Adequacy of Existing Aerosol Samplers for Monitoring NORM Exposures

O. Witschger

Institut National de Recherche et de Sécurité, Vandoeuvre, France

1. Abstract

While for years now, many efforts have been made in the optimisation to keep external radiation exposures as low as reasonably achievable (ALARA), very, very few efforts have been devoted to put into practice the ALARA approach for internal exposures. However, in many workplaces, the most significant exposure pathway is the internal exposure via inhalation of particulate airborne contaminants. In particular, it is the case for the industries involved with naturally occurring radioactive materials (NORM) or for the nuclear fuel handling 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 to assess 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 recently brought to the front with the publication of the Council Directive 96/29/EURATOM (1996).

For radioactive aerosols, the use of the aerosol sampling as a method for internal dose (via inhalation of radioactive particles) assessment is in debate 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 in term of dose.

2. Aerosol sampling in the industrial hygiene context

In the general context of industrial hygiene, the aerosol sampling is most often accomplished in order to estimate the exposure (via inhalation) of the concerned persons and to compare the measured concentrations to the permissible exposure

Marilee Williams, Editor
Proceedings of the 9th EAN ALARA Workshop
18-21 October 2005
Augsburg, Germany

limit to the considered air contaminant, expressed as time-averaged concentration values for a given conventional particle-size sampling fraction.

Figure 1 shows the three conventional sampling fractions (inhalable, thoracic and respirable) internationally agreed 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)).

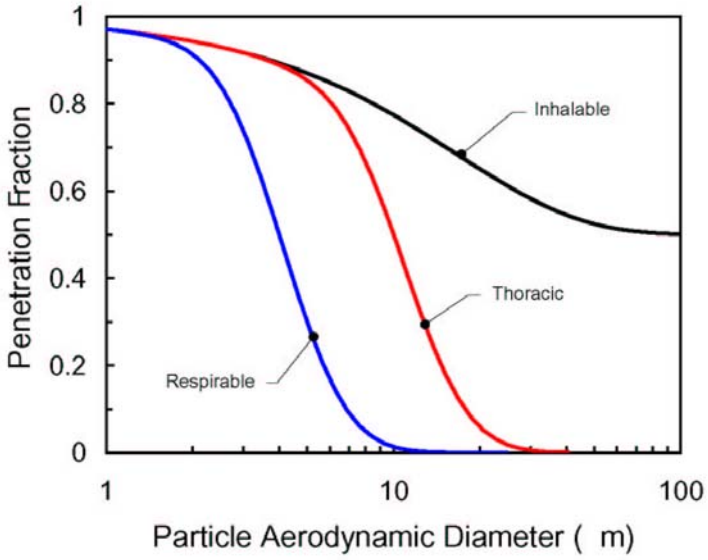

Figure 1. Particle size fractions (i.e. inhalable, thoracic, respirable) for health-related sampling in workplaces that have been internationally agreed by CEN, ISO and ACGIH.

These curves are used as a guidance to assess worker exposure, depending on the relative toxicity of the air contaminant deposited in each pulmonary region. These curves are also used as a reference for the development of aerosol sampling systems as they constitute internationally agreed target sampling criteria.

3. Aerosol sampling in the radiological protection context

The situation is somewhat different when dealing with radioactive aerosols, as in the NORM or the nuclear industries as the primary component to assess is the effective dose, and secondly 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 (or total aerosol). Thus, it is desired to sample the true 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%.

To illustrate the implication of the importance to well know the aerosol sampler performance and the particle size distribution, calculations have been made, with the results shown in Figure 2.

