Annex 3, Appendix 1

Sampling for particulate airborne contaminants

Review and analysis of techniques

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1. BACKGROUND

While for years now, many efforts have been made in optimisation to keep external radiation exposures as low as reasonably achievable (ALARA), very few efforts have been devoted to put into practice the ALARA approach for internal exposures. However, in some workplaces, the most significant exposure pathway is the internal exposure via inhalation of particulate airborne contaminants. In particular, this can be the case for the industries involved with naturally occurring radioactive materials (NORM) or for the nuclear industries. A rough estimate for the total number of workers potentially exposed to internal radiation in the EU lies in the range 5000 to 10000 persons (van der Steen *et al.,* 2002). For those persons, internal exposures situations differ considerably with respect to workplaces conditions and particulate airborne contaminants characteristics (referred as aerosols to hereafter). One way for assessing the effective dose resulting from the worker's inhalation of airborne radionuclides is to use aerosol sampling results, including those of the particle size distribution and particle concentration. This issue has been highlighted with the publication of the Council Directive 96/29/Euratom (1996).

In 1996 the European Commission created a European ALARA Network (EAN), to further promote European research on topics dealing with optimisation of all types of occupational exposure, as well as to facilitate the dissemination of good ALARA practices within all sectors of the European industry and research. The EAN organized at Neuherberg in November 1999 a workshop on "Managing Internal Exposure" from which the third following recommendation to the European Commission has been made (Lefaure *et al., 2000*): *"…there is a need to pursue efforts to improve the quality and accuracy of internal dose monitoring techniques (particularly personal air sampler) to fit with the specifications needed for analytical task dosimetry. The meeting recommend to the Commission and regulatory bodies, that they support research in that area."* As a result, and part of the 5th Framework Programme, the European Commission (D.-G. Research), ordered under contract n° FIGM-CT2001-00076 the project entitled SMOPIE to start in November 2001. The final objective of SMOPIE (Strategies and Methods for Optimisation of Internal Exposures of workers) is to recommend monitoring strategies and methods for optimising internal exposure in a wide range of situations of predictable occupational exposures (van der Steen *et al.,* 2002).

One of the work packages of the SMOPIE project is devoted to the evaluation of monitoring strategies, methods and tools (WP4) with the objective to critically review potentially useful monitoring strategies and methods and associated analytical tools. The contractor CEPN (Centre d'Etude sur l'Evaluation de la Protection dans la domaine du Nucléaire) asked to the IRSN (Institut de Radioprotection et de Sûreté Nucléaire) to be sub-contractor as being recognized to have an expertise in monitoring devices used for sampling particulate airborne

contaminants. The review defined in the WP 4 is first based on the state of art from the relevant literature, and second on specific laboratory or field tests that would be devoted to the evaluation of sampling performances of selected devices.

The present document exposes the first part of the defined work: the review, from the appropriate literature, of the monitoring devices and methods to be used in aerosol sampling studies in workplaces for exposure assessment.

2. INTRODUCTION

Particulate airborne contaminants in workplaces are the sources of a high proportion of potential occupational illness. In particular, occupational lung disease is associated most with worker exposure to aerosols in the form of dusts, fumes, mists, and smokes. The respiratory tract is also an important route for particulate radionuclides to enter the human body. Inhalation of radioactive aerosols poses a potential health hazard to workers in the nuclear industry and other industries involved with naturally occurring radioactive materials at large. For this reason, the worker monitoring of exposure to intakes via inhalation of radionuclides is a subject of considerable interest. One of the ways for the estimation of the committed effective dose is to make measurements of the characteristics of the inhaled radioactive aerosols (concentration and particle size distribution), and to use these results combined with calculations using a respiratory tract deposition-retention-dosimetric model for radioactive substances like the one's proposed in the ICRP publication 66 (1994) or more recently proposed by the NRCP (1997). As a consequence, sampling of radioactive aerosols for the purpose of predicting or assessing radiation doses now becomes an important issue in radioactivity-related occupational hygiene. In particular, in the European countries, the issue has been recently brought to the nuclear fuel handling industry's attention with the publication of the Council Directive 96/29/Euratom (1996). In particular, it is specified in chapter II (article 24 and 25) that measurement results from aerosol sampling can be used for assessing the individual dose when the individual biological monitoring is not possible or gives insufficient results. To assess the effective dose resulting from the worker's inhalation of airborne radionuclides by aerosol sampling, two types of result are needed: the particle activity concentration and the particle size distribution.

Sampling and measurement of radioactive aerosols mostly involves the *traditional* aerosol instrumentation used in the aerosol sampling studies in the workplace. Therefore, many of the information given in thisdocument comes from the industrial hygiene literature. The advantage of the unique radioactive property of radioactive aerosols that makes them essentially easier to detect once sampled and subsequently collected on medium is only reviewed in very few documents.

This document is presented in three chapters. The first sets out the basic sampling philosophy and objectives. The second chapter exposes the current status of practical sampling instrumentation for the measurement at workplaces. The third and last chapter provides a quick overview on analytical considerations that are specific to the measurement of radioactive aerosols.

3. SAMPLING AGAINST EXPOSURE

In the nuclear sector, the use of the aerosol sampling as a method for internal dose (via inhalation of radioactive particles) assessment has been debated for many years (Britcher and Strong, 1994). It is clear now that for insoluble particles that are retained in the human body, the aerosol sampling method could be a much more adequate way for operational dosimetry than in vivo and/or bioassay methods. In particular, it has been recently shown that the limit of detection of bioassay methods are very high resulting in doses comparable to the annual dose limit (Degrange *et al.,* 1999), and, in comparison, that traditional aerosol sampling methods may lead to lower limits of detection in term of dose.

The first part of the overall process of aerosol exposure is the entry by inhalation of particles from ambient air into the respiratory tract. Once inhaled, aerosols are fractionated during penetration through the airways, and the particles deposited at different levels can cause various health effects, which depend on their (radio) toxicological properties and on their deposition site (Fabriès, 1992). In consequence, health-related aerosol sampling criteria should first reflect the aerodynamic process by which particles initially enter the body during the act of breathing (through the nose and/or the mouth), and by which they are subsequently deposited in the various part of the respiratory tract. An ideal aerosol sampler should follow these sampling criteria. However, in practical, each sampler has its own behavior with regards to many factors. Thus, it is really important to evaluate the deviations by making specific experimental tests.

in industrial hygiene the primary component to assess is the worker exposure to aerosols. As it will be shown latter in chapter 3.1.2, the situation is somewhat different in the nuclear sector and NORM industries as the primary component to assess is the effective dose. The assessment combines measurement results and calculations using a respiratory tract deposition-retention-dosimetric model like the one's proposed in the ICRP publication 66 (1994) or by the NRCP (1997). In particular, these two models require for the calculation of the suitable dose coefficient, the aerosol characteristics of the ambient aerosol.

That means that, if one wants to use directly its results to estimate effective doses, an ideal particulate sampler should follow the 100% sampling efficiency criteria. In practical, there is no sampler with such a performance. Thus, in the radiation protection dosimetry context, it is also extremely important to evaluate the deviations in term of sampling performance against the 100% sampling criteria.

3.1. Particle size selective sampling

The sampling method to use for aerosol measurement should be based on criteria, which relate to the reason for which the measurement was initially considered necessary. Since, the aim is the evaluation of the aerosol intake for potential health-related risk assessment, the measurement criteria should be based on consideration relative to exposure and dose. In this chapter, are presented the different existing sampling criteria for workplace and environmental sampling that have been scientifically discussed for several years, later accepted and recently standardized to be applied in the generic industrial hygiene world. However, the particular situation for the nuclear sector, which makes some significative differences is also presented.

3.1.1. Criteria for workplace sampling

For many years, discussions addressed the question of what should be the basis of health-related exposure assessment and, in turn, aerosol sampling standards. These discussions were based on experimental measurements of the aspiration of aerosols (inhalation) from the ambient air into the top of the respiratory tract (mouth or nose), and of respiratory tract deposition. If the first measurements (for the inhalation) were conducted with the use of (rotating) mannequins, the second measurements used several approaches with human volunteers or laboratory model simulations. As a result, an international agreement between CEN [Comité Européen de Normalisation, CEN (1993)], ISO [International Organization for Standardization, ISO(1995)] and ACGIH [American Conference of Governmental Industrial Hygienists, ACGIH (1996)] has been achieved on a common set of particle size-selective aerosol sampling criteria. These specify that health-related sampling should be based on one or more of the three, progressively finer, particle size-selective fractions: inhalable (the aerosol fraction which enters the nose and/or the mouth during breathing), thoracic (the sub fraction of inhalable aerosol which penetrates into the respiratory tract below the larynx and respirable (the sub fraction of inhalable aerosol that penetrates down to the alveolar region of the lung). These fractions are expressed as curves, which relate the probability of inhalation, or of penetration to the thoracic or alveolar regions, as functions of particle aerodynamic diameter. The particle-size dependent curves are plotted in percentage in Figure 1. The choice of the aerosol fraction to be measured in a specific workplace depends on regional aerosol toxicity. For some type of aerosols, particles constitute a risk to health regardless of where they are deposited in the respiratory tract, like for the lead or cadmium which are highly soluble. For the health-related measurement of aerosols containing such toxic particles, it is then appropriate, and widely

accepted in the generic industrial hygiene context, to sample according to the inhalable convention.

The fate of inhaled particles, once deposited at their initial site of deposition in the lung, includes different complex processes like clearance, dissolution, re-distribution, retention… For example, three modes of clearance having different time constants have been defined that correspond to different compartments in the lung: the fast-clearing mode, the medium-clearing mode, and the slow-clearing mode. The kinetics governing the effects of the deposited particles depend of the structures the particle interacts with at the site of deposition within the respiratory tract, and obviously of the particle size, shape, solubility, surface chemistry… For the radioactive particles, the potential health effect will depend on whether the particle is deposited in the deep lung (alveolar region) or in the periphery of the lung (extrathoracic) and whether it is insoluble or not.

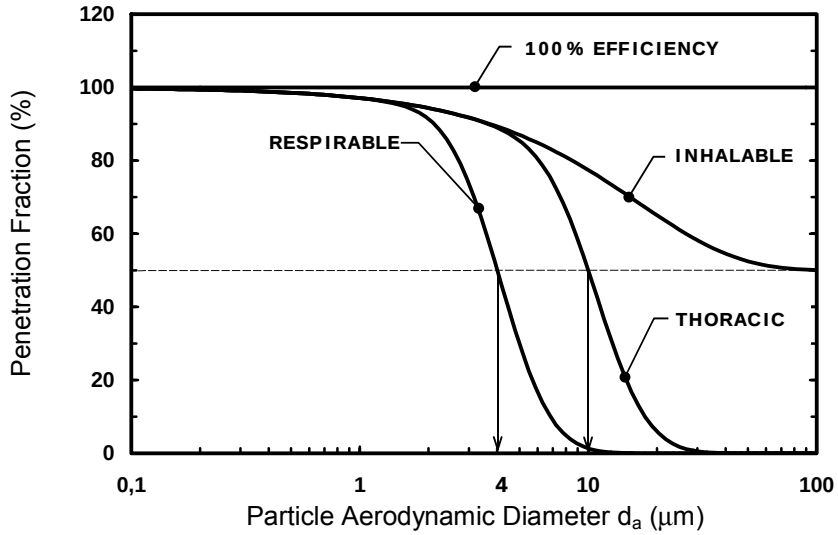

Figure 1 : Particle size fractions (i.e. inhalable, thoracic, respirable) for health-related sampling in workplaces that have been internationally agreed by CEN, ISO and ACGIH.

The inhalable conventional fraction is described by the following expression:

$$I(da) = 0.5 \times \left[1 + \exp\left(-0.06da\right)\right]$$

where the aerodynamic diameter *da* is expressed in µm. This expression is valid for particle diameters up to 100 µm and for air velocities between 0.5 and 4 m/s. This inhalable convention

assumes that all orientations of the worker with respect to the wind direction are equally represented and that the aerosol source is remote and the cloud uniform.

To illustrate the implication of the different fractions in the particle size distribution of an aerosol, a calculation has been made, with the results shown in Figure 2. An ambient aerosol with an activity concentration A = 1 Bq/m^3 is characterized by a lognormal distribution with an activity median aerodynamic diameter AMAD = 10 μm and a geometric standard deviation GSD = 2. Based on the particle size distribution and the three conventional curves in Figure 1 the particle size distributions of the three sampled fractions of this aerosol are calculated, and their characteristics determined. It is shown that the inhalable aerosol is slightly finer than the ambient aerosol with an AMAD = 9.1 μm and an activity concentration of 0.76 Bq/m^3, which is –24% compared to the ambient aerosol. The more it penetrates in the respiratory tract, the finer is the aerosol. At the end, in the illustration, the respirable aerosol is characterized by an AMAD = 3.9 μm and an activity concentration of 0.13 Bq/m^3, which is –87% compared to the ambient aerosol. This in turn means that if a sampler has a sampling efficiency which carefully follows the conventional curve corresponding to, for example, the inhalable fraction, and if this sampler is used for sampling in an ambient aerosol characterized by an AMAD = 10 μm and a GSD = 2, the activity concentration calculated from its measurement would be equal to 0.76 Bq/m^3. If it was thought that this sampler measures well the ambient aerosol, the bias (relative error) in the concentration measurement would be of –24 %! In order to be used in a dosimetric estimate, the result of activity concentration measurements following ideally the inhalable, thoracic and respirable conventionnal curves should be thus respectively corrected by a factor of 1.3, 2.1 and 7.7 (1/0.76, 1/0.47 and 1/0.13).

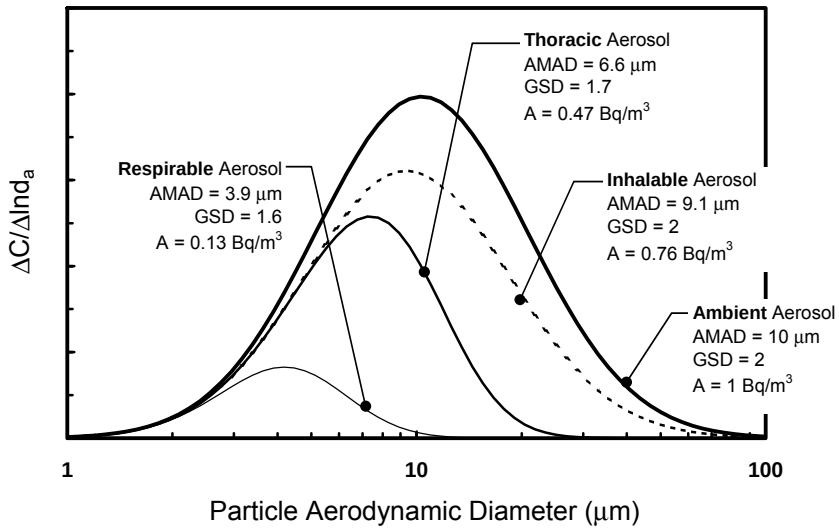

Figure 2: Particle size distributions and normalized concentrations for the ambient aerosol and the three conventional fractions (inhalable, thoracic and respirable). Ambient aerosol: activity median aerodynamic diameter AMAD = 10 μm, geometric standard deviation GSD = 2, activity concentration A = 1 Bq/m3.

These curves should be used as “yardsticks” for the sampling performance characteristics of aerosol samplers. That is, the sampling efficiency curve of any “ideal” aerosol sampling instrument should follow with no deviation the corresponding aerosol fraction. This has implications first on the performance evaluation of samplers, in particular for the “old” samplers that have been used (and still used and marketed in some cases). Also, these conventional curves are important for the new development of aerosol samplers. It should be noted here that in the industrial hygienists context, the threshold limit values (TLVs) for chemical substances refer to airborne concentrations of substances within a given size-fraction (inhalable, thoracic or alveolar). For example, for crystalline silica, the particle size selective TLV is based on the respirable mass concentration in recognition of the well-established association between silicosis and respirable mass concentrations. Obviously, in the radiation protection dosimetry context the philosophy is not the same but as long as there are no marketed samplers (and especially no personal samplers) dedicated to the measurement of radioactive particles only, it is thus better to use what has been already done and will be developed in the close future for the industrial hygiene purpose, and therefore to profit by the existing knowledge in that field, for the development of a sampler with very good sampling performances is very costly, even if the shape is simple.

