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Personal Thoracic CIP10-T Sampler and its Static Version CATHIA-T

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A specific version of the personal aerosol sampler CIP 10 was designed, named CIP10-T, for sampling the conventional CEN thoracic fraction. A static sampler, named CATHIA, was also designed. It uses the same sampling head, but the size selected particles are collected onto a filter. The combined particle efficiency of the aspiration slot and the selector was measured in a horizontal wind tunnel at low air velocity, close to 16 cm s^{-1}. The flow rate of both samplers was fixed at its nominal value, i.e., 7 l min^{-1}. Two different methods were used: the former was based on the Aerodynamic Particle Sizer (TSI); the latter used the measurement of particle size distribution of the collected samples by the Coulter technique. For the CIP10-T sampler, the particle collection efficiency onto the rotating cup was also measured. For both samplers bias and accuracy maps have been calculated, following the recommendations of a new CEN standard about sampler performance. The bias does not exceed 10% in absolute value for both samplers, within a large range of particle size distribution of the total aerosol. For the CIP10-T sampler, the accuracy map exhibits a large area where the accuracy is better than 10%, corresponding for example to 4 μm $\leqslant$ MMAD $\leqslant$ 14 μm for GSD = 2. For the same geometric standard deviation, the accuracy is still better than 20% for 15 μm $\leqslant$ MMAD $\leqslant$ 21 μm. For the CATHIA-T sampler, the accuracy map can be roughly divided into two parts. The accuracy remains better than 10% for MMAD $\leqslant$ 12 μm, and it remains between 10 and 20% for coarser aerosols, with 13 μm $\leqslant$ MMAD $\leqslant$ 20 μm, provided GSD $\geqslant$ 2.

INTRODUCTION

The assessment of workers' exposure to airborne particles is a major task in occupational hygiene, to evaluate the hazards due to dispersed harmful and toxic substances. The development of personal samplers during recent decades has improved the representativeness of exposure measurements, as aerosol sampling is performed in the immediate vicinity of the upper respiratory airways of the workers, and the sampling time usually covers a large part of the whole time shift. Many personal samplers have been designed for sampling the conventional inhalable and respirable fractions of an aerosol: 25- and 37-mm cassettes (Vincent, 1989; Jensen and O'Brien, 1993; Vincent, 1995); 7-hole sampler (Health and Safety Executive, 1986); IOM-sampler (Mark and Vincent, 1986); cyclones: Higgins and Dewell cyclone (Higgins and Dewell, 1967) and its further modifications (Lidén, 1993), 10 mm nylon Dorr-Oliver cyclone (Caplan *et al.*, 1977). Another type of sampler was developed in France, based on the collection of airborne particles in a rotating cup containing a porous polyurethane foam. The rotation of the foam also ensures air movement inside the sampler, avoiding the use of an external pump, thus yielding a very compact equipment. The CPM3 respirable sampler was the first sampler of that type, using a cyclone for particle size selection (Fabriès *et al.*, 1987). Its flow rate is 5 l min^{-1}. A more compact respirable sampler, named CIP 10, was then designed (Courbon *et al.*, 1988). Its flow rate is 10 l min^{-1}, and particle size selection is obtained by combining impaction of the aspirated particles onto a small large pore foam and filtration through a static cylindrical foam.

As conventions (Comité Européen de Normalisation, 1993) define three aerosol fractions of interest in relation to health effects of inhaled particles, namely the inhalable, thoracic, and respirable fractions, the hygienists are confronted by the relative lack of availability of thoracic samplers. The thoracic fraction is

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relevant in the case of substances having a local effect on the conductive respiratory airways, like irritation, or in the case of toxic substances which can slowly diffuse into the blood through the tracheo-bronchial walls.

A few samplers only can meet the new specifications of the thoracic convention. One of the oldest samplers which was designed to ensure a particle-size selection close to the actual thoracic convention was the vertical elutriator (Lynch, 1970). This sampler is still widely used, particularly in the United States, for measuring aerosol concentration in the cotton industry. A prototype personal sampler, capable of collecting the three conventional fractions simultaneously, was derived from the original IOM sampler (Aitken *et al.*, 1993). It is based on selective penetration of particles through porous foams.

As the CIP 10 sampler is now widely used in its respirable version (now named CIP10-R), it appeared very attractive to design a thoracic version by modifying the upper part of the sampler. A first tentative prototype sampler was tested (Fabriès *et al.*, 1989), and more recently improved. The new version is named CIP10-T. This paper presents this latter version, and its sampling features. Furthermore, a static version of the CIP10-T sampler has also been made, combining the use of the sampling head of the CIP 10 sampler with a collection stage based on a filter. This static version is also presented.

TECHNICAL FEATURES OF THE PARTICLE-SIZE SELECTOR

The particle-size selector is presented in Fig. 1, mounted inside the sampling head of the CIP10-T instrument. It is a conical stainless steel piece, with eight cylindrical orifices of 1.6 mm in diameter, through which the aerosol is forced to flow. Coarser particles are deposited mainly by inertial effects onto the conical surface of the selector, in the vicinity of the orifices, and onto the external surface of the lower base. Finer particles are not retained by the selector, and thus they can reach the lower stage of the sampler, which contains the rotating cup with its cylindrical foam responsible for the air movement, and they are finally trapped by the foam. The flow rate was fixed at the value of 7.0 l min^{-1}, close to the flow rate obtained naturally by simply replacing the respirable selector by the new thoracic selector. However, a fine adjustment by means of the potentiometer acting on the voltage input of the DC motor is necessary to get the right value. The flow rate and the orifice diameter have been both optimised during several consecutive experimental trials, yielding for each configuration a complete set of sampling efficiency values. The experimental methods used are described in the experimental section.