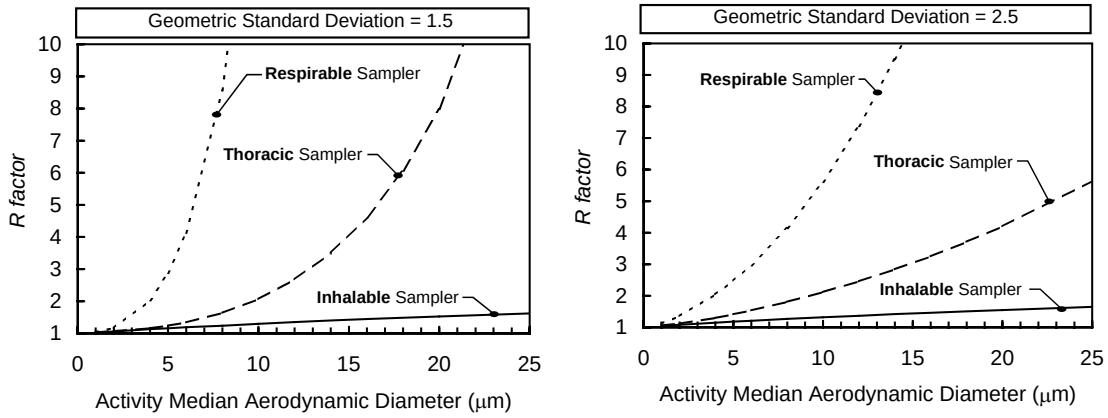

Figure 2. R_X factor to employ for the estimation of the true total (or ambient) aerosol concentration from the measured aerosol concentration corresponding to the inhalable, thoracic or respirable fraction, as a function of the activity median aerodynamic diameter (AMAD) and for two geometric standard deviations (GSD).

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 characterised 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 from the measurement of the C_X. 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.

$$C_{AMBIENT}(AMAD, GSD) = R_X(AMAD, GSD) \times C_X(AMAD, GSD) \qquad (Bq/m^3)$$

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 characterised by an AMAD equal to 10 μm and a GSD equal to 2.5.

Figure 2 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 analysing the data.

Also, the R_X factor can also be used to evaluate the exposure (by inhalation) X_{TRUE} (Bq) of a worker as:

$$X_{TRUE} = C_X(\text{AMAD, GSD}) \times R_X(\text{AMAD, GSD}) \times B \times t_E \quad \text{(Bq)}$$

where B and t_E represent respectively the ventilation rate of the worker (m^3/h) and the duration of the exposure (h).

When the final information is the true effective dose, the situation is more complex. The true effective dose corresponding to the inhalation of a polydisperse aerosol of a specific radioactive compound 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 $e(\text{AMAD, GSD,...})$

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.

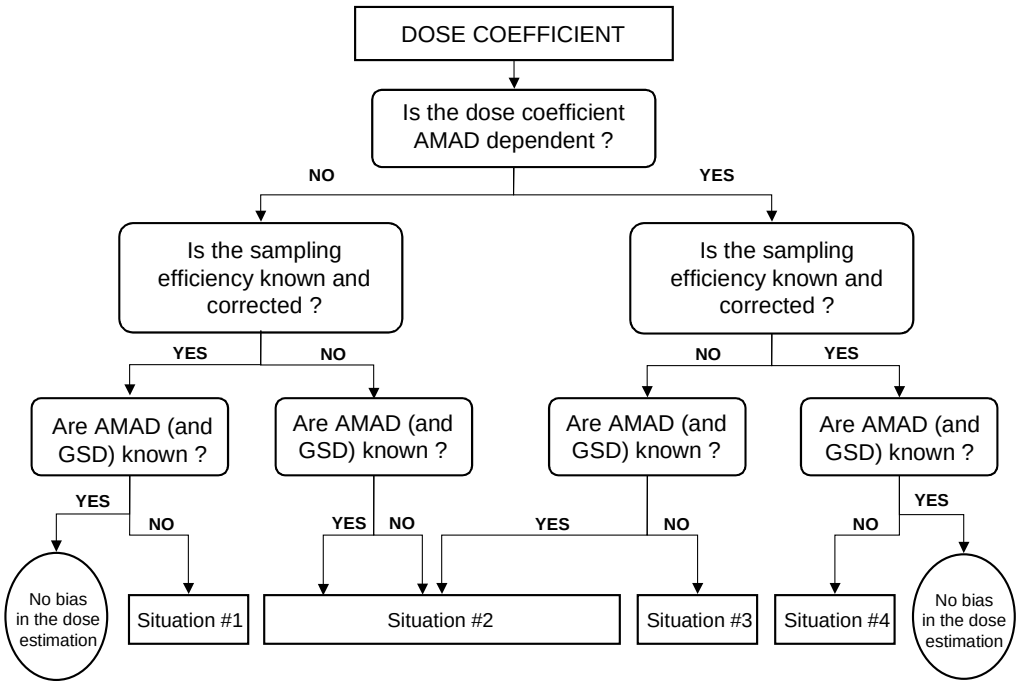

Figure 3. Schematic describing the different situations occurring in relation to aerosol sampling in the radiation protection dosimetry context, and that lead to bias in the dose estimation.