It is important to note here that the conventional curves in Figure 1 are the latest, and that the evolution has led to some confusion on terminology, and now the general agreement is to use the three terms inhalable, thoracic and respirable to name the above fractions,. Moreover, the adoption of the new sampling criteria replace the old “total” aerosol that was previously used. By definition, the “total” aerosol would be the *true total aerosol (also referred as to ambient aerosol in Figure 2 and hereafter)*, i.e. the aerosol with all particle sizes. It should be known that several aerosol samplers have been sold commercially (and are still sold at this time) with this designation, but without any regard to specific appropriateness to the *true total aerosol.*

3.1.2. The radiation dosimetry context

The radiation dosimetry context differs from the generic industrial hygiene context for two reasons. First, due to the description of the model proposed by the ICRP publication 66 (1994) , and secondly due to the final information targeted: the committed effective dose.

The human respiratory tract model for radiological protection proposed by the ICRP publication 66 (1994) already includes, as a first step, the inhalation of particles, as well as the transport and deposition processes in the following stages of the pulmonary tract . It means that the entry parameters of the model regarding the aerosol characteristics necessary to evaluate the suitable dose factors must be the aerosol characteristics of the true total (or ambient) aerosol. **Thus, in turn, to use this model, it is desired to sample the *true total (ambient) aerosol*, i.e. particles of all sizes with 100% efficiency or to correct for the sampling efficiency of the aerosol sampler if it differs from 100%!**

As the reader will notice in the paragraph 3.2, each aerosol sampler has its own sampling performance, which most of the time, is strongly dependent of the particle size, as well as other external parameters like the wind velocity, etc. That means that an ideal aerosol sampler that samples the ambient aerosol with a sampling efficiency equal to 100%, whatever the particle size is, does not exist. Hence, for a given ambient aerosol, an R_X *factor* can be defined which relates the concentration measured by the sampler C_X to the concentration of the ambient aerosol $C_{AMBIENT}$ as below:

$$C_{AMBIENT}(AMAD, GSD) = R_X(AMAD, GSD) \times C_X(AMAD, GSD)$$

This R_X *factor* is function of the sampler type (X = inhalable, thoracic or respirable) and of the particle size distribution of the ambient aerosol.

To illustrate the implication of the importance of knowing the aerosol sampler performance *and* the particle size distribution, calculations have been made, with the results shown in Figure 3. For the calculations, the working hypothesis was made that three aerosol samplers differ with their sampling efficiency curves following exactly each of the three conventional curves (inhalable, thoracic and respirable) as shown in Figure 1. The three different aerosol samplers are used for measuring the concentration of the same ambient polydisperse aerosol characterized by an activity median aerodynamic diameter (AMAD) and a geometric standard deviation (GSD). Based on this, the calculations have been made to define the R_X factor to employ for the estimation of the $C_{AMBIENT}$ from the measurement of the C_X. The calculations were made for GSD = 1.5 and 2.5. As an example, the concentration measured by an inhalable, a thoracic or a respirable sampler should be multiplied by respectively 1.3, 2.1 or 5.6 for estimating the ambient aerosol $C_{AMBIENT}$ characterized by an AMAD equal to 10 μm and a GSD equal to 2.5.

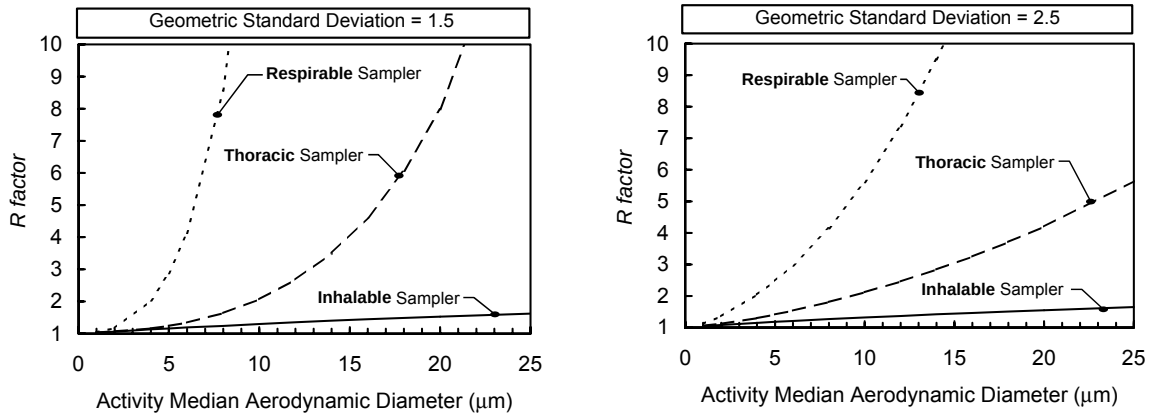

Figure 3: R_X factor to employ for the estimation of the true total (or ambient) aerosol concentration from the measured aerosol concentration corresponding to the inhalable, thoracic or respirable fraction, as a function of the activity median aerodynamic diameter (AMAD) and for two geometric standard deviations (GSD).

Also, Figure 3 shows clearly that the R_X factor is "AMAD dependent" and that this dependence differs from one aerosol sampler to another one. Moreover, for each sampler, the dependence is less important for the larger GSD value. *It means that there is no unique R_X factor*. Therefore, in theory, each concentration measurement should be associated with a particle size measurement in order to determine with the best precision the R_X factor to employ for the calculation of the ambient aerosol concentration. But in the reality of the field (or the workplaces) studies, particle size measurement is not always performed in parallel with concentration measurement. This is due to some degree to the difficulty of performing such measurement, and analyzing the data.

Therefore, in the hypothesis where the particle size characteristics are not (or unperfectly) known and if the final information targeted is the $C_{AMBIENT}$ concentration (or the activity intake), it is better to select an aerosol sampler type which does not show much AMAD dependence, like for example an inhalable aerosol sampler, rather than an aerosol sampler that is AMAD dependent, like for example a respirable sampler or a thoracic sampler.

However, when the final information targeted lin the radiation dosimetry context is the committed effective dose, the problematic is more complex. As already mentioned in the introduction, the committed effective dose may be estimated on the basis of aerosol sampling measurement results. In that case, the "true" effective dose E_{TRUE} is given by:

$$E_{TRUE} = e(AMAD, GSD, ...) \times C_X(AMAD, GSD) \times R_X(AMAD, GSD) \times B \times t_E \quad \text{(Sv)}$$

Where B and t_E are respectively the ventilation rate of the worker (m^3/h) and the duration of the exposure (h), and $e(AMAD, GSD, ...)$ is the dose coefficient for intake by inhalation of a given radionuclide. It corresponds to the committed effective dose resulting from the intake by inhalation of 1 Bq of a specific radionuclide, under a given chemical and physical form. This dose coefficient is a complex function of the particle size characteristics (AMAD and GSD) as well as other parameters related to the clearance from the lung and absorption into blood (by dissolution and uptake) of the inhaled particles. These dose coefficients can be calculated using the recent Human Respiratory Tract (HRT) Model for Radiological Protection (ICRP publication 66, 1994). Depending of the radionuclide absorption rate, the dose coefficient can be more or less AMAD (and GSD) dependent. The ICRP publication 68 (1994) gives a comprehensive list of dose coefficients for inhalation for about 800 radionuclides. For each, the dose coefficient has been calculated using the HRT model with two log-normally distributed ambient aerosols with AMAD of 1 μm and 5 μm, and GSD of 2.5. The 5 μm AMAD is a default value considered to be representative of workplace aerosols. Although to be recommended as a default value by the ICRP publication 66 (1994), it should be emphasised that this value is not always conservative (Dorrian and Bailey, 1995). For exposure of the public to radioactive aerosols in the environment, the 1 μm default AMAD is recommended by the ICRP publication 66 (1994). Here also, this value will not always be conservative. It is for example the case when people are exposed to material resuspended into atmosphere by wind, and where a larger AMAD has to be considered (Dorrian, 1997).

Different possibilities can occur depending on the AMAD (and GSD) dependency of the dose coefficient as well as the knowledge on (and correction for) the sampler performance (sampling efficiency) and particle size characteristics of the ambient aerosol (AMAD and GSD). All the possibilities are exposed in Figure 4. From these, different situations leading to different bias in the dose estimation have been defined. It can be noted that only two situations lead to no bias in the dose estimation, both including the knowledge (and correction) of the sampling efficiency and the AMAD (and GSD).

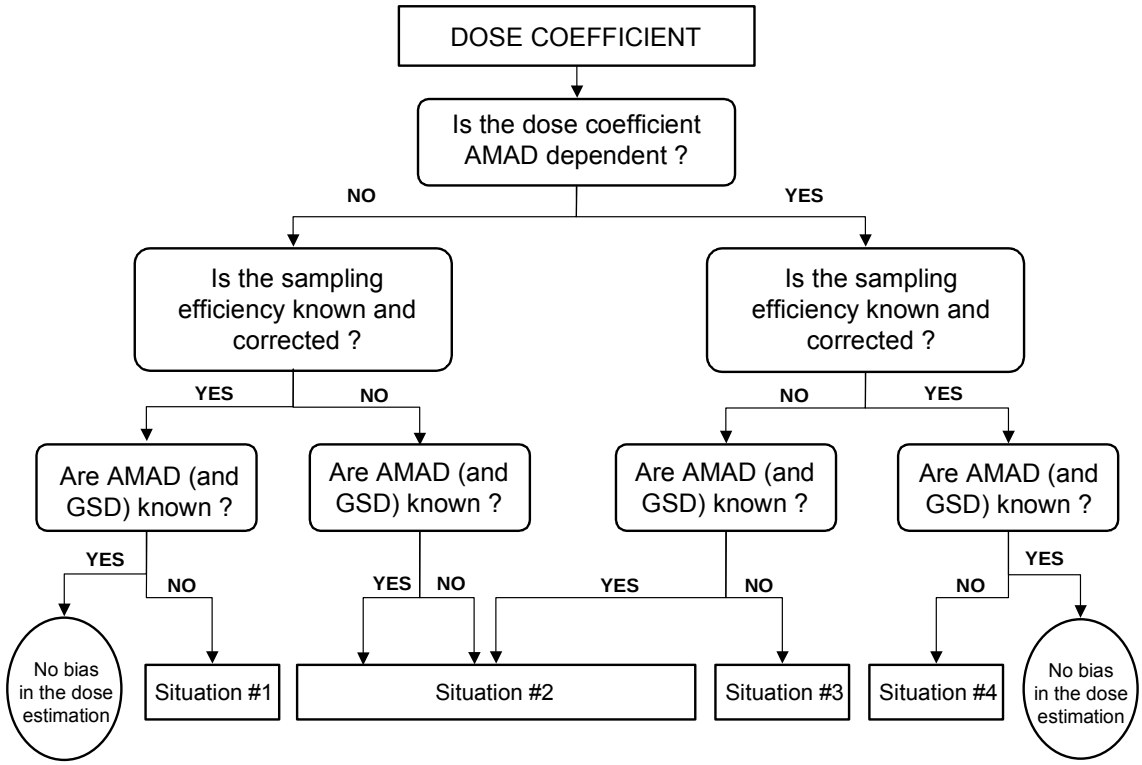

Figure 4: Schematic describing the different situations occurring in relation to aerosol sampling in the radiation protection dosimetry context, and that lead to bias in the dose estimation.

In the following, it has been defined the $AMAD_D$ and GSD_D that correspond respectively to the default AMAD and the default GSD. These default values are the values taken into account for the calculation when the particle size characteristics (i.e. the AMAD and the GSD) of the ambient aerosol are not (perfectly) known.

FromFigure 4, and for a given situation, it can be defined the bias, which express the relative difference between the dose estimated for the given situation and the dose to be estimated, i.e. the "true" dose.

The bias in the dose estimation for situation #1 is:

$$\mathrm{Bias}(\mathrm{situation\#1}) = \frac{R_X(AMAD_D, GSD_D) - R_X(AMAD, GSD)}{R_X(AMAD, GSD)} \times 100$$

The bias in the dose estimation for situation #2 is:

$$\mathrm{Bias}(\mathrm{situation\#2}) = \frac{1 - R_X(AMAD, GSD)}{R_X(AMAD, GSD)} \times 100$$

The bias in the dose estimation for situation #3 is:

$$\text{Bias}(\text{situation}\#3) = \frac{e(AMAD_D, GSD_D, ...) - e(AMAD, GSD, ...) \times R_X(AMAD, GSD)}{e(AMAD, GSD, ...) \times R_X(AMAD, GSD)} \times 100$$

The bias in the dose estimation for situation #4 is:

$$\text{Bias}(\text{situation}\#4) = \frac{e(AMAD_D, GSD_D, ...) \times R_X(AMAD_D, GSD_D) - e(AMAD, GSD, ...) \times R_X(AMAD, GSD)}{e(AMAD, GSD, ...) \times R_X(AMAD, GSD)} \times 100$$

For the situation #1 and situation #2, the bias is dependent on particle size distribution of the ambient aerosol, particle size distribution of the default aerosol, and sampling efficiency of the sampler.

For the situation #3 and situation #4, the bias is also dependent of the radionuclide and its solubility (for the calculation of the dose coefficient).

To illustrate the implication of the different described four situations, calculations have been performed to present and compare for a given situation the AMAD dependency of the bias for three types of samplers (inhalable, thoracic and respirable) and four default AMAD values: 1, 5, 10 and 20 μm. The GSD was equal to 2.5 in all four cases. As seen above in the four bias expressions, only two (#3 and #4) are dependent of the dose coefficient, and then need to be calculated with a specific radioactive compound. For these two situations, calculation of the dose coefficient for intake by inhalation has been made for a compound of U_{234} and considering a slow rate of adsorption (type S). To do this, the LUDEP 2.2 code (Jarvis, 1993), that implements the HRT Model for Radiological Protection (ICRP publication 66, 1994), has been used. Figure 5 shows the dose coefficient (in Sv/Bq) of an insoluble compound of U234 as a function of the AMAD (GSD = 2.5).

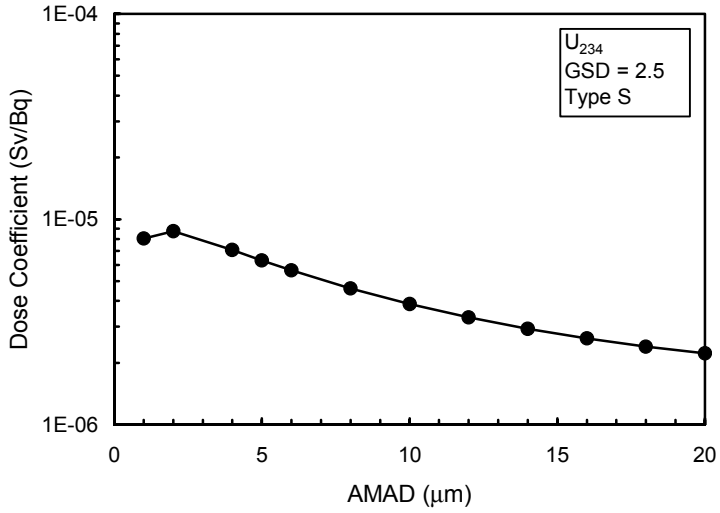

Figure 4 : Dose coefficient for intake of U_{234} by inhalation as a function of the AMAD. Calculations have been made for a insoluble compound of type S (slow rate of absorption), a GSD of 2.5, and based on the biokinetic information of the ICRP publication 30.

The results of bias calculations in the four described situations that have been highlighted in the schematic description of Figure 4 are respectively presented on the Figure 5, Figure 6, Figure 7 and Figure 8.

In Figure 5, which corresponds to the situation #1, the inhalable and respirable sampler are the ones that respectively minimise and maximise the bias, whatever the default AMAD considered. When one expect to use and correct the results of a sampler with a known sampling efficiency, for estimating the committed effective dose associated with the inhalation of radioactive compound with a dose coefficient that presents a weak dependency with the aerosol granulometry characteristics (AMAD and GSD), the use of an inhalable sampler can be advised in order to minimise the bias associated with the insufficient knowledge of the AMAD for the considered sampling period. One must remind however that the residual bias decreases with the true value of the AMAD and also that it may reach respectively –32%, –24%, 26% and 48% for default AMAD values of 1, 5, 10 and 20 μm.