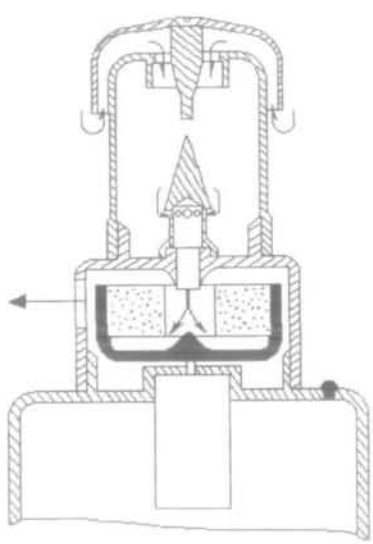

Fig. 1. General scheme of the sampling head of the CIP 10-T sampler with its thoracic particle-size selector.

PERSONAL CIP10-T AND STATIC CATHIA-T THORACIC SAMPLERS

The CIP10-T sampler, equipped with the new selector, is a personal thoracic sampler, having the same practical advantages as that of the respirable version. It operates independently for a long period, generally more than 30 hours with the new DC motor (Escap™ ref. M2R 112101, Portescap, La Chaux de Fonds, Switzerland). When the sampler is intended to be used in the textile industry, it should be preferably equipped with a protection grid (Görner and Fabriès, 1994), to avoid the possible clogging of the selector holes by long fibres. This grid has a total area of 21 cm^2, and is made of 0.2 mm stainless steel wire with a 0.8 mm square mesh. The main difficulties encountered with the sampler are the sensitivity of the cup with its collection foam to air humidity, much higher than that of many filter types, which can alter the precision of direct weighing of the cups, and the extraction of collected substances from the foam.

According to our experience, when new foams have been correctly washed and conditioned in the laboratory before weighing, the standard 95% confidence interval around a collected mass is ± 0.4 mg, provided three blank cups have been weighed in parallel with the active cup and foam. For comparison, the same interval would be approximately ± 0.022 mg for Nuclepore filters of 37 mm in diameter. Therefore, it is generally required to sample a sufficiently significant mass of particles, at least 1 mg, to prevent an inaccurate concentration result. On this basis, and for a sampling duration of 8 hours, the limit of detection would be approximately 0.12 mg m^{-3}, and the limit of quantification 0.3 mg m^{-3}. These limits would be lower for a longer sampling time.

The extraction of inorganic substances (for example quartz dust) is possible by foam incineration. However this method cannot be applied to organic substances. In this case the direct extraction of the col-

lected substances is possible by using an appropriate solvent, which does not damage the polyurethane foam itself. The direct resuspension of collected particles into a liquid phase is difficult, and this operation is sometimes of very poor efficiency.

The personal CIP 10 sampler should therefore preferably be used when the requirements concerning foam weighing or the extraction of the collected substances can be met. Because of these limitations, it appeared useful to design a new complementary instrument, which would take advantage of the design and performance of the respirable and thoracic selectors This sampler, named CATHIA (acronym meaning in French Thoracic, Inhalable, and Respirable Aerosol Sampler), combines the sampling head of the CIP 10 sampler and a more classical filter system for collecting particles. It has to be connected to an external pump, and is therefore a static sampler. Its schematic diagram is given in Fig. 2 (thoracic version), and a general view in Fig. 3. Its total height is 22.3 cm, the external diameter of the cylindrical part between the filter and the sampling head is approximately 38 mm, and the internal diameter 32.0 mm. The internal surface is polished for the stainless steel version, whilst a thin layer of nickel is deposited on all surfaces for the brass version. A standard 37 mm Millipore cassette can be mounted as a filter-holder. An intermediate metallic ring can be inserted, pressing the filter to avoid any electrostatic effects. Another metallic filter-holder was designed. It can be purchased on special request from the manufacturer (Arelco A.R.C., Montreuil, France). The tubing length as well as the angle of the upper conical part located below the selector stage was optimised in order to obtain a uniform particle deposit on the filter surface. Recently, the use of the CATHIA-T sampler for the assessment of the airborne concentration of asbestos fibres was tested (Kauffer *et al.*, 1996) and compared with other samplers. In this study, it was also shown that collected particles obtained by sampling an aerosol of dolomite dust yielded a uniform surface density.

The experimental results of sampling efficiency for both samplers and the experimental methods are detailed in the next paragraphs.