In this context, the big issue is to know which aerosol sampling technique is likely to minimise, for a given radioactive compound, the bias between the measured effective dose and the true effective dose. The answer is complex for it depends not only on the knowledge (and eventual correction) of the sampling performance of the chosen technique but also on the knowledge of the particle size characteristics (measured or considered as a default) of the aerosol (AMAD, GSD,…) and on the relationship between the dose coefficient and these latest data. Figure 3 presents the different situations that can occur and lead to different bias in the effective dose estimation.

Only two situations lead to a non-biased estimation of the effective dose, where the sampling efficiency is known and corrected for and the particle size characteristics of the ambient aerosol (AMAD and GSD) are perfectly known. Bias in the other situations may be minimised by the choice of the sampling fraction.

For example, in Situation #4 – where the dose coefficient is AMAD (and GSD) dependent – the sampling efficiency is known and corrected but the AMAD (and GSD) is not (perfectly) known, the bias may be expressed as:

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

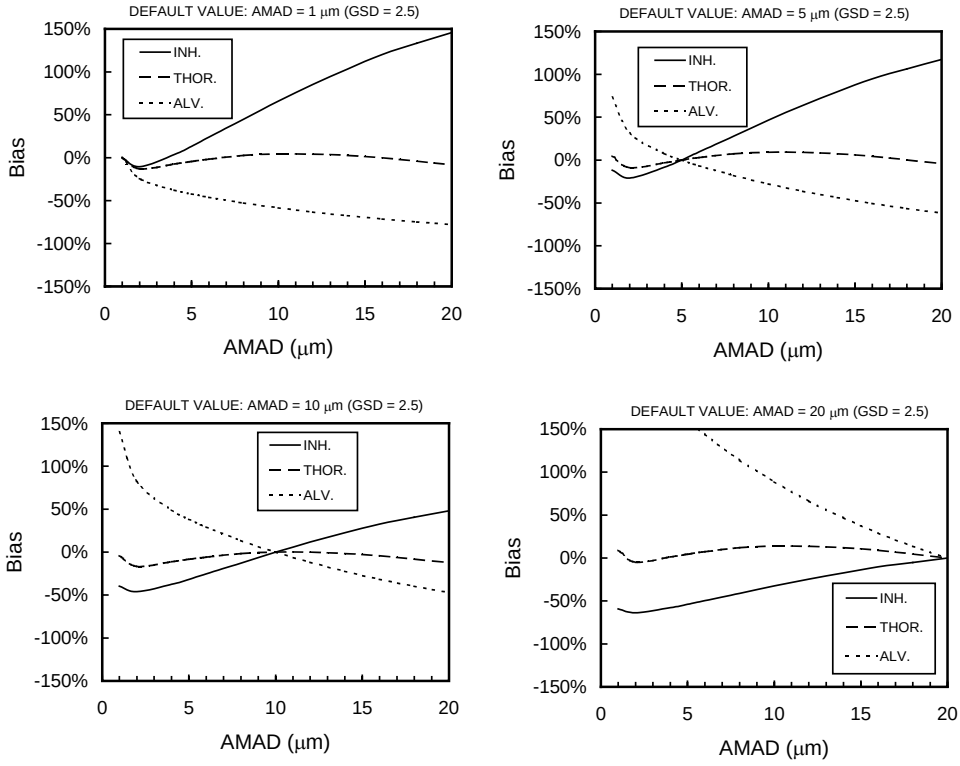

Figure 4. Bias between the estimated dose and the true dose in Situation #4. The calculations have been made for four Default AMAD (1, 5, 10 and 15 μm) and for the Default GSD of 2.5. The bias in the situation#4 depends of the radionuclide, which is considered. Therefore the calculations have been made for the intake of U_{234} by inhalation and considering a slow rate of absorption (Type S).

To illustrate, calculations have been performed and results shown in Figure 4, which compare, for Situation #4, the AMAD dependency of the bias for three ideal aerosol samplers (inhalable, thoracic and respirable) and four default AMAD values: 1, 5, 10 and 20 μm (GSD = 2.5 in all cases). 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) using the LUDEP 2.2 code (Jarvis, 1996) that implements the HRT Model for Radiological Protection (ICRP publication 66, 1994).