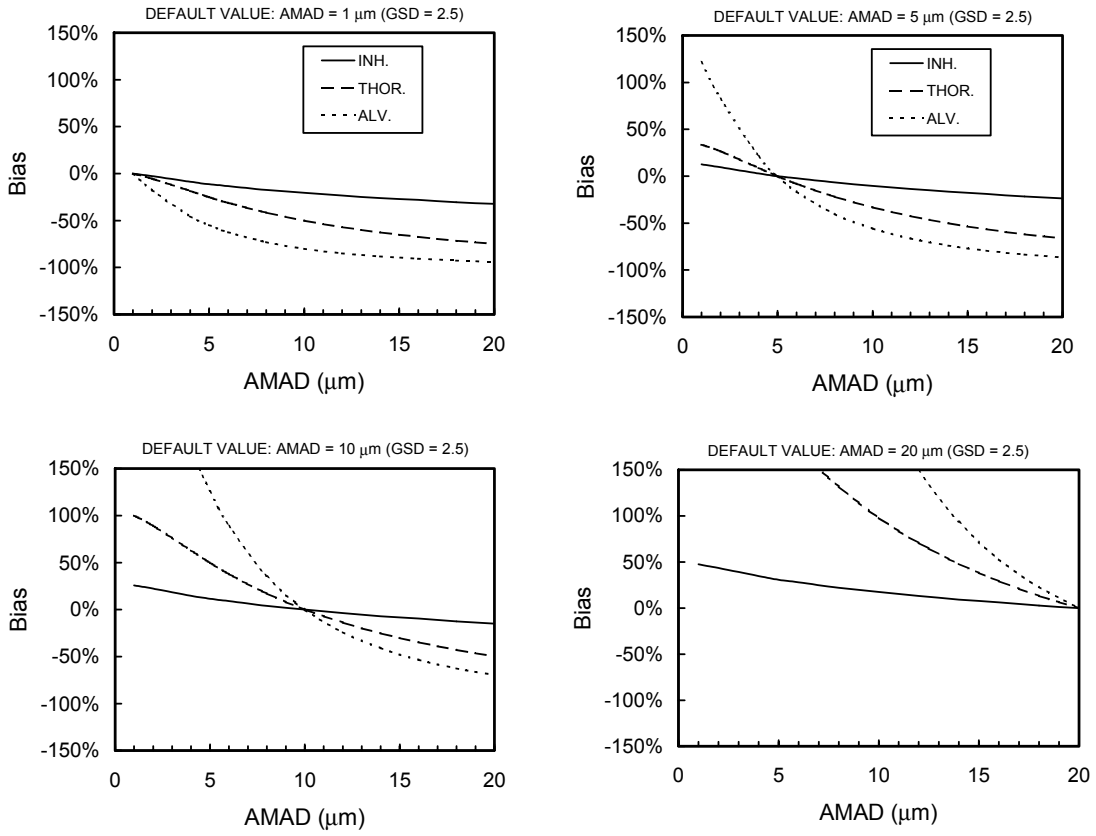

Figure 5 : Bias between the estimated dose and the true dose in situation#1. The calculations have been made for four Default AMAD (1, 5, 10 and 15 μm) and for the Default GSD of 2.5. The bias in the situation#1 does not depend of the radionuclide which is considered.

In Figure 6, which corresponds to the situation #2, the inhalable and the respirable sampler are again the ones that respectively minimise and maximise the bias, whatever the default AMAD considered. When one expects to use the results of a sampler with no correction of the sampling efficiency, for estimating the committed effective dose associated with either the inhalation of a radioactive compound whose dose coefficients presents a weak dependency with the aerosol granulometry characteristics (AMAD and GSD) or the inhalation of a radioactive compound with a dose coefficient that presents a significant dependency with the aerosol granulometry characteristics (AMAD and GSD) but for which these characteristics are perfectly known, the use of an inhalable sampler can be advised in order to minimise the bias associated with the lack of correction of the sampler sampling efficiency. One must remember however that the residual bias increases with the true value of the AMAD and may reach -35% for a true value of AMAD equal to 20 μm.

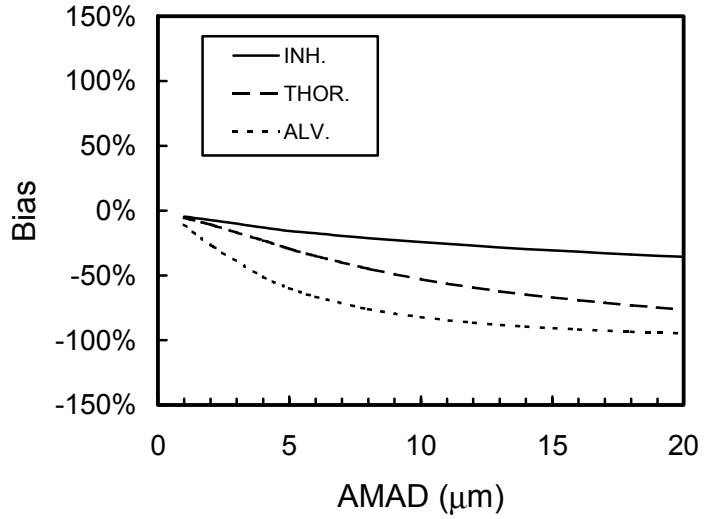

Figure 6 : Bias between the estimated dose and the true dose in situation#2. The calculations have been made for four Default AMAD (1, 5, 10 and 15 μm) and for theDefault GSD of 2.5. The bias in the situation#2 does not depend of the radionuclide which is considered.

In Figure 7, which corresponds to the situation #3 , the conclusions are more constrasted. While the use of an inhalable sampler leads to a bias that increases with the true value of the AMAD and may either underestimate or overestimate the true value of the dose, depending on the considered value for the default AMAD, the use of thoracic and respirable samplers lead to a bias that systematically underestimates the true value of the dose whatever the considered value for the default AMAD and the true value of the AMAD. In this case the thoracic sampler is the one that minimises such a systematic bias. When one expects to use the results of a sampler with no correction of the sampling efficiency, for estimating the committed effective dose associated with the inhalation of a radioactive compound with a dose coefficient that presents a strong dependency with the aerosol granulometry characteristics (AMAD and GSD) but for which these characteristics are not perfectly known, the use of a thoracic sampler can be advised in order to minimise the bias associated with both the lack of correction of the sampler sampling efficiency and the lack of knowledge of the true value of the AMAD. One must remember however that the residual bias, that does not vary significantly with the true value of the AMAD but slightly decreases with an increasing value of the AMAD considered as default, may reach values up to -77 % for a default value of AMAD equal to 20 μm.

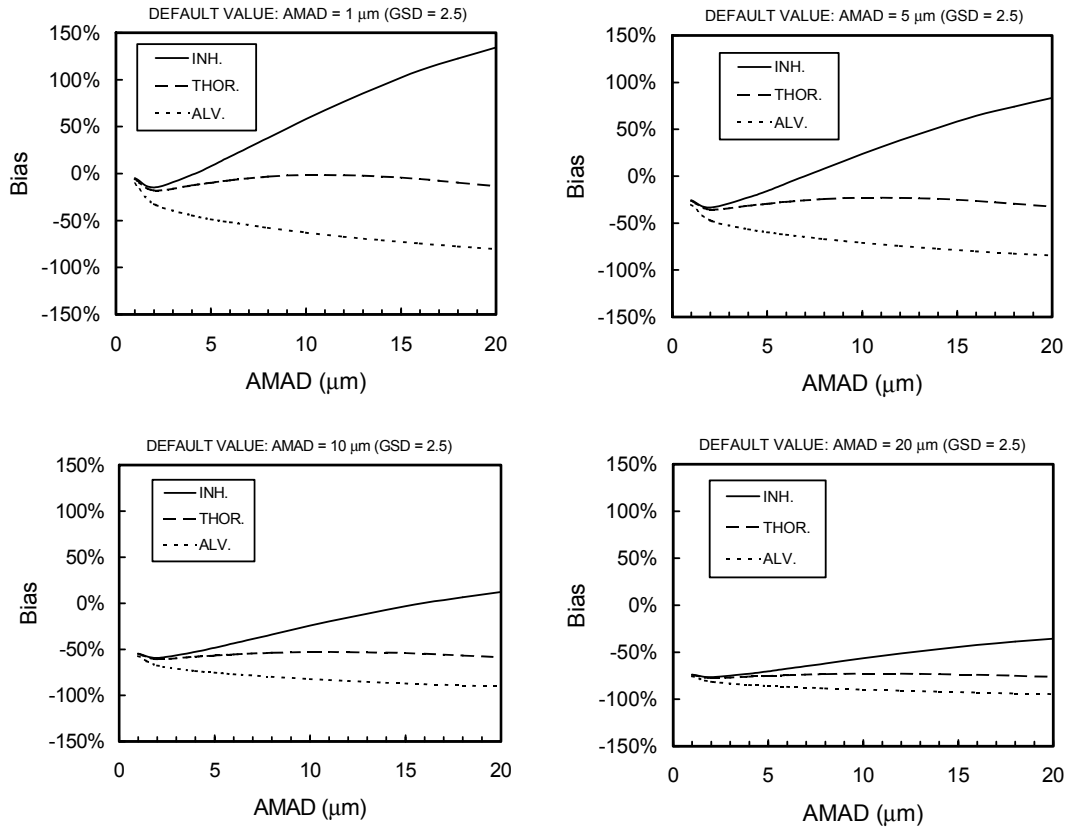

Figure 7 : Bias between the estimated dose and the true dose in situation#3. The calculations have been made for four Default AMAD (1, 5, 10 and 15 μm) and for the Default GSD of 2.5. The bias in the situation#3 depends of the radionuclide which is considered. Therefore the calculations have been made for the intake of U234 by inhalation and considering a slow rate of absorption (Type S).

In Figure 8, which corresponds to the situation #4, the thoracic sampler is again the one that minimises the bias, whatever the default AMAD considered. When one expect to use the results of a sampler with a correction of the sampling efficiency, for estimating the committed effective dose associated with the inhalation of a radioactive compound whose dose coefficients presents a strong dependency with the aerosol granulometry characteristics (AMAD and GSD) but for which these characteristics are not perfectly known, the use of a thoracic sampler can be advised in order to minimise the bias associated with the lack of knowledge on the aerosol granulometry characteristics (AMAD and GSD). One must remind however that the residual bias, that does not vary significantly with the true value of the AMAD, may reach -16% for a default value of AMAD equal to 10 μm.

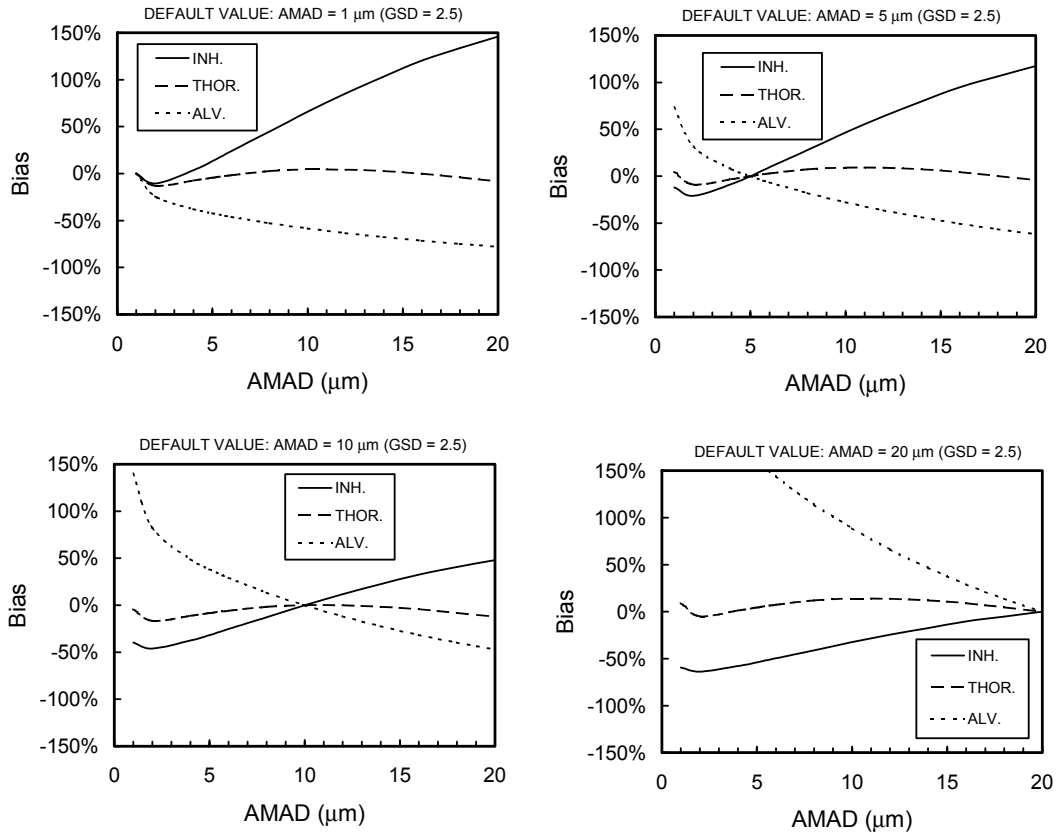

Figure 8 : Bias between the estimated dose and the true dose in situation#4. The calculations have been made for four Default AMAD (1, 5, 10 and 15 µm) and for the Default GSD of 2.5. The bias in the situation#4 depends of the radionuclide which is considered. Therefore the calculations have been made for the intake of U234 by inhalation and considering a slow rate of absorption (Type S).

As a conclusion, if one wants to minimise, for a given radioactive compound, the bias associated with the estimation of the committed effective dose on the basis of air sampling results, one should carefully select the most suitable sampling characteristics (sampling of the inhalable, thoracic, or respirable fraction) of the sampler, depending on the degree of dependency of the compound dose coefficients with the aerosol characteristics (AMAD and GSD), as well as the knowledge (and correction) of the sampling efficiency and the knowledge of the true aerosol characteristics during the sampling period.

One must remember however that, in situations where the measurement sensitivity may be an important factor, the sampling of the inhalable fraction will always lead to a higher amount of activity deposited on the filter (and thus a higher measurement sensitivity) than the sampling of the thoracic or alveolar fraction.

3.1.3. Criteria for environmental sampling

For environmental sampling (indoor and outdoor), the size selective sampling criteria are not the same as the criteria for sampling in the workplace. Figure 9 shows the comparison between the respirable and the thoracic fractions as defined in the text above with the two environmental conventions promulgated by the U.S. Environmental Protection Agency: PM2.5 and PM10 (ACGIH, 2001). These fractions are now adopted worldwide.

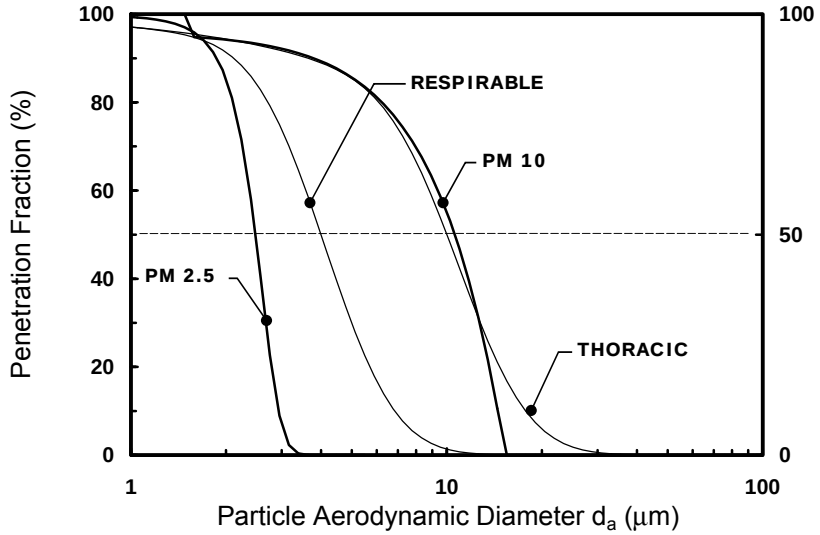

Figure 9: Comparison of the thoracic and respirable fractions for sampling in the workplaces and the EPA recommendations for the PM2.5 and PM10.