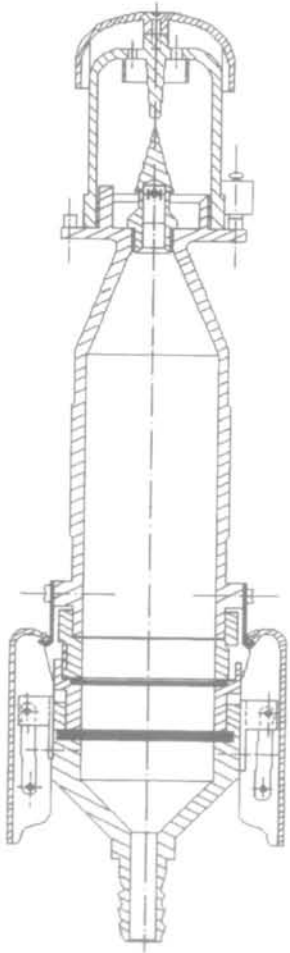

Fig 2 Cross sectional view of the static CATHIA-T sampler. thoracic version. Nominal flow rate 7.0 l min^{-1}, to be ensured by an external flow controlled pump

EXPERIMENTAL ASSESSMENT OF SAMPLING EFFICIENCY

The combined particle penetration efficiency through the aspiration slot and the selector was measured in a horizontal wind tunnel (Fabriès *et al.*, 1984), using two methods differing from each other by the analysis technique. The sampler under study was placed in static configuration in the test section of 1 × 1 m^2 cross-sectioned area. Two test aerosols of aluminium oxide (Aloxite 175, Société Abrasifs Mercier, Ivry, France) and dolomite (Dolomie DRB-4/15, Société des Blancs Minéraux de Paris, Chatou, France.) were generated in the tunnel by using a fluidised-bed generator. A ^{85}Kr radioactive source was used as an electrostatic charge neutraliser downstream of the generator. Each aerosol flowed horizontally with a uniform flow velocity of approximately 16 cm s^{-1}. Owing to the low air velocity, the test conditions can be regarded as corresponding to calm air. Particle-size distributions of both dusts were unimodal, and their main characteristics are tabulated in Table 1

Within a small particle size interval, particle penetration efficiency through the sampler, excluding the collection stage, is the ratio of the concentration of particles having passed through the aspiration slot and the selector, to the undisturbed upstream particle concentration of the test aerosol. If the collection stage is able to capture all coming particles, then penetration efficiency is identical to sampling efficiency. If particles are partly rejected by the collection stage, as it is the case with the CIP 10 sampler, then sampling efficiency is the product of penetration and collection efficiencies.

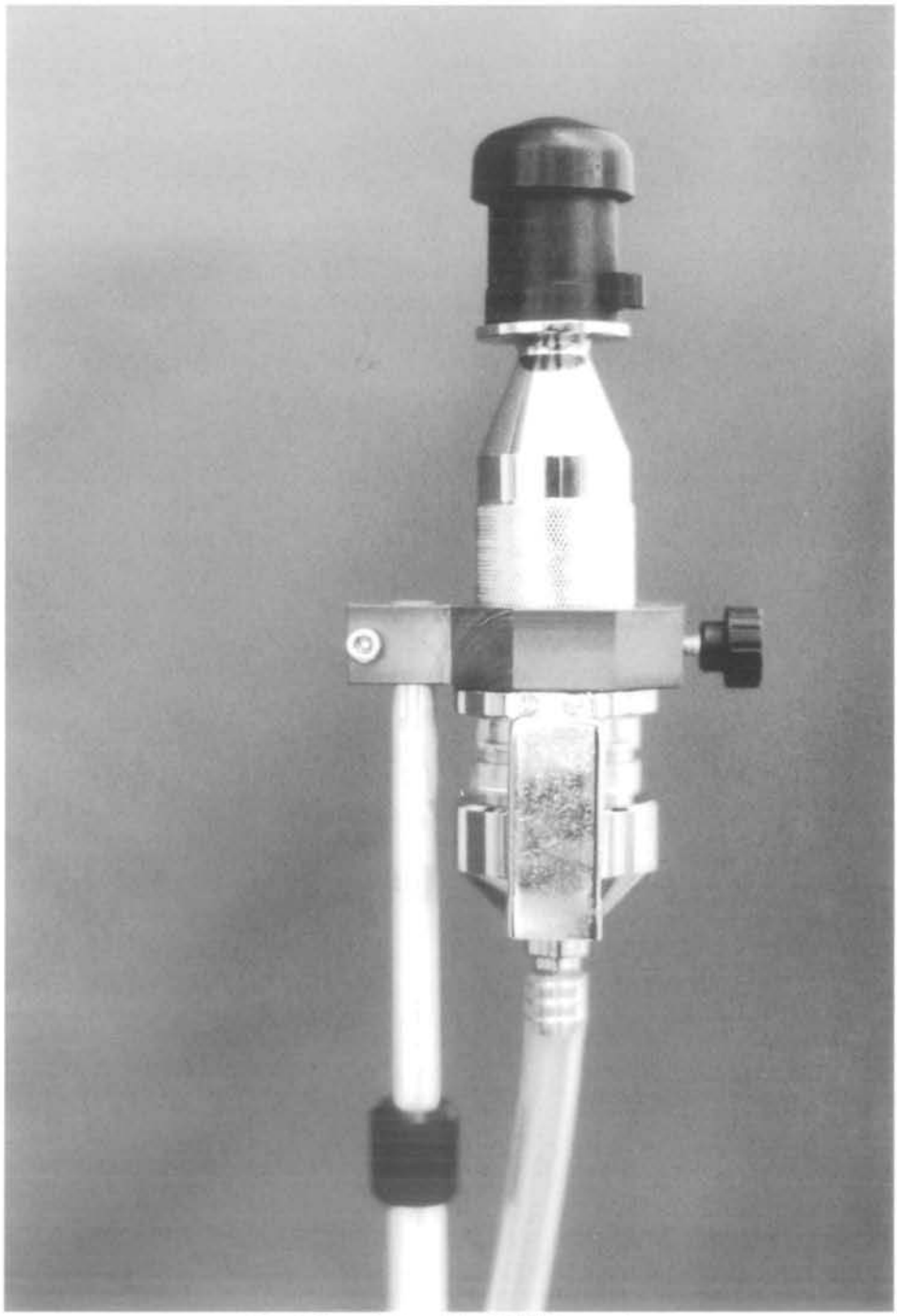

Fig. 3. General view of the CATHIA sampler.