According to the Figure 4 the thoracic aerosol sampler is the one that minimises the bias, whatever the default AMAD considered. But as a general rule it can be easily deduced from the above equation that the sampler that minimises - for a given radioactive compound - the bias on the effective dose due to the fact that the particle size distribution (AMAD) of the ambient aerosol is not (perfectly) known, is the sampler which dependency with AMAD of the sampling efficiency follows as closely as possible the AMAD dependency of the considered compound dose coefficients. Or in other words, to minimise the impact on the effective dose of the uncertainty associated with the particle size distribution (AMAD) of the ambient aerosol, (whatever the true AMAD of the ambient aerosol and the value of AMAD and GSD considered as a default) different aerosol sampler could be chosen according to the solubility of the considered compound.

4. Aerosol samplers for measuring in workplaces

Ideally, one wishes to characterise the microenvironment in the breathing zone of the worker to evaluate its specific exposure (see Figure 5). Regarding the “strategic” question of how best to assess 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. In this paper, we are focusing on the techniques.

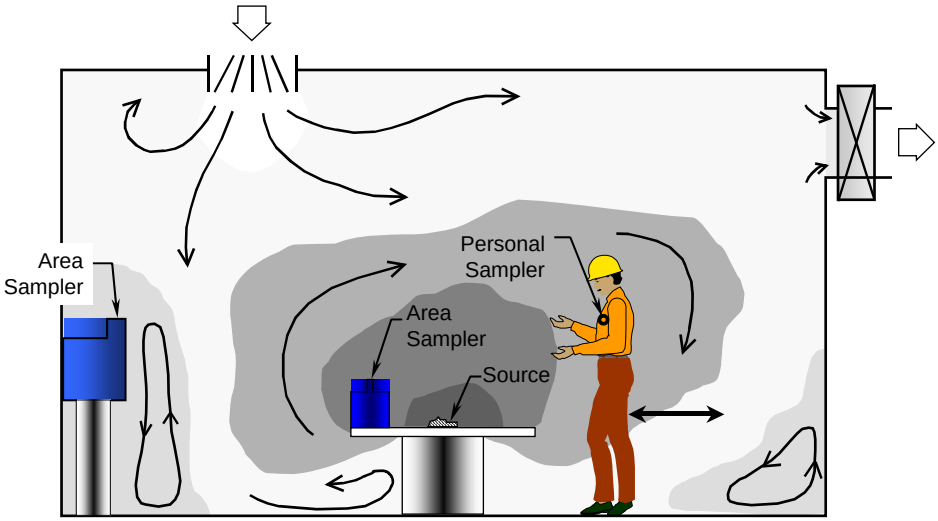

Figure 5. Illustration of the nature of the dispersion of the contamination in an indoor workplace.

There are two types of measurement that can be carried out in the workplace:

1) 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, and

2) 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 flow rates (tens of L/min), 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, there 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 centred on the mouth and nose and having a radius of about 30 cm (Vincent, 1995), as it is illustrated in Figure 6. 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

Figure 6. Location on worker of personal sampler with the predominant facing to the dust source direction.

A number of aerosol personal samplers exist now in the market, some of them being old, some new (summarised by ACGIH, 2001; Baron and Willeke, 2001; Hinds, 1999; Kenny *et al*., 1997; Maynard and Jensen, 2001; Vincent, 1995 and Witschger, 2000). But not all of these personal 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.

As a example, Figure 7 shows the average sampling efficiencies of the IOM, Button and closed-face 25-mm Millipore filter holder samplers operating in a low air movement environment facing the aerosol source measured by Witschger *et al*. (2004). The points represent our experimental data obtained for six aerodynamic particle diameters: 6.9, 14.1, 28.4, 38.7, 60.1 and 76.0 μm. Each data point was determined as an average value of at least three replicates with the standard deviation calculated for the 95% confidence interval (shown as error bars). The internationally standardised ISO/ACGIH/CEN Inhalability Convention (ACGIH, 1999; CEN, 1993; and ISO, 1995) and the recently proposed "low-wind inhalability" curve [Eq. (2), Aitken *et al*., 1999] are also plotted in Figure 7.

The comparison of the sampling efficiency of a specific sampler with the 100% level is meaningful for radioactive aerosols found in the NORM industries since the effective dose of radionuclides is assessed through the "total" ambient aerosol concentration. Comparing to this "target" level, the IOM sampler over-samples the particles of MMAD = 5 – 15 μm by 29 – 47%, while the 25mm cassette under-samples them by 33 – 67%. The efficiency of the Button Inhalable Aerosol Sampler is only slightly below 100%: the bias ranges from -3% to -12%.

