumed default working year (usually 2000 hours) and by the respective Annual Limit on Intake (ALI) for the nuclides of concern. In both cases worker occupancy data needs to be recorded so that estimated intakes are proportionate to the actual time spent in the workplace. The estimated intakes will need to account for potential underestimations of the SAS measurement by the application of correction factors (sometimes known as Dilution Factors, Breathing Zone Factors or PAS Factors).

PAS are more often used for assessing worker intakes. The results of the PAS are assumed to be directly related to the worker’s intake, modified by the ratio of sample flow-rate and presumed default breathing rate. A review of assessed intakes of plutonium and higher actinides at Harwell was carried out ^[17]. This enabled a comparison to be made between intakes assessed from bioassay sampling with initial estimates based on PAS activities. The correlation between the two estimates of intake was found to be close to zero. A similar result was found for acute and chronic intakes for individuals at Sellafield ^[18]. However, the same authors state that there is better agreement when groups of workers are considered. A survey of activities in routine faecal samples for groups of workers at Harwell ^[19] showed a clear correlation with average PAS activities for the group. There are various other studies which have attempted to compare intake estimates by PAS and bioassay measurements; however, the authors are not aware of any published studies which have attempted a rigorous (if any) uncertainty analysis for both techniques, although both techniques will be subject to substantial, but different, sources of uncertainty. Therefore any attempt to draw meaningful conclusions would seem to be of dubious value.

It is noted that there is a marked difference between sampling for plutonium and sampling for uranium. 1 DACH of pure Pu239, with an AMAD of 5 microns, corresponds to about 250 particles. Sampling statistics therefore place severe constraints on the accuracy of air sampling. However, 1 DACH of natural uranium aerosol with the same size distribution corresponds to $2.8\text{E}6$ particles and sampling statistics are likely to favour more accurate estimates of intake. Air sampling data at a plant where

uranium mining concentrate is converted to UF₆ has been found to correlate with uptakes calculated from urine analyses ^[20].

Analysis of bulked PAS data for a group of workers might be useful to provide further indications of the potential probability distributions of worker intakes.

Despite the outstanding questions regarding the correlation between individual air sampling results and bioassay, air sampling still has an important role to play as a trigger for undertaking dose assessments. A preliminary survey of Harwell data ^[21] shows that about 25% of 81 investigations into potential acute exposures, where an acute intake was confirmed by positive follow-up bioassay measurements, were triggered by a high air sampling result (either PAS or SAS); this compares to about 50% detected by routine bioassay programmes (either urine or faeces). However, the air sampling measurements have a lower 'false trigger' rate than the bioassay programmes: i.e. an air sample trigger is more often confirmed as an intake than an investigation triggered by routine bioassay (53% compared to 29%, respectively). These analyses will be subject to further study.

OPTIMISATION: CO-ORDINATED STRATEGIES, QA & COSTS

Air sampling is not generally recommended as a means for assessing internal doses for operations where internal doses greater than 6 mSv per year are likely ^[16]; in such circumstances routine bioassay programmes should be considered. Where less significant doses are likely - typically in the range 1 mSv to 6 mSv per year - then air sampling might be used as the primary means for the routine assessment of intakes. However, the impact of the uncertainties implicit in air sampling programmes should be noted: these uncertainties can be significant but are often unknown and unknowable. Therefore it is general practice that air sampling, when used for this purpose, is supported by a 'reassurance' or 'confirmatory' bioassay programme ^{[22] [23]}. Such programmes need not be as extensive or frequent as those used to directly assess routine doses. The basic strategy within Nuvia Approved Dosimetry Services is to recommend 'reassurance' bioassay programmes designed to detect doses greater than 6 mSv per year. The inference from this strategy is that we claim that the 'reassurance' bioassay programmes provide confidence that doses are less than 6 mSv per year, and that the air sample programme (usually PAS) provides the best point estimate of the dose.

For operations where significant intakes are not expected – i.e. when prior risk assessments have deemed that routine intake and dose estimates are not required - then carefully designed air sampling programmes are an effective means for providing measurements to check and validate this risk assessment. However, the complexities and uncertainties implicit in the relationship between air sample measurements and worker intakes means that intakes cannot be directly quantified with any reliability. There are various approaches which might be considered to address this issue.

- (a) *extended characterisation of air sampling uncertainties*: as discussed previously this is not a trivial undertaking; however, it might be cost-beneficial for large-scale processes, where operations are reasonably routine and uniform, and potential release-exposure scenarios are fairly simple and of limited scope. This might be an especially cost-effective solution where large numbers of workers are involved.
- (b) *supporting 'reassurance' or 'confirmatory' bioassay*: this avoids the need for extensive characterisations; and might be more cost-effective for smaller work groups, and where exposure-release scenarios can be more varied.
- (c) *task specific or campaign monitoring*: a limited bioassay study might be conducted to assess the potential intakes arising from a specific process ^{[16] [22]}. For example, at the start of a new process a limited study, involving bioassay and air sampling, can establish a partial characterisation of the air sample programme – e.g. to derive Investigation Levels. The air sampling programme then continues to monitor for any significant change in conditions; if a change is detected then the study is repeated and reviewed.

Other issues which will affect how costs are optimised also include:

- existing and available facilities, or the need to set-up from new: e.g. a bioassay laboratory will be substantially more expensive to establish from new; an extended air sampling programme will have significant costs to purchase the sampling and monitoring devices;
- technical capability of techniques & facilities for the expected exposure hazards; and also the availability and skills of staff
- knowledge (or ability to gain knowledge) of air activity characteristics: this is likely to change with time, so periodic reviews would be undertaken to see if the overall strategy needs to 'evolve';
- assessed risks of chronic and acute exposures: the assessed risks will also change with time and experience, so should also be subject to periodic reviews
- nuclide mixes (air sampling is a gross measure, whereas bioassay is normally nuclide-specific)

CONCLUSIONS

It's complicated! However, it is believed that an objective awareness of the issues associated with air sampling can lead to substantial opportunities for optimising both the effective radiological protection of the workforce, and the costs of running monitoring programmes.

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Forthcoming reference material (in preparation)

ISO 16639: Surveillance of the activity concentrations of airborne radioactive substances in the workplace of nuclear facilities

European Commission: Technical Recommendations for Monitoring Individuals for Occupational Intakes of Radionuclides