Aerodynamic particle sizer method

The sampling head of the CIP10-T was placed vertically in the working section, and connected to an Aerodynamic Particle Sizer APS 3300 (TSI, Inc., St Paul, MN, USA) instrument. The APS instrument was connected to the outlet of the sampler selector stage by means of an intermediate flow adapter. The latter was designed to adapt the flow rate of the sampler to that of the APS (total flow rate 5 l min^{-1}, 4 l min^{-1} being used as sheath air after filtration and 1 l min^{-1} as the flow rate to be analysed). A part of the sampled aerosol corresponding to 2 l min^{-1} was withdrawn to compensate the flow rate difference. The aerosol concentration in the wind tunnel was limited to a few mg m^{-3} so that the coincidence errors related to high particle counts were negligible. For dolomite, the particle number concentration was approximately 100 part.cm^{-3}.

The properties of the test aerosol were measured in another successive sequence by simply disconnecting

Table 1. Experimental and calculated values of the ratio C/C_0 for the CATHIA sampler and for various experiments performed with both test aerosols. C: mass concentration of the sampled aerosol, C_0: mass concentration of the total aerosol; C^*: mass concentration of the conventional CEN thoracic fraction Δ: bias = $(C-C^*)/C^*$

Aerosol	MMAD μm	σ_g		C/C_0 experimental CATHIA	C/C_0 calculated CATHIA	C^*/C_0 calculated CEN convention	C/C^* calculated	Δ %
				0.4544	0.4511	0.3880	1 16	16.3
				0.4751	0 4931	0.4350	1.13	13.4
				0.4612	0.4430	0.3800	1 17	16.6
				0.4138	0 4257	0.3619	1.18	17.6
dolomite	11.4	2.16						
			mean	0.4511	0 4532	0.3912	1.16	16.0
			st dev	0.0263	0 0286	0.0312	0 02	1.8
				0.1645	0 1490	0.0874	1 70	70.5
				0.1826	0.1540	0.0918	1 68	67.8
				0.1994	0 1469	0.0866	1 70	69.6
				0.2644	0.1641	0.1019	1.61	61.0
Aloxite 175	17 9	1.35						
			mean	0.2027	0 1535	0.0919	1.67	67.2
			st. dev.	0.0435	0.0077	0.0070	0 04	4.3

the sampler from the vertical tube to the APS, and by inserting a sampling orifice of 16 mm in diameter, oriented upwards. The flow rate was maintained at the same value of 7.0 l min^{-1}. These conditions ensure a sampling efficiency close to unity up to a particle aerodynamic diameter of 15 μm. Using the model developed by Grinshpun *et al.* (1993), it can be shown that the aspiration efficiency is still 0.988 at 30 μm. Furthermore, as the same experimental set-up between the sampling point and the detection zone of the APS instrument was used for both the sampler and the reference sampling orifice, the eventual particle losses inside the vertical line and the APS instrument itself were the same, introducing no additional bias into efficiency measurement.

As it was expected that the particle collection stage of the CIP10-T, corresponding to the rotating cup compartment, was not completely efficient for the finest particles, its collection efficiency was measured. The air outlet was directly connected to the APS analyser by using the same experimental set-up. The collection efficiency was calculated for each particle-size interval by dividing the number concentration of collected particles (i.e the difference between the concentration of particles leaving the selector stage and the concentration of particles leaving the CIP10-T outlet) by the concentration of particles leaving the selector compartment.

For each configuration of the experimental set-up (reference sampling, behind the selector stage or the air outlet of the CIP10-T sampler), five consecutive measurement cycles were achieved, and the efficiency was calculated from average particle number concentrations measured for several adjacent particle-size intervals. The stability of the test aerosol concentration was checked during the experiments. The relative standard deviation of particle number concentration was about 4% near 1 μm in particle aerodynamic diameter, 4% near 5 μm and 14% near 10 μm. The variance of efficiency was also calculated by analysis of error propagation (International Organization for Standardization, 1993) applied to all experimental variables including particle counts and flow rates.

Particle density and shape corrections were performed by means of an additional software aiming at recalculating the calibration curve of the APS instrument, associating the particle aerodynamic diameter with the channel number of the instrument accumulator. These corrections are generally required as the particles exiting the APS nozzle do not fulfil the Stokes regime. The procedure used enables to create a table which is directly used by the original BASIC software used by the instrument Particle density corrections were performed following Wang and John, 1989. Particle shape corrections were performed from the calculation of the drag coefficient according to Haider and Levenspiel, 1989. These authors proposed to introduce a sphericity factor for isometric and disk-shaped particles. The correspondence between sphericity factor and dynamic shape factor is given by the equation from Pettyjohn and Christiansen, 1948. For either dolomite or Aloxite dust, the dynamic shape factor K_{rv} was determined by comparing particle size distributions measured both with a Coulter counter and a Marple 8-stage personal impactor (Grazeby Andersen, Model 298, Smyrna, GA), after particle volume diameter D_v-aerodynamic diameter D_{ae} conversion by using the equation given by Hinds, 1982. K_{rv} was calculated vs. particle diameter. Its numerical values lie within 1 0 and 1.25 for Aloxite over 10 μm $< D_{ae} <$ 30 μm, and within 1.3 and 1.0 for dolomite over 7 μm $< D_{ae} <$ 26 μm. However, the relative standard deviation was estimated to be close to 10%. Therefore, taking into account the accuracy corresponding to the instrument calibration by means

of monodisperse latex microspheres (Duke Scientific Co., Palo Alto, CA), which corresponds to a standard deviation $\sigma_{cal}(D_{ae}) = 0.1\,\mu m$, the overall uncertainty in D_{ae} (95% confidence interval) due to calibration and errors in K_n is estimated to be $\pm 1.0\,\mu m$ in the vicinity of $10\,\mu m$.