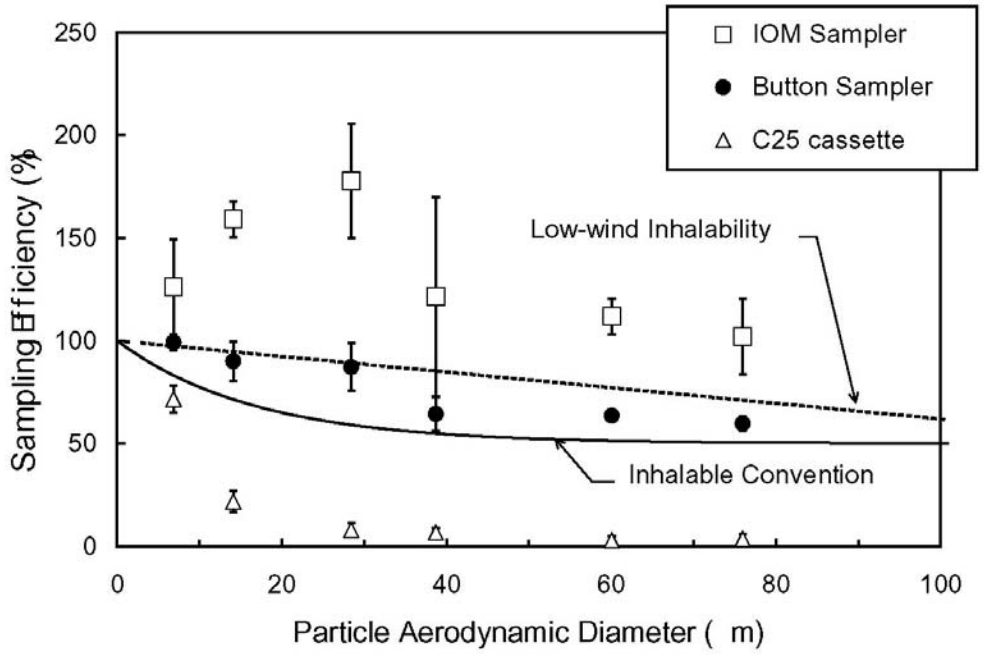

Figure 7. Sampling efficiencies of the IOM Inhalable Sampler, Button Personal Inhalable Aerosol Sampler, and 25-mm closed-face cassette in very slow moving air near the source (Witschger *et al*. 2004). Average values are presented with their 95% confidence interval. The solid curve represents the international Inhalability Convention (ACGIH, 1999; CEN, 1993; and ISO, 1995); the dotted line represents the Low-wind Inhalability curve (Aitken *et al*., 1999).

5. Conclusion

Protection of workers against hazardous airborne dusts has received considerable attention as an important part of the overall objective to minimise occupational exposures. The airborne aerosol sampling in workplaces (personal and stationary) has become a key issue in occupational hygiene, since the data collected on the particle size and concentration are used by regulatory agencies for exposure and risk assessments in various industries.

Recognising the importance of this issue for the NORM industries, a collaborative research effort is being conducted through the European ALARA Network community to "…improve the quality and accuracy of internal dose monitoring techniques" (Lefaure *et al*., 2000). As a result, a European project (SMOPIE) started in November 2001.

Among the strategies adopted throughout the industries in different countries to adequately assess the true exposures of individual workers to occupational hazards, the personal measurement (with an aerosol sampler mounted on the worker's body) is frequently recommended. A wide variety of personal samplers capable of extracting the inhalable aerosol fraction have been developed over the last three decades. Resulting from the differences in their inlet design and operational parameters (e.g., the sampling flow rate), the samplers exhibit significantly different performance characteristics. Experimental and theoretical evaluations of the sampling efficiency of personal aerosol samplers revealed that – at least, for the majority of available samplers – the sampling efficiency is a strong function of the particle size and the ambient air velocity.

Within the SMOPIE project, a generic method to facilitate the identification of the particle size aerosol sampler to select for minimising the respective biases between the true and estimated exposure and the true and estimated effective dose associated with exposure by inhalation to any radioactive compound. It is thought that this method should benefit any industry from the nuclear or non-nuclear sector that have or may have potential occupational exposures to radioactive aerosols.

This work was partially supported by the European Commission DG Research within the framework of the 5th PCRD (SMOPIE project: "Strategies and Methods for Optimisation of Internal Exposures of workers from industrial natural sources").