Like the thoracic sampling, the PM10 (Particulate Matter with a cut off size of 10 μm in aerodynamic particle diameter[1] is based on those particles that penetrate beyond the larynx (to the thorax). If the cut off sizes are the same for the two conventions, the two curves are different especially for the particle diameter larger than 15 μm. Inevitably, this has implication on the comparison of samplers.

3.1.4. Issues relative to the inhalability

[1] See 4.4.2 for definition

There are two emerging issues relative to the inhalability. The first relates to the inhalability in low wind environment, the second concerns the inhalability for the large particles.

3.1.4.1. Inhalability in low wind environments

There is now evidence that the air speeds in indoor workplaces rarely exceed 0.2 m/s. In a survey of air velocities measurements covering 55 work areas over a wide range of indoor workplaces, Baldwin and Maynard (1998) found that the vast majority of the background air velocities were below 0.3 m/s and were typically less than 0.1 m/s. Their work includes the relative motion between workers and their environment as they accomplish their task. The authors specified that these air movements represent the conditions that most of the workforce is exposed to for the majority of time. Whicker *et al.* (2000) made measurements of air speeds at the height of a worker's breathing zone inside a nuclear laboratory. Results show the same trend with a median velocity less than 0.2 m/s. Aitken *et al.* (1999) is the first (and still the only one) to have considered new experiments to extend the definition of the inhalability in *very slow moving air* (referred to calm air or low wind hereafter). They investigated several oral breathing rates. The curve that is proposed is shown in Figure 10. The low wind inhalability curve is significantly greater than that in moving air and defined by the inhalable convention. This curve is thought to correspond to the "worst case" situation (oral breathing of 20 l/min). Although it is at present too early to take this relation as firm, certainly because it needs independent and new experiments, such suggestion for low-wind inhalability can be used in comparison studies with inhalable sampler efficiencies that would be measured in such equivalent calm air conditions.

3.1.4.2. Inhalability for large particles

The second emerging issue relates to the position of the worker from the contamination source (dust source). Observations suggest that in most of the situations encountered in workplaces, the location of the worker is close to the contamination source. Moreover, the worker many times faces the source. That means the orientation is 0°. For work situations where the environment is very dusty (like for example mining griddling, etc.), large particle (above 100 μm) can be inhaled by the worker, posing a potential health risk. But, the inhalable fraction (convention) is not defined above 100 μm, because there were not published data. Kennedy and Hinds (2002) recently investigates the inhalability of large and solid particles with diameters up to about 150 μm. The curve that is proposed is shown in Figure 10. The orientation averaged inhalability curve generates by the recent study shows a significant deviation from the inhalable

convention. According to the author, the source of the difference is unknown, but may be related to differences in experimental setup! It needs further investigation.

A specific and very surprising issue relates to the ICRP publication 66 (1994). The inhalability expression, which is taken into account in the publication, is not the one given by the inhalable convention. Although the experimental data base, on which the two expressions are fitted, is exactly the same, the resulting fitting curves are not the same! It should be noted here that the inhalability expression is only used for the calculation of the dose coefficient (Sv/Bq) through the HRT Model.

This difference is well observed in Figure 10 with comparison to the inhalable convention. It is beyond the scope of the present document to argue about the differences between curves. In general, the approach followed in standards for occupational health and hygiene measurements, is to tend toward the “worst case” condition. Following this philosophy, the curve for the low wind proposed by Aitken *et al.* (1999) suggests a strong basis for modifying the inhalable convention. However, it should be noted here that this is the only available published data showing this tendency, and therefoe before making any recommendation, further data are clearly needed. The other philosophy would be to generalize the inhalable convention and produce a modified single convention which encompassed all windspeeds from very low wind to large wind.

On the comparison between the curve entitled “Large Particles” and the inhalable convention, the source of the difference is unclear. Kennedy and Hinds (2002) indicate some possible explanations: the difference on the methods used to determine orientation-averaged inhalability, the charge of the particles of the test aerosols (they neutralize the charge but the data used as basis for describing the inhalable convention were obtained without neutralization of the test aerosols), the difference in the bearthing mechanism of the mannequins used for the experiments (the mannequin used by Kennedy and Hinds inhaled and exhaled through the same path but mannequins used in the previous studies inhaled through the mouth and the exhaled air exited either through the back of the head or through the nostrils), the differences in the complex (and often unique) experimental facilities. All theses differences coud result in lower values for inhalability for the measurement conducted by Kennedy and Hinds. However, it should be noted that the investigators took care to minimize sampling errors and have confidence in the data, and so do we. One very new result concerns the inhalability for particles larger than 100 μm. Certainly, the observed tendency is a result of the competition between horizontal velocity and settling velocity. Here again it is clear that further investigations are needed.

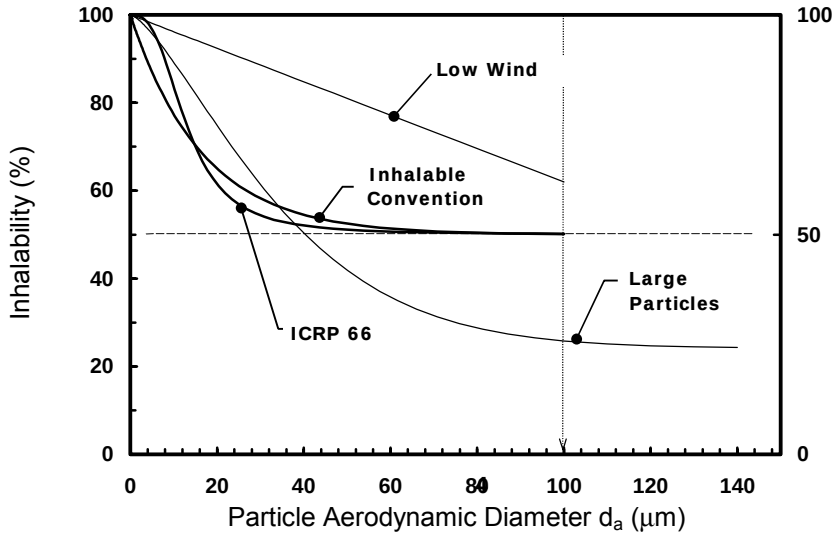

Figure 10 : Comparison of the inhalable convention (as defined by the CEN, ISO and ACGIH) with the proposition for low wind inhalability (Aitken *et al.,* 1999) and inhalability for solid large particles (Kennedy and Hinds, 2002), and the inhalability curve in the ICRP publication 66.

To illustrate the implication of the different curves presented in Figure 10, calculations were made and the results are presented in the Table 1. It can be observed that the fractions calculated with the different inhalability expressions show significant differences already for the finest aerosol with a AMAD of 5 μm. It is also observed that the low wind inhalability fraction stays very close to the true total fraction even for the larger aerosol.

Table 1: Calculations of concentration fractions relative to the *true total (or ambient) aerosol*. Fractions were calculated with the inhalable convention, the low wind inhalability (Aitken *et al.,* 1999), the large particles inhalability (Kennedy and Hinds, 2002) and the inhalability for the ICRP publication 66. Calculations are made for three log-normally distributed aerosol size distributions with a geometric standard deviation (GSD) of 2. Calculations are made for three log-normally distributed aerosol size distributions with a geometric standard deviation (GSD) of 2.

AMAD (μm)	5	10	15
Convention	0.85	0.76	0.70
Low wind	0.98	0.95	0.93
Large particles	0.94	0.86	0.78
ICRP 66	0.92	0.81	0.72

To summarize, the different criteria for sampling in the workplaces (Figure 1) or for environmental sampling (Figure 9) are important as they are standards to which aerosol samplers should conform. However, several problems still remain in particular with the implementation of the inhalable convention with the need to improve the relevance of the convention in more realistic working conditions (Kenny, 2000), i.e. corresponding to calm air environments or to situations where the worker is close to a dust source that disperse in the atmosphere large particles than can be inhaled.

3.2. Performance consideration for workplace aerosol samplers

In the early 1980's, the aerosol science and in particular the industrial hygiene community became aware that aerosol sampling for aerosol exposure assessment was not as simple as previously thought. In particular, it was realized that simply drawing air through a filter and measuring the particle matter that is collected is not truly representative of either true total ambient aerosol or what workers are actually exposed to.

3.2.1. Factors influencing the sampling performance

Short discussion of sampler performance can begin by referring to Figure 11, which represent the air flow and particles trajectories near an aspirating inlet at a given direction with respect to the external incoming air flow. The most important aspect of the performance of an aerosol sampler is the *sampling efficiency* with which particles are transferred by aspiration from the air outside the sampler and into the sampler through its one or more entry orifices. The sampling efficiency is the product of the *aspiration* efficiency and the *transmission* efficiency (also called *penetration* efficiency). The aspiration efficiency is a strong function of particle size, sampling flow rate, wind velocity, sampler orientation, sampler size and shape. After aspiration, the particles are usually transported through some sort of duct to a filter or to a sensing zone (for direct-reading aerosol instruments). During such transport, deposition on the internal walls of the sampler may take place by a variety of mechanisms (sedimentation, inertial impaction, electrostatic attraction). Altogether, these numerous mechanisms contribute to generate a bias between the ambient aerosol in which the sampler operates and the actual aerosol which is collected on the filter (or measured in the sensing zone). Finally, the aerosol sampler might overestimate (means that the sampling efficiency is above 100 %) or underestimate (means that the sampling efficiency is below 100%) the true concentration. The sampling efficiency depends on the balance between the aspiration and the deposition, the latter always contributing to the under sampling due to the losses inside the sampler lines. For further information, an important review of sampling theory and practice is well compiled in a book by Vincent (1989).

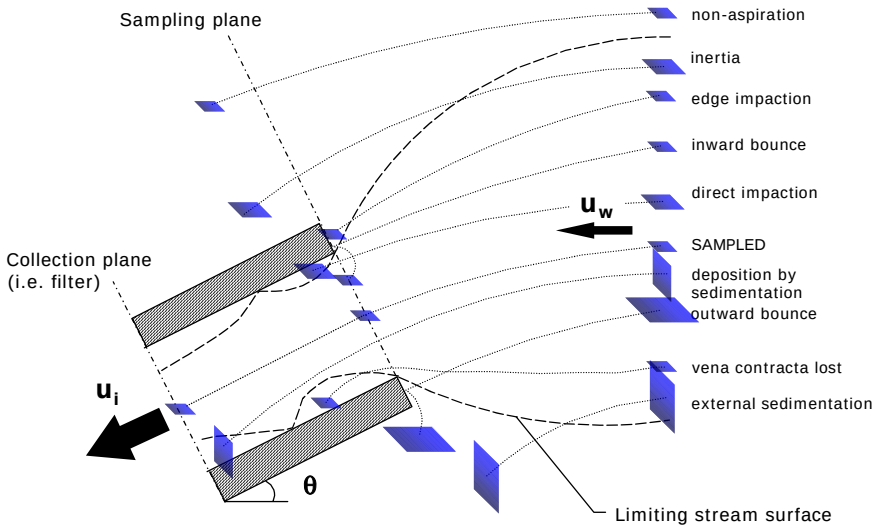

Figure 11 : Schematic representation of the different mechanisms that affect the sampling efficiency of an inlet. The drawing is made for an inlet with an aspiration velocity higher than the air velocity outside, and with an angle between the inlet axis and the incoming air flow.

The Table 2 presents a list of the principal factors known to influence more or less significantly the sampling performance of aerosol samplers (Witschger, 2000).

Table 2 : Compilation of factors that influence the sampling performance of aerosol samplers.

Factor	Nature of Effect	Sampler types
Particle size	Size-dependent selection of particles (aspiration, deposition)	All samplers
Wind speed	Affect aspiration of particles (large particles)	Any sampler not having an isokinetic[2] inlet (for moving air)
Wind orientation	Affect aspiration of particles (large wind speed)	Any sampler not having an omnidirectional inlet
Nearby human body	Affect flow field near inlet	Many inhalable samplers
Wind turbulence	Variability of the aspiration	All samplers having wind speed and orientation dependence
Aerosol composition	Particle bounce or re-entrainment Breakdown of agglomerates	All samplers having large bluff body

[2] an isokinetic inlet is an inlet in which the air flow is characterized by the same velocity and direction as the ambient air flow.

Humidity	Mass variation of filter cartridge	All sampler using a filter cartridge system
Inlet shape	Orientation-dependency and deposition of particles Over sampling of very large particles Passive sampling	Especially inhalable samplers
Inlet-filter geometry	Transmission losses Uniformity of sampled aerosol	Many samplers
Filter sealing	Particle deposition on the periphery of the filter may be lost	All samplers using filters
Sampler integrity	Particles my be lost due to leakage especially around filter	Any sampler not airtight
Sampler handling	Variability of the results due to difficulties during disassembling	Any samplers not user-friendly
Specimen variability	Small dimensional differences may cause large aerodynamic effects	e.g. cyclones, impactors
Sampled aerosol mass	Collection efficiency changes for heavily loaded surfaces	e.g. impactors, samplers using porous foam as selector
Electrostatic charge	Attraction to and repulsion from surfaces	Any sampler build with non-conducting material
Flowrate variation	Particle separation mechanism strongly flow-dependent	e.g. cyclones, elutriators, impactors
Surface treatments	Collection efficiency depends on collection surface or medium	e.g. impactors, impingers

Finally, each sampler has its own specific behavior, which is defined by its own sampling efficiency. As said previously, this function is complex and involves many factors related to the environment as well as the ambient aerosol and sampler working conditions like stability of the flowrate, size of the entry orifice, or position on the worker etc.

3.2.2. Evaluation of sampling performance in laboratory

The evaluation of the sampling performances in a laboratory requires a well defined experimental protocol. The traditional protocols use either a wind tunnel, for evaluation in moving air with velocity > 0.5 m/s (like the one's developed by Witschger *et al.,* 1997), or a so-called calm air chamber, for evaluation in low wind with air velocity < 0.1 m/s, like the one's developed by Kenny *et al.* (1999). Since the introduction of the inhalability concept, the aerosol samplers devoted to personal sampling should be tested when mounted on a mannequin. This configuration is essential only for moving air, where the presence of the mannequin affects significantly the airflow around the personal sampler (Witschger *et al.,* 1998), but not in calm air environment. The moving air conditions are rarely encountered in the reality, except where forced ventilation is employed or close to open doors. The same is true for the aerosol, which is used in these performance tests, always homogeneous, and thus representative of contamination source that are far from the exposed simulated worker. All of this implies that new development of more realistic protocols are needed and some of them are currently under way.

3.2.2.1. Moving air

In moving air, there are two approaches.

The first approach is known as the Simplified Test Protocol. It was designed with the intention to simplify and reduce the cost of the experiments. It uses as a basis a simplified torso and was initially proposed by Witschger *et al.* (1998). The rationale behind the simplified test torso is to simulate the middle part of the human torso where inhalable dust samplers are usually mounted. It is a three-dimensional rectangular body having rounded corners to simulate the effect of the human body on the sampler. This Simplified Test Protocol has been since successfully adopted for evaluating a number of inhalable sampler performances in moving air (Aizenberg *et al.,* 2000a; Kennedy *et al.,* 2001) but always in large cross-section wind tunnel. The final step of validating the Simplified Test Protocol has been recently carried out by

Aizenberg *et al.* (2001), where it was used in a small wind tunnel capable of generating very large particles (> 100 μm) at wind velocities of 0.5 and 1.0 m/s.

The second approach uses scaling relationships to prescribe experimental conditions and small-scale sampler design that can be tested in small wind tunnel (Ramachandran *et al.,* 1998). At this time, the latter approach has not been further investigated or applied.

3.2.2.2. Calm air

Observations of what is currently done in the calm air sampling tests (that is very few) put forward the clear need to design a new experimental protocol for measuring the sampling efficiency of aerosol samplers in very slowly moving air and near a dust source. With the intention from the beginning to design something easily duplicable by any laboratory in order to carry out in the very close future similar work to compare with their experimental results, Witschger *et al.* (2002a) have recently proposed a new experimental sampling test protocol. The simple arrangement consists of a generation system that continuously rotates and gently disperses in an omni-directional way the test aerosol being transported by turbulent diffusion and natural convection to the samplers to be tested and to the reference samplers. It uses classic equipments, providing a low-cost method. The close source of the test aerosols in our test system resembles a point or area source as it is observed at workplaces rather than the homogeneous cloud used in traditional evaluating protocols. Moreover, the direction facing to the source (facing or referred to as 0°) of the samplers to be tested is representative of what it is seen at indoor workplaces: worker usually faces the major dust source.