Other authors dealt with the problem of particle density and shape corrections: Brockmann and Rader, 1990; Cheng *et al.*, 1990; Cheng *et al.*, 1993.

Coulter method

The sampling head of one specimen of CIP10-T sampler was mounted as previously described, but a pre-weighed filter was placed inside a filter-holder just downstream of the aerosol exit. It was of polycarbonate Nuclepore type, 25 mm in diameter, pore size $0.8\,\mu m$. An external pump ensured the right flow rate; the latter was controlled by a regulated mass flow meter (Bronkhorst, Ruurlo, The Netherlands). The test aerosol was measured in parallel with the sampler by means of a 25 mm open filter-holder facing the aerosol movement inside the wind tunnel, containing a $0.8\,\mu m$ Nuclepore filter, with a flow rate of $16\,l\,min^{-1}$. As previously it was verified that these conditions fulfilled the requirements for no bias sampling. Both filters were pre-weighed with 3 blank filters. For the reference sampling line, a similar mass flow meter was used to maintain the flow rate at a constant value, whatever the pressure drop across the filter. The flow meters were calibrated before their use

With a concentration of the test aerosol close to $11\,mg\,m^{-3}$ for dolomite dust and $6.7\,mg\,m^{-3}$ for Aloxite dust respectively, the duration of each sampling phase was fixed approximately at two hours. Thus the mass of particles collected on the filter located downstream of the sampling head was at least 1 mg. The mass of particles collected on the reference filter was much higher, about 21 mg for dolomite. It was measured after charge neutralisation on a microbalance of sensitivity $1\,\mu g$. The average mass variation of blank filters was taken into account. The 95% confidence interval around each experimental value of collected mass was estimated.

After weighing, the filters were carefully washed with an electrolyte solution (Isoton II®, Coultronics France S.A.), where some droplets of tensio-active agent were added. The resulting suspensions, the volume of which was recorded, were analysed by the Coulter technique (Silverman *et al.*, 1971) by means of a Multisizer® instrument (Coulter Electronics Ltd , Luton, UK). An aperture diameter of $50\,\mu m$ was used for dolomite, with a working range expressed relative to particle volume diameter of approximately [1–$30\,\mu m$]. For Aloxite, an aperture diameter of $100\,\mu m$ was used, with a working range of [1.9–$60\,\mu m$]. The suspensions were diluted down to the right particle concentration with known volumes of pure electrolyte, so as to minimise the coincidence errors, keeping however the highest possible concentration to limit the errors in counting within each channel due to Poisson law. Equivalent volume particle diameter D_v was measured, and particles were counted in 32 channels. Aerodynamic diameter D_{ae} was calculated from D_v by using correction factors for particle density and shape. The shape factor was estimated by comparing directly the particle size distributions of the test aerosol measured by both the reference filter associated with the Coulter technique and a Marple 8-stage personal, as previously described. As for the APS instrument, the 95% confidence interval around Dae = $10\,\mu m$ is estimated to be close to $\pm 1.0\,\mu m$. It would be closer to $0.2\,\mu m$ with spherical particles.

In order to check the validity of the method, it was also applied in our study to measure the sampling efficiency of the vertical elutriator (Lynch, 1970), which was originally designed for sampling airborne dust in cotton industry. The results obtained yielded a 50% cut-off aerodynamic diameter of $10.7\,\mu m$ (Görner *et al.*, 1994), whilst for example Rubow and Marple (1983) found a value of $10.5\,\mu m$.

RESULTS AND DISCUSSION

Figure 4 shows the experimental values of the penetration efficiency through the selector of the CIP10-T sampler, and the collection efficiency of its rotating cup, measured by both the Coulter and APS methods. The data are plotted with their 95% confidence interval. The error bars relative to particle aerodynamic diameter are not plotted to avoid figure overloading. The values obtained with dolomite dust by both methods seem to be consistent each other, at least on the graphical representation. The upper limit of the APS instrument is practically $18\,\mu m$ in diameter. Even for the Coulter results, the accuracy is poor above the same limit, due to the lack of particles of this size in the aerosol. For dolomite, the cumulated fraction of particle number concentration is about 0.5% for $D_{ae} > 10\,\mu m$, and 0.15% only for $D_{ae} > 15\,\mu m$ The aerosol of Aloxite 175 was chosen in order to get other more precise data for larger particles; these points are very important because they bring more information on the sampler behaviour in the tail area of the efficiency curve. As the particle-size distribution of Aloxite dust is very narrow, the precision on penetration efficiency is poor below $10\,\mu m$, and the corresponding data points are not close to that obtained with dolomite dust.