6. References

ACGIH (1996). Threshold Limit Values for Chemical Substances and Physical Agents and Biological Exposure Indices. American Conference of Governmental Industrial Hygienists, ACGIH, Cincinnati, Ohio.

ACGIH (2001) Air Sampling Instruments for evaluation of atmospheric contaminants. 9th Edition, ACGIH, Cincinnati, Ohio, 740p.

Aitken, R.J., Baldwin, P.E.J., Beaumont, G.C., Kenny, L.C., Maynard, A.D. (1999) Aerosol inhalability in low air movement environments. J. Aerosol Sci., 30, 613-626.

Baron, P.A. and Willeke, K..(2001): Aerosol Mesurement. Principles, techniques and Applications. Second edition. Edited by Baron and Willeke, Wiley-Interscience, New –York, 1131p.

Britcher, A.R. and Strong, R. (1994) Personal air sampling – a technique for the assessment of chronic low level exposure*? Rad. Prot. Dosim., 53, 59-62.*

CEN (1993). Workplace atmospheres: Size fraction definitions for measurements of airborne particles in the workplace. *CEN standard EN 481. CEN, Bruxelles, Belgium.*

Council Directive 96/29/EURATOM (1996) Council of the European Union laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionising radiation. *Council Directive 96/29/EURATOM of 13 May 1996.*

Degrange, J.-P, Gibert, B., Basire, D. (1999) A radiological protection study in a french uranium refinement plant. *3rd ALARA Network Workshop on Managing Internal Exposures, Neuherberg, November 1999, 10p.*

Hinds, W.C. (1999) Sampling for Inhalable Aerosols. Particle size-selective sampling for particulate air contaminants. ACGIH, Cincinnati, Ohio.

ISO (1995). Air quality - Particle size fraction definitions for health-related sampling. *International Organization for Standardization, ISO standard 7708, ISO, Geneva, Switzerland.*

ICRP publication 68 (1994) International Commission on Radiological Protection: Dose coefficients for Intakes of Radionuclides by workers. Replacement of the ICRP publication 61. Volume 24, Nos 4. Pergamon, Elsevier Science Ltd., Oxford.

Jarvis N.S., Birchall A., James, A.C., Bailey, M.R., Dorrian, M.D. (1996). LUDEP 2.0, Personal computer program for calculating internal doses using the ICRP publication 66 respiratory tract model. NRPB-SR287 (Chilton: NRPB).

Kenny, L.C., Aitken, R., Chalmers, C.P., Fabriès, J.F., Gonzales-Fernandez, E., Kromhout, H., Lidén, G., Mark, D., Riediger, G., Prodi, V. (1997) A collaborative European study of personal inhalable aerosol sampler performance*. Annals of Occupational Hygiene, 41, 135-153*.

Lefaure, C., Croft, J., Degrange, J.-P (2000) Observation and recommendations of the 3rd European ALARA Network Workshop on Managing internal Exposures. *European ALARA Newsletter, issue 8, May 2000, 2-6.*

Maynard, A.D. and Jensen, P.A. (2001) Aerosol measurement in the workplace. IN: Aerosol Measurement: Principles, Techniques, and Applications, Second Edition, Edited by Paul A. Baron and Klaus Willeke. Wiley InterScience, Inc., New York, USA. pp. 779-799.

NCRP (1997) report n°125 Deposition, retention and dosimetry of inhaled radioactive substances. National Council on Radiation Protection and Measurements, Bethesda, MD.

Van der Steen, J., van Weers, A.W., Lefaure, C., Degrange, J.-P;, Vaillant, L., Shaw, P.V., Witschger, O. (2002) Strategies and Methods for Optimisation of Internal Exposures of workers from industrial natural sources (SMOPIE), IAEA Conference on Occupational Radiation Protection, 26-30 august 2002, Geneva, Switzerland

Vincent, J.H. (1995). Aerosol Science for Industrial Hygienists. *Pergamon, Elsevier Science Ltd., Oxford.*

Witschger, O. (2000) Sampling of airborne dusts in workplaces atmospheres. Kerntechnik, 65, 28-33.

Witschger, O., Fauvel, S., Basso, G., Grinshpun, S.A. (2004) Performance of Personal Inhalable Aerosol Samplers in Very Slowly Moving Air When Facing the Aerosol Source. Ann. Hyg., Jun 2004.