The test system has been used to evaluate sampling performances of existing personal samplers. This test protocol is thought to be also applicable for testing area (or static) samplers.

Although it is expected that the sampling efficiency of the selected sampler measured during laboratory tests is as close as possible to the corresponding conventional curve (inhalable, thoracic or respirable), some deviations between both functions may be generally observed (through specific experimental tests), leading to some bias between the measured concentration (by the selected samplers) and the conventional concentration. The bias expresses the degree of conformity of the sampler to the sampling convention.

3.2.3. Field tests

Field tests are carried out primarily for comparisons of various samplers. Analysis of data from a field study allows a correction function to be obtained that relates aerosol concentrations measured by a given sampler to those measured by another sampler taken as reference. It is important to have in mind that the correction function (or factor) is specific to the workplace activity(ies) included in the field study, and cannot be assumed to apply to different circumstances. Because of the typical variability of aerosol concentration in the field, it is difficult to use these situations for accurate assessment of sampler performance. However, field studies are important to verify the overall performance of a sampler, and to indicate specific sampler problems that are usually highlighted only in the field (and people that operate in the field know well that usually it never goes the way it is first thought!). Usually, these field studies suffer from the lack of enough repetitive measurements or additional measurements that can be used for the analysis, like air velocity measurements (a good index for the migration - transfer of the contamination) or like the existence of any predominant direction to the source, etc.

To conduct a study for sampling performance evaluation is still a big challenge. However, these studies are extremely important in order to analyze measurements in the field studies.

3.3. Sampling strategies for exposure assessment

Short discussion of sampling strategy can begin by referring to Figure 12. Regarding the question of how best to reflect the true exposure of a worker (or a group of worker), it is far beyond the scope of the present document to expose all the different concepts in order to select an homogenous group of workers, frequency of measurements, duration etc. The reader is invited to read the brief paper from Gardiner (1995) or the more recent (but more complicated) from Tielemans *et al.(*1998).

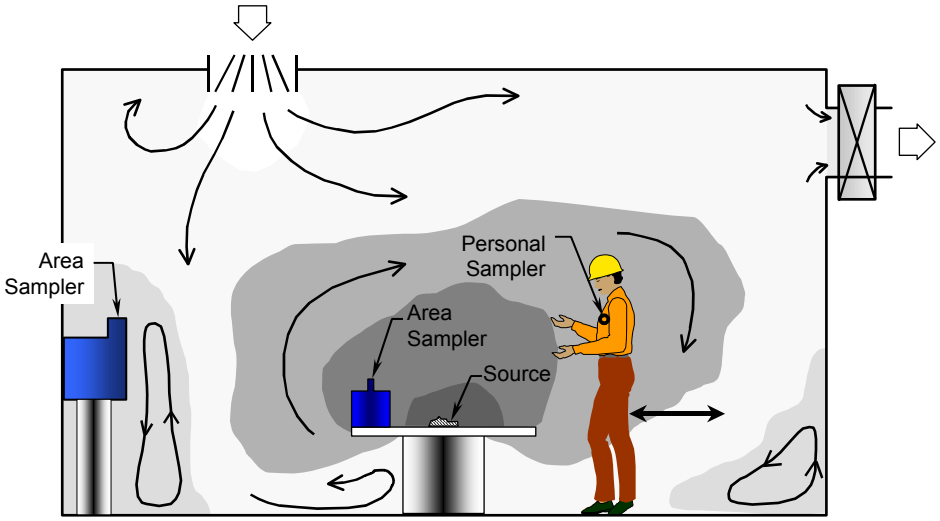

Figure 12 : Illustration of the nature of the dispersion of the contamination in an indoor workplace.

3.3.1. Area vs. personal sampling

The placement of an area sampler in the workplace when the measurement is intended to be representative of the aerosol to which a worker is exposed to is strategic. Ideally, one wishes to characterize the microenvironment in the breathing zone of the worker to evaluate its specific exposure. There are two types of measurement that can be carried out in the workplace:

- *area* (also called *static* or *at fixed position*) measurement where the chosen aerosol sampler is placed *somewhere*, its location being thought to be *relevant*, meaning that the concentration measured is *representative* of the ambient aerosol,

- *personal* measurement where the sampler is mounted on the body of the worker, thus moving all the time with the worker; the aspiration orifice of the sampler is placed in the "breathing zone" of the worker.

One advantage with the area samplers is that they have high flowrates, making them attractive where the level of the particulate contamination is low, because a large amount of material can be sampled in a short period. Moreover, they are usually easy to use.

The use of personal samplers is more labour intensive and require the cooperation and efforts from the workers themselves. However, it is now widely accepted that the health-related sampling in the workplace should be conducted by personal samplers mounted on the workers. The location of the personal sampler should be in the "breathing zone", a region of the body defined as an hemisphere centered on the mouth and nose and having a radius of about 30 cm (Vincent, 1995), as it is illustrated in Figure 13. *But here, it is extremely important to understand that it is not because the personal sampler is located in this region that the sample will be representative. If the personal sampler has a poor sampling performance, the measurement will not be representative.* Thus, once again, the most important information to know when using a personal sampler is its sampling efficiency (with the remarks made in the previous chapter 3.2).

Figure 13 : Location on worker of personal sampler with the predominant facing to the dust source direction.

The results from the field studies usually reveal significant differences in the aerosol concentration when comparing a personal sampler to an area sampler, but also area samplers between them as recently reviewed by Witschger (2000). This is attributed to two phenomena: the particle transport from the dust source to the sampling point (where the given sampler aspirates the aerosol laden air) and the sampling efficiency of the given sampler. The airborne particle transport throughout the workplace is strongly dependent mostly on the source

characteristics and the airflow pattern in the environment. In turn, the placement of the aerosol sampler, and specifically when it is an area aerosol sampler, is also an important issue especially if the need is to get a rapid and reliable detection of contamination release for alarm (Whicker *et al.,* 1997).

3.3.2. Transfer studies and modelling

In order to identify which of the above two phenomena had the most significant effect on the noticed difference, and also to understand why, specific studies can be carried out in the workplaces to evaluate the transport phenomena or migration (also called *transfer*) of the contamination (Boulaud *et al.,* 1994). These transfer studies are usually based on the use of a tracer gas (like He or SF_6). Hence it is theorized that the tracer gas mimics well the transfer process of the contamination of interest, which is not always true for an aerosol. Since an aerosol consists of particles suspended in the air, it is expected that the behavior of an aerosol will be highly dependent on the behavior of the air itself, and in that sense it is true, and the use of a tracer gas brings some information. But due to the particulate phase, the evolution and the behavior of an aerosol change in many ways from those of the air. Moreover, the evolution can be due to many phenomena like growth by coagulation, agglomeration, condensation, sedimentation, turbulent diffusion etc. Obviously, these phenomena depend on the aerosol concentration, particle size, level of charge, material, generation process, etc.

In particular, in the transfer studies that have been conducted at this time, aerosol sedimentation and wall deposition by turbulent diffusion are the two phenomena that limit the use of tracer gas to measure the aerosol transfer. For example, in a recent study carried out in a laboratory ventilated room, it was clearly demonstrated that for aerodynamic particle diameters greater than about 5 to 10 μm, the transfer studies should use particles as tracer (Bemer *et al.,* 2000). There is therefore a need to develop reliable and simple tracer solid particles generation systems that could be used directly in the workplaces, the traditional aerosol generation systems being mostly applicable only for laboratory experiments or for liquid particles. A new way of development concerns also the particle detection in real time with very low concentration level for these transfer studies. As a example, a new system has been developed and is currently under testing in order to measure in real time the particle concentration using the fluorescence detection (Prevost *et al*., 1997).

Also, the development of numerical simulation tools costing less make these tools attractive in transfer studies, and then for exposure assessment studies, especially to examine effects of different variables of interest that are difficult to test in the workplace (Bennet *et al.*, 2000). However, boundary conditions as well as calculation for real workroom configurations make

these numerical studies at this time not as easy as first thought, but also not completely reliable. Hence, research is needed to confirm the ability of numerical calculations to represent indoor aerosol dispersion through validation with experimental reliable data.

4. AEROSOL SAMPLING IN THE WORKPLACES

Aerosol sampling for radioactive particles can be used to determine whether the confinement of radioactive particulates is effective (for example from a glove box), to warn of significantly elevated levels of radioactivity in the air, to determine what protective equipment and radioprotection measures are appropriate, to demonstrate compliance with regulatory requirements, to predict or assess radioactive doses to the respiratory tract (Perrin *et al.,* 2002). This chapter deals only with the latter aspect. Therefore, it includes a review of different techniques that are intended to measure health related aerosol characteristics. A number of aerosol samplers exist now in the market, some of them being old, some new. But not all of these samplers have been yet tested either against the sampling conventions or against the 100% efficiency curve. Also, very surprisingly, there are samplers that are used without knowing their aerosol sampling performance.

The review is focused of the major aerosol samplers that are used more or less widely in the industrial hygiene as well as some specific instruments used by some of the partners involved in the SMOPIE project. As the radioactive aerosols are of concern, and for the reason exposed in chapter 3.1.1, the chapter deals specifically with the inhalable samplers as, it is thought that the convention describing the inhalable fraction is appropriate. However, the reader is invited to consult the comprehensive list of air sampling instruments encountered in the industrial hygiene world edited regularly by the ACGIH (2001).

4.1. Aerosol concentration, particle size and shape

For workplace aerosols, in industrial hygiene, the aerosol concentration is usually expressed in terms of particulate mass per unit of air volume. The level of the mass concentration ranges over orders of magnitude from hundreds of mg/m^3 down to few $\mu g/m^3$. A related property is the number concentration ($particles/m^3$). In the nuclear sector, the concentration is expressed in terms of activity per unit of air volume (Bq/m^3). Calculations can be made to express the concentration in one of these units. However, care must be taken as these calculations require hypothesis that are sometimes difficult to verify. A good example is the change from $particles/m^3$ to mg/m^3, which require to know the individual mass of the particles and then the density of the particles, the particle size distribution, and the shape. These important parameters are often not well characterized.

An extremely important property of the aerosol particles is their size. It describes the particle behavior and residence time in the air environment. But it is a property whose definition is not always as simple as might at first appear. The simplest case is to consider that the particle is perfectly spherical. It is rarely the case in the workplaces where the shapes that can be encountered vary from regular/isometric to platelet of fiber. Only for liquid particles, the hypothesis to be a sphere can usually be done. For combining many aspects of the airborne behavior of the particles, the *aerodynamic particle diameter* is the most widely *equivalent diameter* used in the industrial hygiene context. It is defined as the diameter of a spherical particle of density 1 g/cm3 (equivalent to that of water) that has the same falling velocity (also called sedimentation velocity) in air as the particle in question. When neglecting the slip correction for the very small particles, the aerodynamic particle diameter da is calculated by the following expression:

$$da = dv \times \left(\frac{\rho_p}{\rho_0 \chi} \right)^{0.5}$$

where dv is the equivalent volume diameter (diameter of the sphere having the same volume as that of the irregular particle in question), ρ_p and ρ_0 the density of the particle and the water (1g/cm3), and χ a correction factor called the dynamic shape factor. The dynamic shape factor can be significantly different from the unity. For example, it is about 1.3 to 1.5 for the alumna fine powder (density close to 4 g/cm^3). The aerodynamic particle diameter is a key parameter for characterizing sampling performances of samplers, but also respiratory deposition, filtration, contamination transfer. Only a few devices using the aerodynamic separation can measure the aerodynamic particle diameter, like for example the cascade impactors or the aerodynamic particle sizer (APS).

Different types of particles lead to different dynamic shape factors. Usually this factor depends on the initial generation process. The measurement of this factor is not trivial and can be performed only by comparing a measurement based on the aerodynamic separation and a measurement of the equivalent volume diameter, like for example with the Coulter technique (Witschger *et al.,* 2002b).

4.2. Aerosol measurement errors

As said previously, the performance testing of an aerosol sampler is still difficult, time consuming and costly, even if new simpler methods are currently under way. However, the sampling performance should be known with the best possible precision , to make the results of the field measurements reliable.

Figure 14 exposes some major sources of biases that may occur in aerosol measurement.

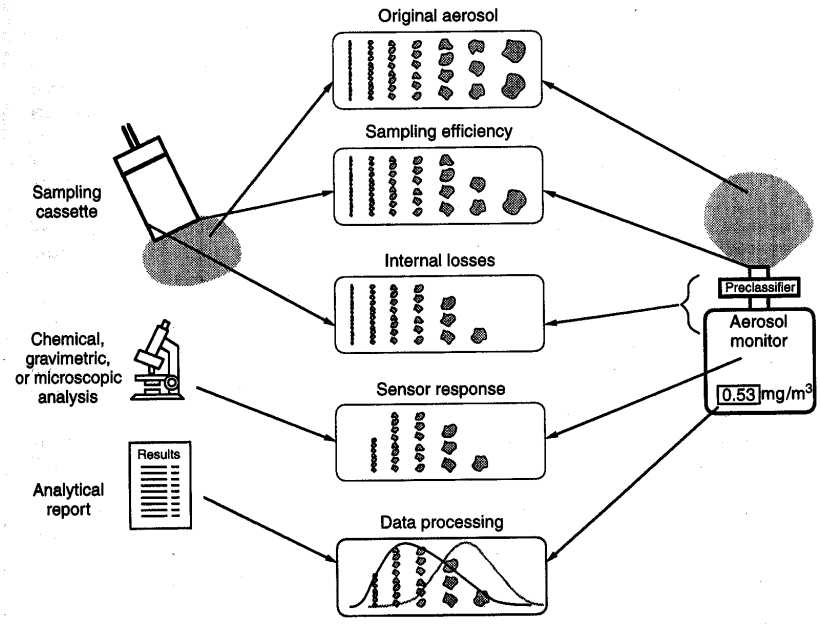

Figure 14 : Schematic representation of some important biases in aerosol sampling (From Baron and Heitbrink, 2001)

Until now, attention has been focused largely on the sampler itself. However, no discussion of aerosol sampling can take place without any reference to flowrate. Accurate aerosol concentration measurement needs accurate measurement of the total volume of air sampled and this total volume is derived from the flowrate and the sampling duration. The measurement of the duration does not need any further explanation except to be careful that its value is really well known! However, flowrate needs calibration. This calibration needs absolutely to be performed *with* the sampler connected and *with* the filter placed in the sampler (to respect the pressure drop effect on the flowrate). Ideally, the calibration should be done under the same conditions of temperature and humidity as those in the workplaces where sampling will be carried out. If the conditions have changed or are expected to change, a correction has to be taken into account to recalculate the real flowrate at the working conditions. Typically, the set

flowrate is expected to be within 5% of the working (or nominal) flowrate. Some of the pumps that are sold with aerosol samplers have their own volume measurement (especially some personal pumps). The more recent ones (but more expensive) regulate the selected flowrate to minimize the impact of changes in temperature, pressure and filter loading on the flowrate and the total volume of air sampled. However it is always recommended to make a calibration of the volume. Some of the pumps may generate pulsations in the flowrate. It has been shown that these pulsations, resulting in the change of the aspiration velocity, have a effect on the sampling performance. It is particularly true for aerosol samplers that are highly dependent of the flowrate, like the cyclones (*Bartley et al.,* 1984). Obviously, flow calibrations need to be performed with calibrated flow meters! Primary standards such as bubble flow meters, commercially available, are preferable.

Even when a given instrument performance is known, and its flowrate calibrated, it is important to remember that, in the workplace, damage and impact from handling are factors that can highly alter the result of the measurement made by the instrument. Therefore, an important issue in the overall performance of an aerosol sampler is the ease with which it can be operated in the field. In particular, the personal sampler must be comfortable for the worker. Moreover, it has to be easy to disassemble in order to replace the filter, etc.

4.3. Personal aerosol samplers

4.3.1. Inhalable Samplers

4.3.1.1. The filter plastic cassettes

The inhalable sampler most widely used in many countries in the world of the industrial hygienists is the 37-mm plastic cassette. This cassette may be used in its open-face version (like in Sweden) or, more commonly, in its closed-face version (like in Britain and U.S.). The latter has a single orifice of 4 mm in diameter through which a fraction of the ambient aerosol is aspirated (see Figure 15, A). When attached on the worker's collarbone, the inlet is always facing downward with its axis at an angle of ≈ 45° to the vertical (Buchan *et al.,* 1986). Figure 15 B presents an holder that is sometimes used in the field to keep the personal sampler in the same position on the worker.