The three series of penetration efficiency data (Aloxite 175 and dolomite dusts for the Coulter method, dolomite only for the APS) were gathered and fitted by a cumulative log-normal equation ($D_{ae} > D_0$, where D_0 means any given diameter value), using a weighted least-squares method (least-squares function χ^2). The details of the method can be found in the appendix. Each deviation between the experimental efficiency and the corresponding calculated efficiency was divided by the standard deviation on the experimental value, estimated by error propagation rules. This weighting technique allows one to attribute

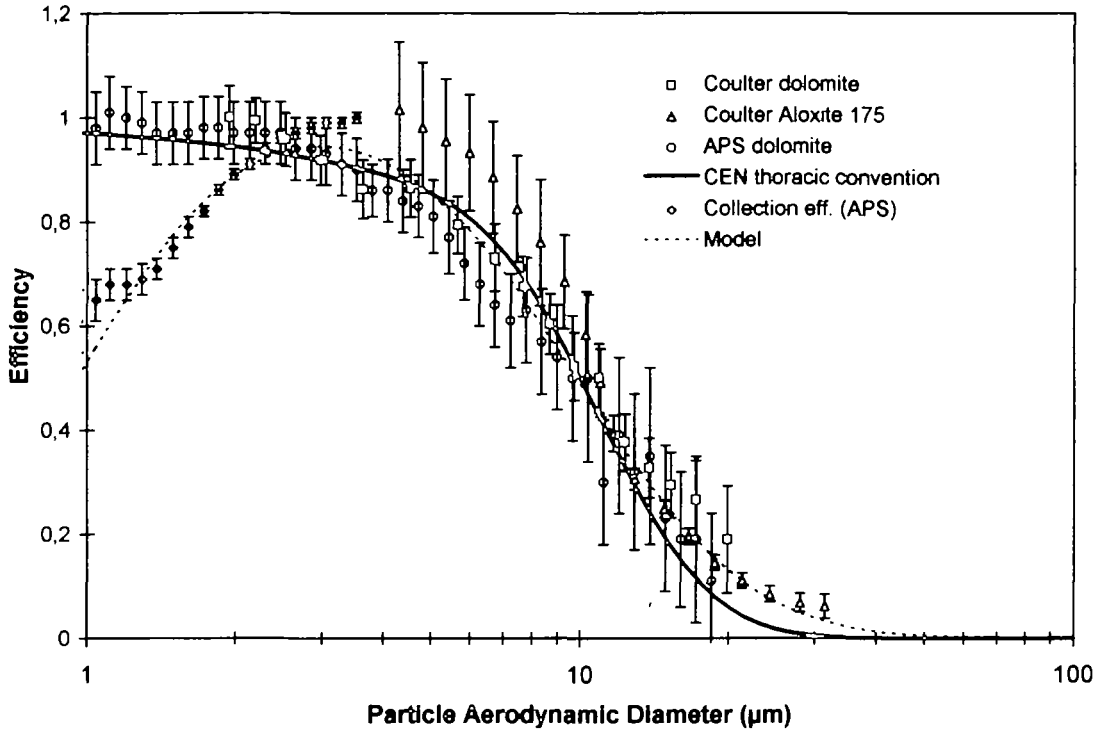

Fig 4. Experimental values of the particle penetration efficiency through the selector stage of the CIP10-T sampler, and collection efficiency of its rotating cup, measured by the Coulter and APS methods. The 95% confidence intervals are also plotted. The dashed line represents the sampling efficiency of the CIP10-T sampler, corresponding to the model with 4 adjusted parameters The continuous line indicates the thoracic convention

relatively more importance to precise data, and not to consider equally all the experimental values. The optimal values of both parameters of the log-normal model are obtained with their standard deviation, as it can be shown from data fitting procedures (Bevington, 1969). However the estimate of the standard deviation of parameters does not take into account the uncertainty in particle aerodynamic diameter. For the penetration efficiency through the particle-size selector, the following two adjusted parameters $D_{50,1}$ and $\sigma_{g,1}$ with their standard deviation were obtained, with a residual χ^2 equal to 229 for 78 data points:

$$D_{50,1} \pm \sigma(D_{50,1}) = 9.82 \pm 0.08\ \mu m$$

$$\sigma_{g,1} \pm \sigma(\sigma_{g,1}) = 1.88 \pm 0.02$$

The χ^2-test yields a maximum value of about 116 ($p < 0.005$), thus indicating a slight remaining lack of fit.

The collection efficiency of the CIP 10 rotating cup was only measured with the APS technique and dolomite dust. The single series of results were fitted by another cumulative log-normal equation ($D_{ae} < D_0$), whose parameters are ($\chi^2 = 150$ for 32 data points, maximum $\chi^2 = 53.7$ at $p < 0.005$):

$$D_{50,2} \pm \sigma(D_{50,2}) = 0.95 \pm 0.02\ \mu m$$

$$\sigma_{g,2} \pm \sigma(\sigma_{g,2}) = 1.80 \pm 0.03$$

The sampling efficiency of the CIP10-T sampler is the product of the penetration and collection efficiencies. It is represented as a dashed line in Fig. 4, along with the CEN thoracic convention (continuous line). It can be seen that the sampling efficiency is close to the convention, except for particle aerodynamic diameters less than about 2 μm, due to the partial rejection of these particles by the rotating cup. The slope of the curve near the median diameter is slightly lower than the conventional one. The capture process of particles in the vicinity of the orifice walls does not appear so stringent as for example particle impaction onto a flat surface in front of an orifice. For particles larger than 10 μm, the sampling efficiency exhibits a slower decrease.