The preferred flow in most of the countries is 2 l/min, but in some countries (like in France) the standardized flow is 1 l/min. In most of the countries, the filter used is 37 mm in diameter (hence, the name of the sampler). However in some European countries, a similar version is used but with a 25 mm filter diameter. The 25-mm filter cassette has exactly the same shape, is

made of the same material, but is smaller. Norway and Denmark use the closed-face 25-mm cassette. France has standardized the closed-face 25-mm cassette with a flowrate of 1 l/min. Moreover, the closed-face 25 mm is the only personal dust sampler described in a French standard (AFNOR, 1988a). It is important to note that, at the time of the writing of the standard, it was intended to collect the “inspirable” fraction (former name for the inhalable fraction). However, in Norway and Denmark as well as in Britain and United States, this sampler is intended to sample the “total” fraction …

For many years, this sampler was used without knowing really what were its sampling performances. Its use was due to its very simple design. There is now a general consensus to say that this sampler shows very poor performances in terms of sampling efficiency but also exhibits specific problems that make this sampler no longer a good one for evaluating the inhalable fraction. The poor performances of the 37 mm cassette are well documented in laboratory experiments using rotating mannequin in moving air (Kenny *et al.,* 1997) or calm air (Kenny *et al.,* 1999). It has been also tested with the Simplified Test Protocol by Aizenberg *et al.* (2000a). Only recently, this sampler has been tested in very slowly moving air and near the contamination source, a situation thought to be representative of most of the exposure situation encountered in the workplaces, by Witschger *et al*. (2002a). From the later study, the bias in concentration relative to the 100% efficiency curve has been estimated to be –33% for a polydisperse aerosol with a AMAD of 5 μm (GSD = 2), and –54% with a AMAD of 10 μm (GSD = 2). Moreover, the sampler results show a large dispersion, making this sampler not really reliable.

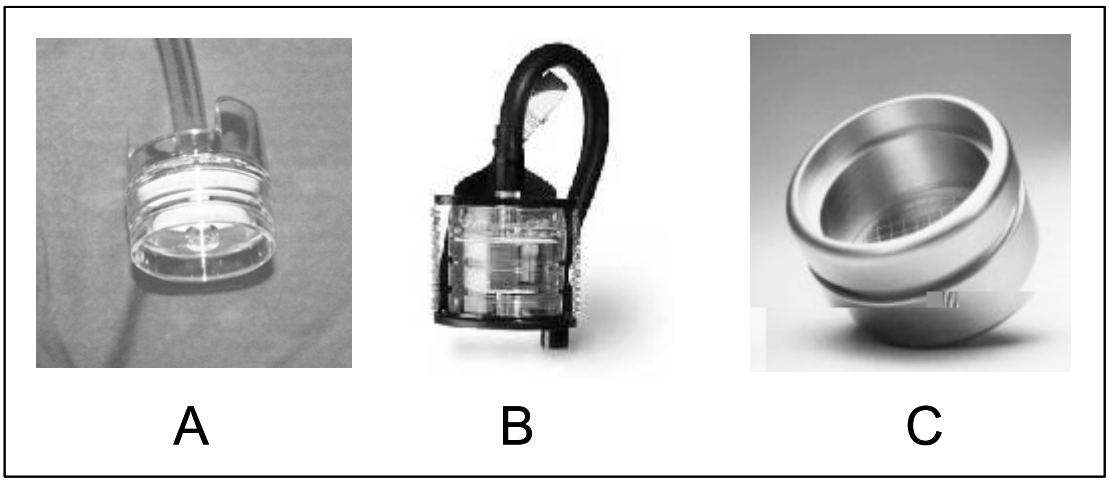

Figure 15 : The 37 mm cassette personal aerosol sampler (shown in the common closed-face version – marketed by Omega Corp. in U.S.). A: placed on a human torso. B: presented with a cassette holder (not a common use). C: metal version of the filter holder to static charges (not a common use).

The working protocol of the cassette indicates that the sampled aerosol is defined on the basis of the aerosol collected on the filter. It is now well established in the hygiene community that this personal sampler has a number of known problems related to its plastic material, cassette

assembly, orientation when attached, uniformity of the deposit on the filter, internal losses etc (Demange *et al.,* 2002; Hinds, 1999; Puskar e*t al.,* 1991). Figure 15 C presents a metal version of the major part of the cassette that may be used to avoid electrostatic problems. However, the performance of this system has not been yet tested. One problem of this sampler that is often underestimated is that it needs to be well assembled to be airtight and therefore to avoid air leakage.

The geometry of the closed-face cassettes causes the deposit to be concentrated on the centre of the filter (due to the high aspiration velocity at the 4 mm orifice and the short distance up to the collection filter), and therefore highly non- uniform on the filter. Therefore, it could be a problem for analysis that requires a good uniformity of the deposit like microscopy or radiation counting. Altogether, use of this sampler is not reliable for aerosol exposure studies.

4.3.1.2. The IOM Inhalable Sampler

The IOM Inhalable Sampler (shown in Figure 17) ,referred to hereafter as IOM, is a device where the aerosol is aspirated through a 15 mm circular protruding inlet at a flow rate of 2 L/min, the particles being subsequently collected on a 25 mm filter or deposited on the internal walls of a lightweight cartridge. *The IOM protocol specifies that both the filter and the cartridge are weighted together* in order to include in the sample any aspirated particles

The cylindrical body of the IOM is made of a conductive plastic. The cartridge is either made of conductive plastic or stainless steel. The latter is preferable to avoid any moisture effect on the gravimetric analysis. When attached on the worker's collarbone, the inlet is always facing forward. Thus, this personal sampler can be subjected to the excessive sampling of particles thrown directly into the inlet. Like for the 37 mm cassette, the IOM has been tested when rotating on a mannequin in moving air by Kenny *et al.* (1997), more recently in calm air by Kenny *et al.* (1999), and with the Simplified Test Protocol by Aizenberg *et al.* (2000a).

In moving air, the results presented by Kenny *et al.* (1997) show that the sampling efficiency curve is quite close to the inhalable convention curve when the sampling efficiency (following the IOM protocol, that is weighing the filter and the cartridge) is presented direction-averaged (means that there is no specific direction referred to Figure 10. In calm air, the IOM direction-averaged sampling efficiency curve is significantly above the inhalable convention curve but is close to the low wind inhalability curve as proposed by Aitken *et al.* (1999).

Due to its geometry, and particularly the open inlet, which protrudes, the IOM is subjected to oversampling, meaning that the IOM sampling efficiency curve is above 100% efficiency curve and then overestimates the true concentration. This has been recently well documented by Roger *et al*. (1998) and Li *et al.* (2000) in laboratory experiments conducted in moving air. Also, in a recent field study, Lidén *et al.* (2000) have shown that the IOM exhibits a significant degree

of oversampling that is attributed to the passive sampling (due to the open inlet). This magnitude of the passive sampling depends on the dust source and the particle sizes. Witschger *et al. (*2002a) have shown that when operating in very slowly moving air and facing the dust source, the IOM sampling efficiency (following the IOM protocol) is well above the 100% sampling efficiency curve for all particles sizes between about 7 μm up to 77 μm. The bias in concentration relative to the 100% efficiency curve has been estimated to be +30% for a polydisperse aerosol with a AMAD of 5 μm (GSD = 2), and +43% with a AMAD of 5 μm (GSD = 2). Also, it was demonstrated that the transmission (or penetration) efficiency curve (see 3.2.1) is about 80% at 7 μm and decreases with size. Moreover, like the filter cassette, the IOM sampler results show large dispersion.

Altogether, that makes the IOM sampler not really adequate for studies that need to analyse the aerosol collected on filters (like radioactive counting).

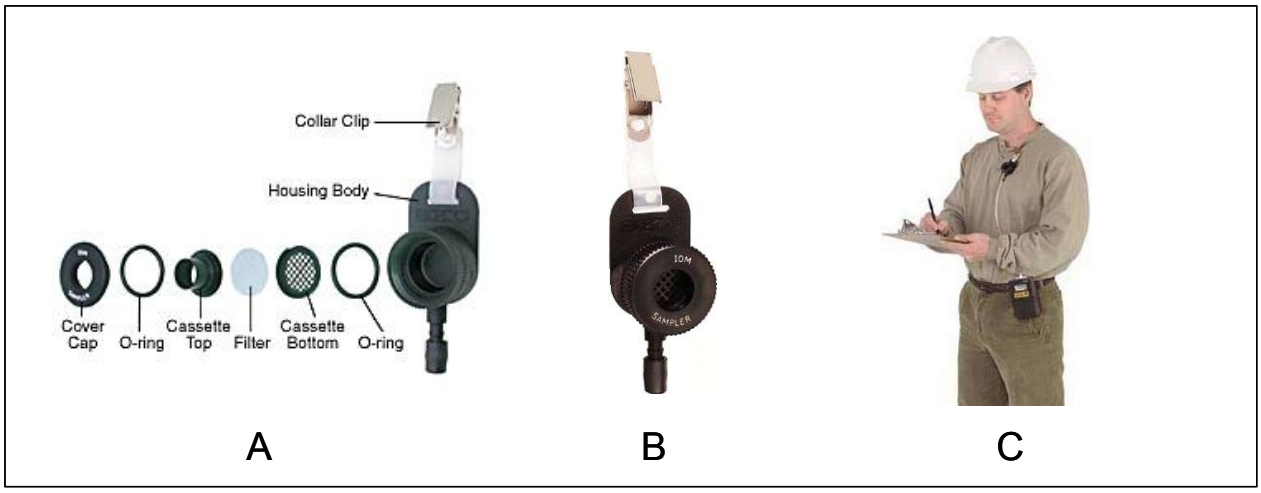

Figure 16 : The IOM Inhalable personal aerosol sampler (marketed by SKC). A: exploded view. B: as isolated with the plastic black cassette. C: placed on a human torso at the lapel level.

4.3.1.3. The Button Inhalable Sampler

Initially and recently developed by Kalatoor *et al.* (1995) the Button Inhalable sampler is presented on Figure 17. The Button sampler is a personal sampler with an aluminum body and with a porous metal screen-like inlet. Its screen is curved and has a subtended angle of 160° and a porosity of 21%. The special arrangement with the numerous 381 μm diameter evenly spaced orifices on the screen produces, at the working flowrate of 4 L/min, a very uniform deposit with the particles collected on the entire exposed area of a 25 mm filter placed directly behind. Aizenberg *et al*. (2000b) have shown that the Button possesses interesting sampling performances like the absence of transmission losses (due to the design) and a low sensitivity to direction and velocity of the incoming moving air (due to the screen). Also, the screen reduces the oversampling due to large particles (like projections).

This sampler is also used for bioaerosol sampling where it was shown as suitable for enumeration of total airborne spores (Aizenberg *et al.,* 2000c). Therefore, it should be suitable for radioactive counting analysis too. Li *et al.* (2000) have tested the Button sampler in moving air (0.5 and 1.1 m/s) and at different orientations compared with the wind. However, the authors used in their study a prototype of the Button sampler, which makes their results not 100% reliable.

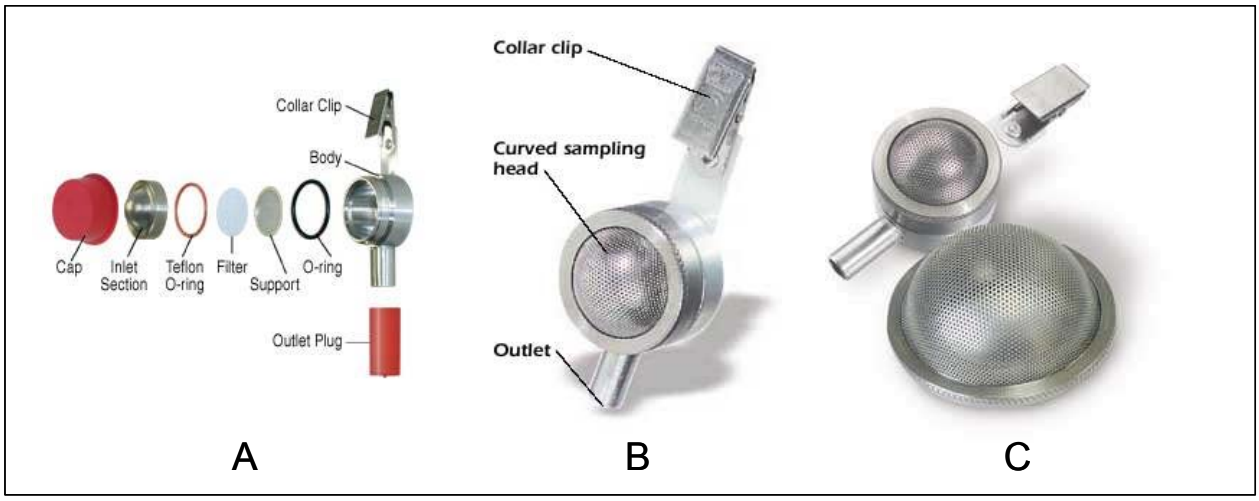

Figure 17 : The Button personal aerosol sampler (marketed by SKC). A: exploded view. B: global view. C: Abrasive blasting sampler.

The first sampling performance evaluation in very slowly moving air and near a dust source has been conducted recently by Witschger *et al.* (2002a). Here, it is clearly shown that the Button sampler has a sampling efficiency that follows very well the low wind inhalability curve (proposed by Aitken *et al, 1999*) and slightly below the 100% efficiency curve. The bias in

concentration relative to the 100% efficiency curve has been estimated to be extremely low -3% (compared to the cassette and the IOM) for a polydisperse aerosol with an AMAD of 5 μm (GSD = 2), and with an AMAD of 10 μm (GSD = 2). Although not measured, it was noticed that no deposition occurs. Moreover, the Button sampler results exhibit small dispersion (on average 3 times lower than the filter cassette). At this time, the Button sampler is certainly the most reliable aerosol sampler in the market.

Recently, the Button has been used for exposure assessment during blasting operations. During these special operations where the concentration of particles is extremely high, problems with overloading of Button screen or direct projections might occur. Therefore, it is recommended to use a sampler's shield that protects the filter from shredding or being overloaded by large particles thrust into the sampler (Aizenberg *et al.,* 2000d).

4.3.1.4. The GSP Sampler

The GSP sampler is shown on Figure 18. The GSP sampler is equivalent to the CIS sampler. The GSP has a conical inlet section with a 8 mm diameter orifice, and the working flowrate is 3.5 l/min. The whole body is molded in a conductive plastic. Once aspirated, the aerosol is collected onto a 37 mm filter that is supported by a grid incorporated in a nylon ring.

The GSP and the CIS protocols requires the filter to be weighed together with the nylon ring and therefore consider all particles that are collected onto the filter and onto the ring to be part of the sampled aerosol. However, in two studies (Kenny *et al.* 1997 and Aizenberg *et al.,* 2000a) that are presented below, the sampled aerosol was determined from the particles onto the filter only.

This sampler has been tested in moving air when mounted on rotating mannequin (Kenny *et al.,* 1997) and with the Simplified Test Protocol by Aizenberg *et al.* (2000a). Both sets of results are similar with an orientation averaged sampling efficiency close to the inhalable convention up to about 30 μm. The GSP underestimates the inhalable convention above 30 μm. However, the GSP has shown in both studies a good precision compared to the IOM or the filter cassette.

Li *et al.* (2000) have measured its sampling performances as isolated for three sampling directions to the incoming moving air (0, 90 and 180°). The same tendency as in the previous study is observed. However, this study shows clearly that the particle losses inside the conical section is significant, especially for the 0° orientation (face to the wind) and for particles larger than 20 μm. Losses in the GSP is due to sedimentation as the average air velocity rapidly decreases when entering in the sampler (due to the conical shape).

Kenny *et al.* (1999) have also incorporated the GSP sampler in their study in calm air onto a rotating mannequin. The sampling efficiency was found to be close to 100% and quite stable

until about 20 μm. Above that limit, the sampling efficiency decreases but stay above the results obtained in moving air until about 50 μm. The observed differences between those results are attributed to the fact that deposition onto the ring is in that case taken into account.

It is clear that the GSP and the CIS samplers need more investigations, particularly in very slowly moving air.

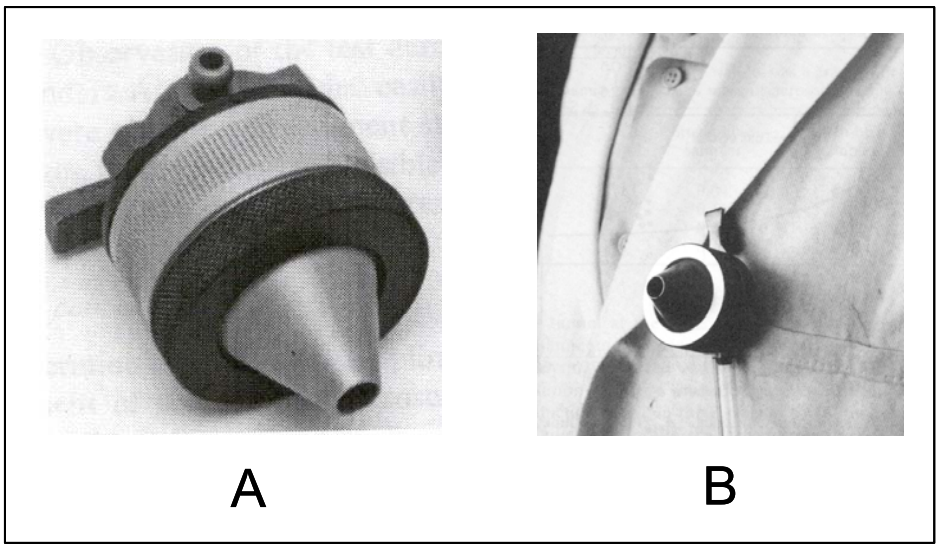

Figure 18 : The GSP sampler (equivalent to the CIS Inhalable Sampler – marketed by BGI). A: global view. B: placed on a human torso.

4.3.1.5. The PAS 6 Sampler

The PAS-6 sampler is a sampler, which is similar in its shape to the GSP, and the CIS sampler. It is an all-metal sampler that collects particles onto a 25 mm filter. The aerosol is aspirated at a 2 l/min flowrate through a 6 mm inlet orifice. The PAS 6 sampler is positioned on the collar bone and the orifice hangs downward, similarly to the filter cassette. It seems that the PAS 6 has only been tested in moving air on a rotating mannequin by Kenny *et al.* (1997). For a wind velocity of 0.5 m/s, the orientation averaged sampling efficiency stays around the inhalable convention up to about 30 μm. Also, the authors indicate that the PAS 6 sampler was found to be more precise than the IOM and 37 mm filter cassette and less than the GSP.

It is clear that the PAS 6 sampler needs more investigations, particularly in very slowly moving air.

4.3.2. Thoracic and Respirable Cyclonic Samplers

Methodology for the sampling of thoracic and respirable fractions in the occupational hygiene utilizes mostly the influence of the centrifugal forces for the particles separation. In the cyclonic samplers, the aerosol stream is drawn into the sampler through a tangential inlet, flows in spiral pattern down inside of the cone walls, reverses direction, spirals upward around the cyclone axis and through an upper centrally located exit. The finest fraction of the aspirated aerosol is finally collected usually onto a filter located above the exit. The larger fraction is impacted onto the inside walls of the cyclone and fall into a cup located downward. Therefore a cyclone gives birth to two aerosol fractions. A number of cyclones exist in the world. They differ by their design and size, some of them (big) are devoted for static sampling, and others (small and lightweight) are dedicated for personal sampling. It is the case for the two selected cyclones presented in Figure 19.

The GK 2.69 cyclone was developed through recent research into a family of tangential flow cyclones by Kenny and Gussman (1997). The GK 2.69 has two versions. At a flowrate of 4.2 l/min, the GK 2.69 is devoted to sample the respirable fraction, while at a flowrate of 1.6 l/min, it is used to sample the thoracic fraction. The aerosol is collected onto a 37 mm filter.

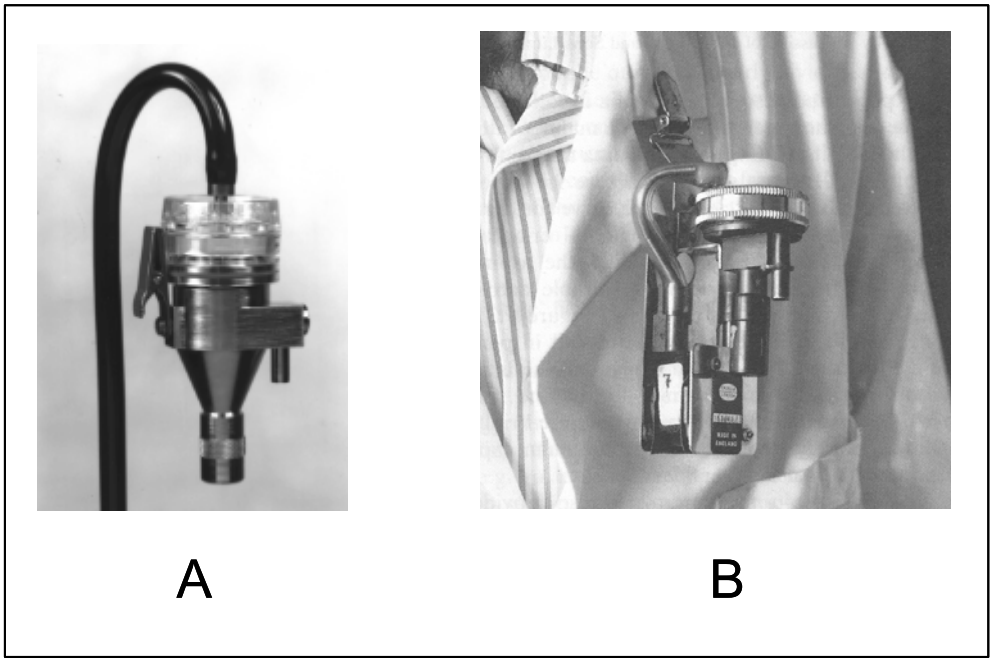

Figure 19 : Cyclonic samplers. A: The GK 2.69 Respirable/Thoracic Cyclone (marketed by BGI). B: the 1.9 l/min Casella Respirable Cyclone (marketed by Casella)

Under calm air simulated conditions in laboratory experiments, Maynard (1999) found that the GK 2.69 cyclone at the working flowrate of 1.6 l/min is in close agreement with the thoracic convention. His works gives functions that can used to model the sampling efficiency of the cyclone. The estimated bias in concentration relative to the thoracic fraction is within the range 0 to +10% for AMADs less than 20 μm and GSDs less than 2.

Görner *et al.* (2001) have presented a study focused on 15 cyclone samplers devoted to measure the respirable fraction. Among the samplers, the Casella cyclone in its plastic version with the working flowrate of 1.9 l/min has been tested. The sampling efficiency is close to the thoracic convention. The estimated bias in concentration relative to the thoracic fraction is within the range -20 to +10% for AMADs less than 10 μm and GSDs less than 3.5.

4.3.3. Environmental Samplers

The sampler presented on Figure 20 is a lightweight personal sampler, which is devoted to measure the PM 2.5 and PM 10 according to the curves presented in Figure 9. The Personal Environmental Monitor (named PEM) consists of a single-stage impactor followed by a filter to collect airborne particles for mass, chemical or radioactive analysis. Aerosol is sampled through the impactor to remove coarse particles larger than the impactor cut-point. Cut off diameters of 2.5 and 10 µm are available for personal PM2.5 or PM10 sampling. Sampling flow rates of 2, 4 and 10 l/min are available.

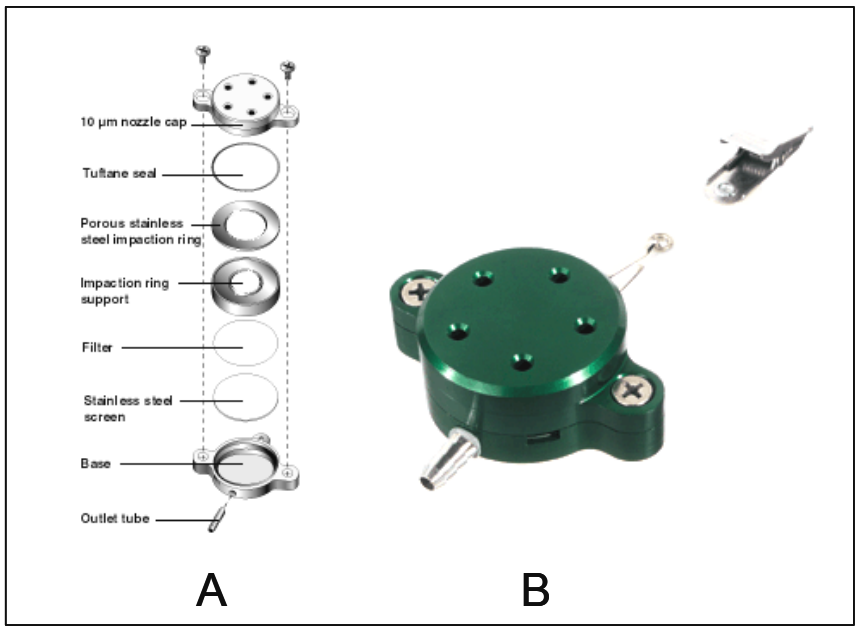

Figure 20 : The Personal Environmental Monitor for measurement of PM10 or PM2.5 in indoor air (marketed by SKC).

Only very limited data have been found regarding the sampling performance of this sampler which is primary devoted for indoor or outdoor personal exposure assessment. Rodes and Wiener (2001) present a graph that indicates the PM2.5 with a flowrate of 2 l/min to be suitable for measuring PM2.5 fraction. According to our knowledge, no more data are available. It is clear that the PEM sampler in its different versions needs more investigations, particularly in very slowly moving air.

4.4. Area aerosol samplers

For many years, the only available and tested area aerosol sampler intended to sample according the inhalable convention was the IOM static inhalable sampler shown on Figure 21. Developed by Mark *et al.* (1985), this sampler was designed to sample at a 3 l/min flowrate that is low for a static sampler. It uses a continuously rotating entry orifice. While used in in Britain, this sampler has never crossed the channel to be used in other European countries.

Figure 21 : The IOM static inhalable aerosol sampler (Vincent, 1989).

A more recent static sampler is the French CATHIA static sampler (French acronym for: thoracic, inhalable, and respirable aerosol sampler). Originally developed at the Institut National de Recherche et de Sécurité (INRS) in France by Fabriès *et al.* (1998), the sampler is a variant of the CIP-10 French personal sampler (widely used in the mines for respirable fraction measurement). The key feature of the CATHIA sampler is the fact that it can be used for measuring the inhalable, thoracic and respirable fraction by easily changing the particle size selector and the aspiration flowrate. The sampling inlet is the same for the three different particle size selectors, and consists of an annular slot designed to follow the inhalable convention (Görner *et al.,* 1996). Thus, the sampler is based on the concept that the thoracic aerosol and the respirable fraction are sub-fractions of the inhalable fraction. Sampled particles leaving the selector travel through a tube down to a 25 mm diameter filter. The tubing length was optimized in order to insure a uniform particle deposit on the filter surface. Figure 22 presents the global view and the schematic of the particle size selector, which is used to sample according to the inhalable convention with a flowrate of 10 l/min. The sampling performance of this new version of the static inhalable sampler has not been yet fully evaluated. Therefore, it is

clear that the static CATHIA inhalable sampler needs more investigations, particularly in very slowly moving air.

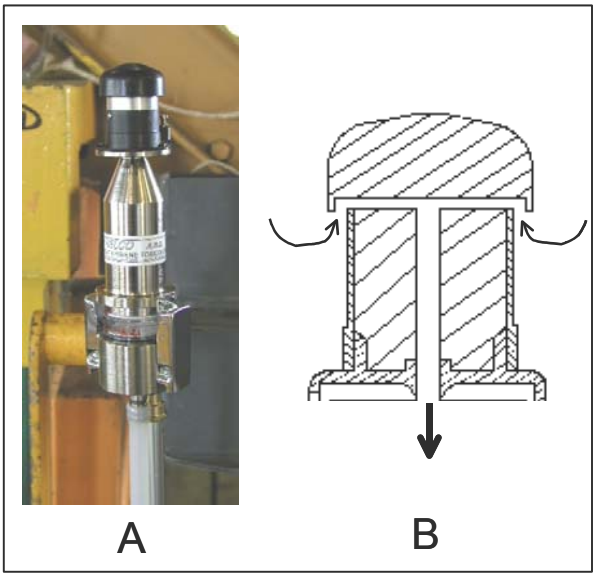

Figure 22 : The CATHIA static inhalable aerosol sampler. A: global view. B: schematic diagram of the particle size selector.

The only static sampler standardized in France is the sampler presented in Figure 23. This sampler (usually named in France, *"AFNOR sampling head"*) consists of an annular omnidirectional slot operating at 25 l/min (AFNOR, 1988b). Once aspirated, the aerosol stream flows inside a vertical tube of 30.5 mm inner diameter up to a 47 mm diameter filter. At this time, no published data are available on the sampling performance of this static sampler. Therefore, it is clear that the static "AFNOR sampling head" sampler needs more investigations, particularly in very slowly moving air.

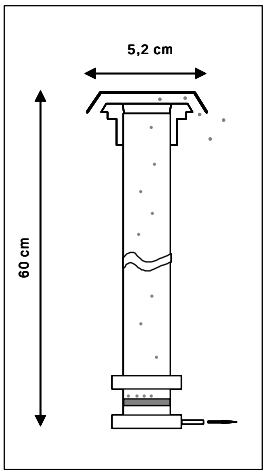

Figure 23 : The AFNOR static aerosol sampling head (French standard NFX43-261)

Several static samplers are available in the market, like for example the one shown in Figure 24. This sampler operates at 10l/min and uses a single stage impactor to remove the unwanted aerosol fraction and subsequently sample either the PM10 or PM2.5 fraction on a 37 mm diameter filter.

Figure 24 : The Micro-Environmental Monitor for PM10 and PM2.5 (marketed by SKC)

One should be careful when using a given static aerosol sampler, as most of the time no data are available on the sampling performance of static samplers. This is particularly due to the fact that in the vast majority of the field studies performed in the non nuclear sector, personal samplers are used, but not often static samplers. A typical example in the nuclear sector is the APA (well known in France) static sampler, for which no available sampling performance data exist.

4.5. Aerosol spectrometer

A full description of the dosimetry of inhaled aerosols requires information, often unappreciated, about the particle size distributions. In particularly the information should be in the form of *aerodynamic diameter*. Therefore, aerosol spectrometers are more versatile that the aerosol samplers that are used routinely in industrial hygiene exposure assessment, which are usually dedicated to a given fraction (inhalable, thoracic or respirable). Among the different options, the cascade impactors are the most useful for the sampling and the classification of particles in the range of particle aerodynamic diameter between about 0.3 (0.05 for the low pressure version) to about 20 μm (Hering, 1996). Here only three devices are mentioned.

Figure 25 presents two well known and used cascade impactors.

On the left side is presented the Andersen 8-stage impactor is certainly the world's reference impactor. It is a static sampler that operates at 28.3 l/min and collects particles onto 8 stages well characterized by their cut off diameter. Coarser particles are stopped on the first stages while the finer particles are stopped on the last stages. It is not the scope of the present document to expose all the careful points related to the use of the impactors. Therefore, the reader is invited to read carefully well known reviews like the one by Hering (1996) or the AIHA practical publication (1995).