For the CATHIA sampler, the sampling efficiency and the penetration efficiency are identical. Figure 5 shows this function as a dashed line, along with the CEN thoracic convention.

In order to check the consistency of the data with measured concentrations, several sampling runs were performed with both test aerosols for the CATHIA-T sampler. At the end of each run, the experimental concentration was measured from the mass of collected particles and the sampled volume of aerosol. On the other hand, the corresponding concentration was calculated (see Appendix, Eqs. (A1) and (A2)) from the sampling efficiency curve and the particle-size distribution of the test aerosol, measured from

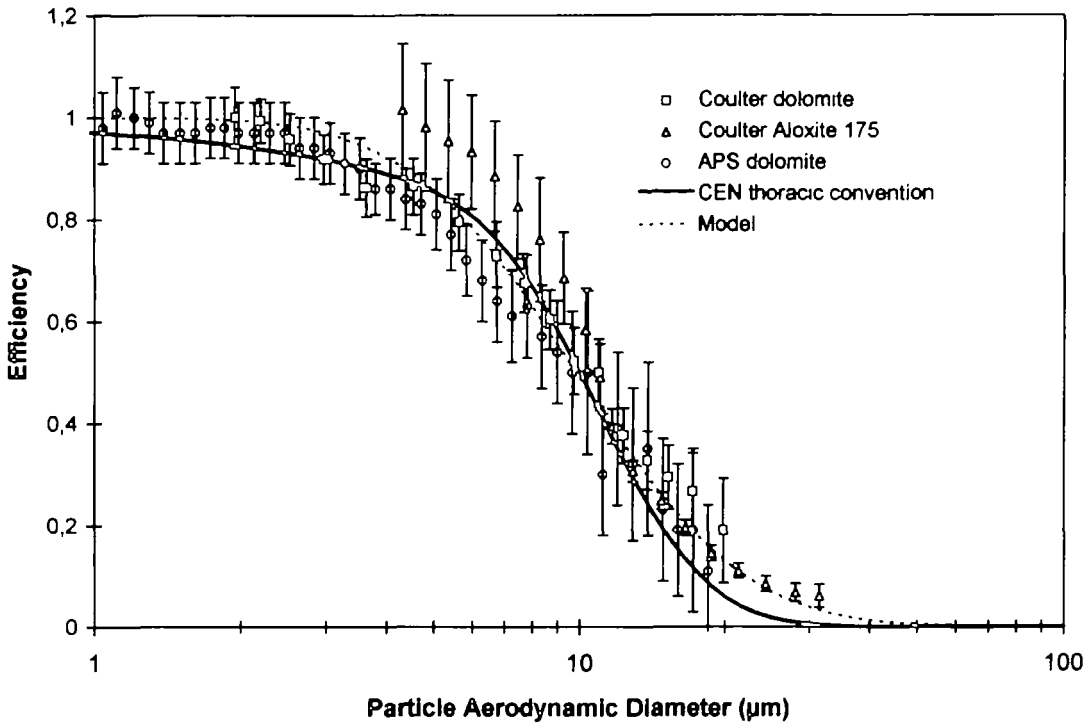

Fig 5. Experimental values of the particle penetration efficiency through the selector stage of the CATHIA-T sampler (or the CIP10-T sampler), equal to those of the sampling efficiency of the CATHIA-T sampler. The 95% confidence intervals are also plotted. The dashed line represents the 2-parameter model. The continuous line indicates the thoracic convention

the reference sample collected by the cassette. The analysis was performed by the Coulter method, after suitable particle density and shape corrections. The concentrations corresponding to the conventional CEN thoracic fraction were also calculated. All the results are given in Table 1. It can be seen that the agreement between experimental and calculated values is better than 1% for dolomite dust, but is poorer for Aloxite dust, with a relative difference close to 25%, as it is very sensitive to the particle-size distribution of the aerosol when the latter is narrow (low σ_g). It is the case for Aloxite. Table 1 also shows that the CATHIA sampler slightly overestimates the conventional concentration by a factor of about 16% for dolomite dust, and 67% for Aloxite 175. The poorer result for Aloxite dust can be easily explained by the fact that its narrow particle-size distribution, centered around 18 μm in aerodynamic diameter, amplifies the role of the tail observed with the sampling efficiency curve for larger particle diameters. Polydispersity of the aerosol induces some damping effect for GSD > 2, and the influence of the curve tail is smaller.

From the model used for the reduction of sampling efficiency data, it is possible to calculate bias and accuracy maps for both instruments. The details of calculations are given in the appendix. The bias map allows one to determine the bias in mass concentration which could be observed between the sampler and an ideal sampler, that would have a sampling efficiency strictly equal to the conventional curve, for an aerosol with a given particle-size distribution (assumed to be log-normal). The accuracy map yields for the same particle-size distribution the fraction X so that the 'observed' mass concentration C, measured by the sampler, is within the interval $C^*(1 \pm X)$ for a given probability level, for example 90%. C^* is the concentration which would be measured by the 'ideal' sampler. The fraction X takes into account not only the bias and its precision due to experimental errors, but also inter-specimen variability and variations that could arise from external factors, like the flow rate. The principles of the evaluation of sampler performance are discussed in a CEN standard project (Comité Européen de Normalisation, 1995). The last version (Comité Européen de Normalisation, 1997) has simplified the procedure, and the accuracy is simply taken as the upper limit of the confidence interval of the bias in absolute value. For an aerosol sampler used in occupational hygiene for exposure assessment, it is required that sampler accuracy X should be better (lower) than 30% (Comité Européen de Normalisation, 1997).