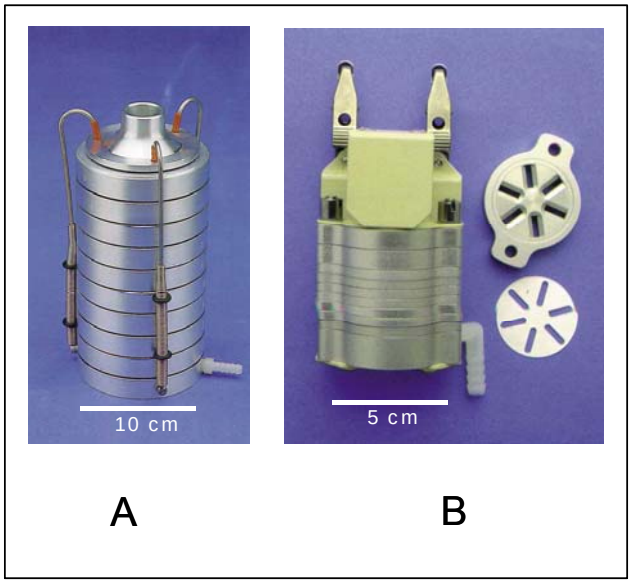

Figure 25 : Cascade impactor. A : the Andersen 8-stage cascade impactor. B: the Marple 290 personal cascade impactor.

On the right side of Figure 25 is also presented a personal version of the cascade impactor: the Marple 290 personal cascade impactor. This small impactor is an 8-stage device that operates at 2 l/min. It has four slot-shaped jets at each stage, where aerosol is collected onto specially designed polycarbonate membrane films. An other personal impactor (not presented here) is the personal inhalable dust spectrometer (referred as to PIDS). Originally developed by the IOM, its performance was found to be in agreement with the inhalable convention (Ramachandran *et al.,* 1996). Therefore, it makes this device attractive as the measurement gives the particle size distribution in aerodynamic diameter and also the total sampled activity (by summing all the acitivities measured on each stage).

Figure 26 presents a new instrument, which is called the Respicon™ particle sampler. This sampler is intended to sample at the same time the three conventional inhalable, thoracic and respirable fractions. This sampler combines inertial classification and filter sampling. The aerosol is aspirated through an omnidirectional slot. The inertial classification is made with three virtual impactor stages in series. The first stage collects particle smaller than 4 μm, the second collects particle between 4 and 10 μm, and the last stage collect particles above 10 μm. The working flowrate is 3.1 l/min. An improved version combines also a direct aerosol concentration measurement using aerosol photometry (three light scattering photometers). Measurements that have been performed in field shown that the instrument is practicable under rough industrial conditions (Koch *et al.,* 1999).

Figure 26 : The Respicon™ Particle Sampler (marketed by TSI)

Li *et al.* (2000) have performed tests to evaluate the capability of the Respicon™ (the filter version only) to measure the inhalable fraction. Experiments performed in moving air at a wind

velocity of 0.5 m/s show that the Respicon™ matches quite well the inhalable convention, if a correction factor of 1.5 is used, i.e. the result given by the Respicon™ should be multiplied by 1.5 to get the true result. Moreover, the authors have noticed particle deposition inside the sampler. They advise to carefully monitor the unit cleaning to prevent plugging of the connecting tube between the first and the second stage.

An other approach for measuring the size distribution uses polyurethane foams placed in series. For example, Vincent *et al.* (1993) presented a work toward the realization of practical sampling devices based on the use of foams. This work has resulted in an improved version of the IOM inhalable sampler for the simultaneous measurement of exposure to inhalable, thoracic and respirable aerosol fraction. However, this type of sampler needs a specific quantitative analysis for extracting the collected particles from the foams without losses.

4.6. Direct-reading devices

All the aerosol samplers presented in the previous chapters are suitable only for time-averaged measurement. Sometimes, there is a need for information about the real time exposure and not only the time-averaged exposure. It is particularly the case when the aerosol is thought to be highly hazardous. Here, an immediate alert to high concentrations is required. Also, it is the case for monitoring in order to examine the effects of adjustments in process or dust control. In these defined situations, the direct-reading, or rapid, devices, are of particular interest as there are variations of the aerosol concentration and particle size distribution with time in workplace. It is reminded that these variations are caused by many factors like forced ventilation in indoor environments, convection in warm environments, wind when outdoor or due to the worker itself.

Direct-reading field instruments for aerosol measurements usually determine total count or mass, and particle size distribution. They are a combination of a sampling instrument and an analytical instrument. Therefore, all the sampling considerations exposed in the previous chapters that can lead to bias in the aerosol concentration or the particle-size distribution are to be taken here into account too. Bias may come from the entry (under- or over-sampling) or from the particle deposition in the lines of the instrument up to the sensing zone. For the user, the instantaneous readout provided by the direct-reading devices often efface this sampling problem. However, it should not be forgotten when analyzing data from these instruments. Moreover, the measurement principles used in the direct-reading devices are usually complex, and therefore, great caution is recommended in choosing a device to perform a particular task for there are many potential traps that are not always seen.

The majority of the instruments fall into five categories: optical, electrical, molecular, mechanical and nuclear. The devices that find an application in the industrial hygiene are the light scattering photometers, the optical particle counters, the condensation nuclei particle counters, the piezoelectric mass balance or the nuclear mass detectors. For a comprehensive review of all of these instruments, it is recommended to read the guide edited by the ACGIH (2001). But surprisingly, within the industrial hygiene community, the use of these direct-reading devices is not well established.

In the following, only two examples are presented.

The first direct-reading device taken as an example is the Grimm spectrometer, shown on Figure 27. It is an instrument that counts and sizes particles using scattered light information from particles illuminated by a laser. In its 1.108 version, the Grimm aerosol spectrometer sizes particles in 15 different channels from about 0.3 μm up to 20 μm, and displays data within six seconds intervals. The measurement made by the Grimm is therefore the time evolution of the count distribution as a function of an optical diameter. The optical diameter can differ from the aerodynamic diameter. Therefore, it is well recommended to calibrate the channels of the Grimm against either calibrated latex particles or other well defined particles. However, the Grimm has an interesting feature: all sampled particles are collected on removable filter for subsequent analysis. Thus, it is possible to compare the time-averaged concentration obtained from the measurement of the collected amount of particles on the filter with the time-averaged concentration obtained from calculation with the stored concentration data, and finally define a “calibration factor”. However, this calibration factor will be representative only of the aerosol that has been sampled and should not be used with an other aerosol type.

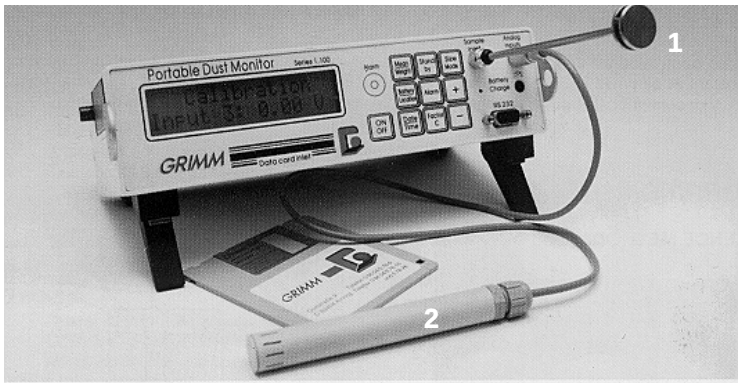

Figure 27 : The Grimm G 1.108 aerosol spectrometer (marketed by GRIMM Technologies, Inc.). 1: Omnidirectional aerosol inlet. 2: Temperature/Humidity sensor.

The aerosol is aspirated through an omnidirectional inlet at a flowrate of 1.2 l/min. As a small light-weighted device, the Grimm spectrometer is an attractive device for monitoring in the workplace the time evolution of the particle size count distribution.

The second instrument taken as an example is a photometer and is presented on Figure 28. The key feature of the Haz-Dust III™ is its portability, making this device a personal photometer. The measurement of aerosol concentration using photometers is based on detection of scattered light by particles simultaneously present in the sensing volume of an optical cell. With the photometers, for the determination of the relative concentrations, the composition of the aerosol (particle size distribution and refractive index) must be constant during the measurements. For absolute measurements of mass concentration, the photometer must be calibrated with the aerosol to be investigated (Görner *et al.,* 1995).

Figure 28 : The Haz-Dust III™Particulate Monitor (marketed by SKC)

A key feature of the Haz-Dust III™ is to have a detachable optical sensor that can be connected in line with different samplers like a 37 mm filter opaque cassette or a cyclone or the IOM inhalable sampler. Thus, a calibration is possible by comparing the response of the device with the mass concentration obtained by analyzing the collected aerosol on the filter. However, there is no available publication presenting the real performance of this new device that is intended to record the respirable, thoracic or inhalable mass fraction.

5. FILTRATION AND QUANTIFICATION OF THE SAMPLED AEROSOLS

Aerosol samples are collected most of the time onto filters. The quantification of the sampled and collected aerosols onto filters can be performed using different methods: gravimetric analysis, chemical analysis or direction radiation counting. The latter is obviously the most adapted method in the radiation dosimetry context.

It should be recalled (see in the previous chapter 4.3) that transmission losses may lead to negative bias in the estimation of the sampled concentration; these losses are not taken into account in the analysis. Therefore it is preferable to choose an aerosol sampler that does not exhibit transmission losses.

5.1. Gravimetric analysis

The measurement of the amount of collected particles is usually performed by weighing the filter on an analytical balance, before and after the experiment. Gravimetric analysis requires a high degree of stability in the environmental conditions in the room where the filters are weighed (particularly the moisture). It is recommended that a number of blank filters (minimum three) are weighed with filters devoted for the measurement. The average variation in mass of the blank filters is then used to compensate for the mass variation of the sample filters.

5.2. Chemical analysis

The measurement of the amount of collected particles can also be performed using specific techniques like, in the nuclear sector, reduction to ash or dissolution for analysis by analytical chemistry or radiochemistry.

5.3. Direct radiation counting

The activity can be measured directly by using radiation counting methods. When alpha particle spectroscopy or alpha total counting is applied, membrane filters with their superior front-surface collection characteristics are preferred over fibre type filters. Although it is not well

documented, the penetration of particles into the filter matrix is a function of the type of filter, and this has a important effect on the radiation detection efficiency (Grivaud and Fauvel, 1996).

Membrane filters have the advantage that they can retain particles effectively on their surface (an advantage for alpha counting and also for optical microscopy), whereas fibre filters have the advantage of providing high loading capacity (an advantage for gravimetric analysis). However, the choice between both should also take into account the pressure drop effect, as membrane filters usually have a higher pressure drop than the other filters.

Most of the time, the collection efficiency of the filters is not an issue as in the range of particles dimension encountered in workplaces, the collection efficiency is usually close to 100%. However, if membrane filters with great pore size are used (like the Nuclepore filters) for pressure drop requirements, some reduction in the collection efficiency can take place.

Some of the samplers, use foams as collector or particle size selector. The foams are usually formed from reticulated polyurethane with a structure consisting of a matrix of bubbles with connection between them. Such samplers cannot be used for alpha counting as the particles are deeply retained inside the foams. However, radioactive measurements could be performed if a reduction to ash method is used.

For some types of filters (like PVC or PTFE), electrostatic charge can present aerosol collection and handling problems, particularly when working in low humidity environments. It is recommended to use a source of bipolar ions to neutralize the sample before weighing.

In the samplers like the impactors, particles are collected onto impenetrable impaction substrates (metal plate coated with a very fine layer of oil). Also, it can be hypothesized that due to the way of the collection of the particles in impactors, there is no penetration concerns even with filters are used as a impaction substrates.

6. CONCLUSION

Protection of workers against inhalation of radioactive aerosols is receiving considerable attention as part of the overall emphasis on the minimization of various occupational exposures. Recognizing the importance of this issue, a collaborative research effort is being conducted through the European ALARA Network community to "…*improve the quality and accuracy of internal dose monitoring techniques".* As a result, a European project (SMOPIE) started in November 2001. As part of the work package n°4, the present review of the monitoring devices and methods to be used in aerosol sampling studies in workplaces for exposure assessment has been made.

The development of a reliable data base on size-selective particle deposition in the human respiratory tract has enabled recently the establishment of a truly scientific rationale for the specification of sampling criteria. However, several problems still remain with the implementation of the inhalable convention (wind dependence, orientation averaged, unspecified above 100 μm). This implies that, in the longer term, the inhalable convention needs revision. But to start an effective discussion, further experimental investigations from different laboratories should be carried out to bring new data.

Aerosol sampling techniques, and especially personal sampling techniques, intended for evaluation of exposure have undergone marked evolution over the past years in the direction of a better sampling performance compare with health-related sampling criteria. However, there are still sampling techniques with bad performances that are used in the *industrial hygiene* world.

To conduct a study for sampling performance evaluation still appears to be a big time consuming and costly challenge. Therefore, studies should be carried out to develop and compare new (but simple and cheap as well) sampling performance tests that insure accuracy and reliability. As a result, performance evaluation of existing samplers could be carried out more frequently, and new samplers developed.

The closed-face filter cassette is now known to be not a reliable personal aerosol sampler, having poor sampling performances and large dispersion. Traditionally, this sampler has been used widely in the workplaces, but it should no longer be used in the future. It is therefore important to conduct field studies in order to better understand the relationship between this sampler and the more recent inhalable samplers like the IOM, the Button or the GSP, like for example the recent one's conducted in the wood industry by Tatum *et al.* (2001).

For the inhalable samplers, it seems that the GSP and the PAS 6 samplers needs further sampling investigations particularly in very slowly moving air, thought to be more representative of real workplace environments.

Direct-reading instruments are very attractive devices as providing more rapid measurements with less effort (and cost) than the traditional approach using the filter collection. However, care should be taken as these optical based instruments (like photometers) may lead, if not well calibrated, to erroneous estimations of the exposure, especially if large particles (above 10 μm) are involved. Therefore, there is a need to develop instruments that extend their application on the large particles, like to estimate the inhalability.

A continuing need exits for simple, cheap and reliable personal samplers. Also, a great deal of progress should be done in reducing the dimension and the weight of the pump to be used with the personal samplers, but to increase their capacities with working flowrate up to about 10 to 15 l/min. The higher flowrate would make the personal aerosol samplers acceptable for very low aerosol concentration or useable with short sampling duration. At this time, the pump flowrate is limited with rarely greater than 4 l/min.

Also, an emerging issue that will grow significantly in the next future concern the measurement of very, very small particles, with diameter less than 0.1 μm (also called ultra fine particles). This measurement requires a specific instrumentation that departs from the traditional sampling approaches that have been presented here. Because, the evidence seems to be that for ultra fine particles, the appropriate health-relevant metric is the number concentration rather then the mass, it is thought that the most promising instrumentation, adapted for measuring in the workplace conditions, would be based on the recent development made for the nano-particles technology.

The particular situation of the determination od internal radiation doses (presented in 3.1.2) imposes that in the context where the aerosol particle size distribution is perfectly known an ideal aerosol sampler would be an aerosol sampler having a 100 % sampling efficiency for all particle sizes. This sampler does not exist in the market. It is therefore recommended to select an aerosol sampler with a very well defined sampling efficiency that is not dependent to factors like external wind, orientation etc., and to associate with the concentration measurement a measurement of the particle size distribution in order to estimate the corrective R factor to be applied for the determination of the ambient concentration (see chapter 3.1.2). In the case of estimating the ambient concentration in absence of particle size distribution measurement in the workplace, it is recommended to go for an aerosol sampler which shows an R factor not strongly AMAD dependent. It is particularly the case of some inhalable aerosol samplers that have been presented in chapter 4.3. In absence of, particle size distribution measurement, the thoracic samplers seem to be a reasonable alternative, as being not much dependent of external factors

like wind or orientation to the source, but it should be used carefully because as soon as the AMAD is above about 5 μm, the R factor to employ is strongly “AMAD dependent”.

Finally, in a situation of imprecise (or uncertain) particle size measurement, a generic method has been presented in chapter 3.1.2, aiming at identifying the aerosol sampling fraction that minimises the impact of this uncertainty on the estimated effective dose, taking into account the AMAD dependency of the considered compound dose coefficients and the estimated AMAD of the aerosol particles.

The review is based on the analysis of about 70 scientific publications (published scientific papers, books and guides), with about 40 being less than 5 years old, and 60 being less than 10 years old. Altogether, the results and their analyses presented of this review, and its implication in the SMOPIE project should benefit any industry from the nuclear or non-nuclear sector that have or may have potential occupational exposures to radioactive aerosols.

The author (now at Laboratoire de Métrologie des Aérosols, INRS, Nancy, France, email: olivier.witschger@inrs.fr) would like to thank Jean-Pierre Degrange from CEPN for his careful reading and critique of this review, which in particular provided most of the development of the chapter 3.1.2.

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