The bias and accuracy maps for either sampler CIP10-T or CATHIA-T in thoracic configuration are shown in Figs 6–9. The bias does not exceed 10% in absolute value for both samplers within a large area of the map. The CIP10-T shows a slight underestimation of the thoracic concentration C^* for MMAD < 8 μm. The bias reaches values less than

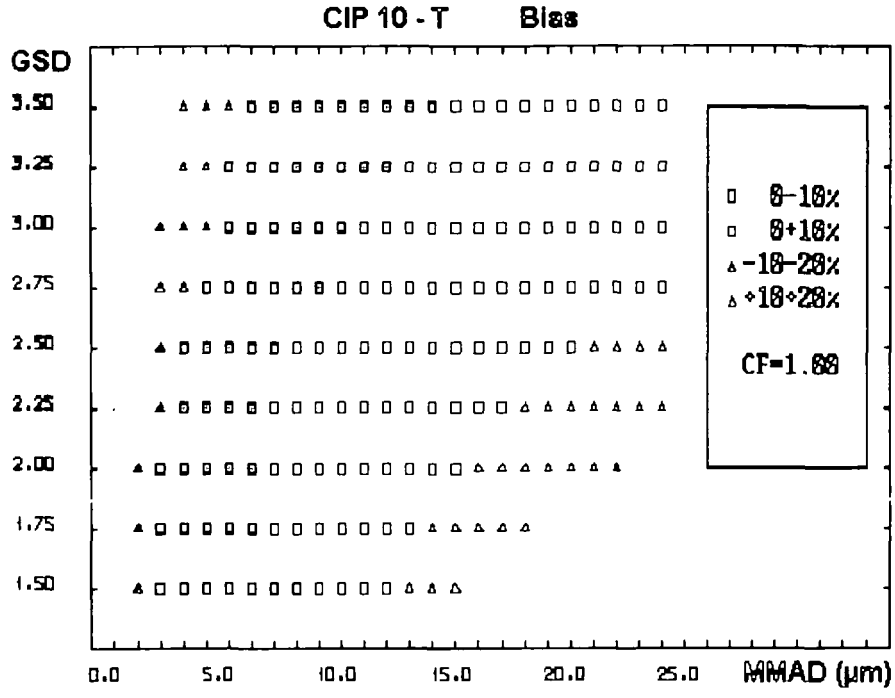

Fig. 6. Bias map of the CIP10-T sampler at 7.01 min^{-1}. The bias values are expressed as percentages. Negative values indicate an underestimation of the conventional concentration; positive values indicate an overestimation. No correction factors (CF = 1) are used.

-10% only for MMAD $\leqslant 2\,\mu m$. This is due to the rejection of fine particles by the rotating cup of the sampler. This phenomenon does not occur with the CATHIA-T sampler, and the bias always remains positive, except for slightly polydisperse aerosols corresponding to GSD $\leqslant 1.50$. For the CIP10-T sampler, the accuracy map exhibits a large area where the accuracy is better (lower) than 10%, corresponding for

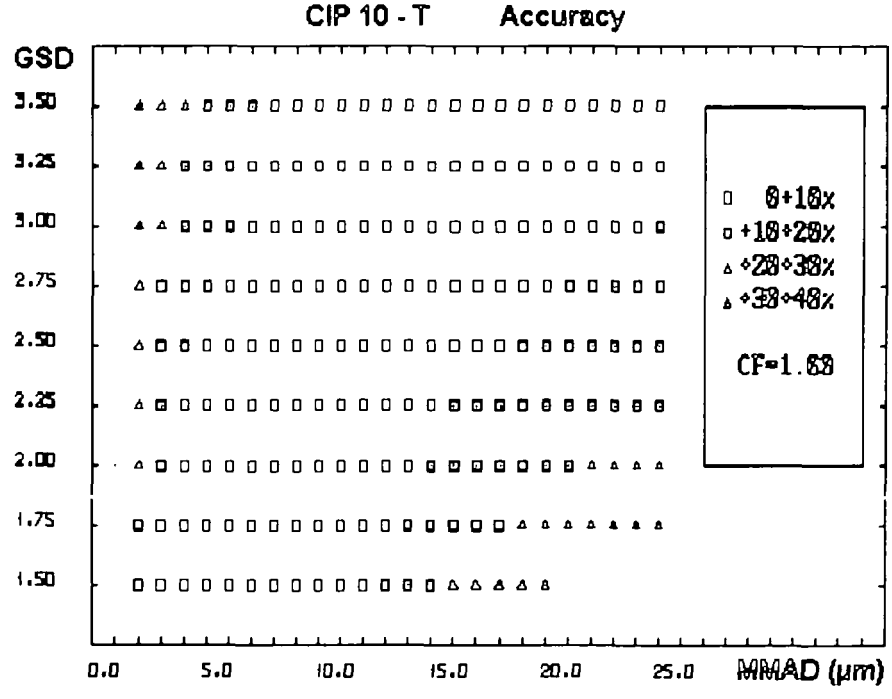

Fig. 7. Accuracy map of the CIP10-T sampler at 7.01 min^{-1}. Each point of the map indicates the accuracy range in percentages. Accuracy is calculated as indicated in annex. No correction factors (CF = 1) are used.

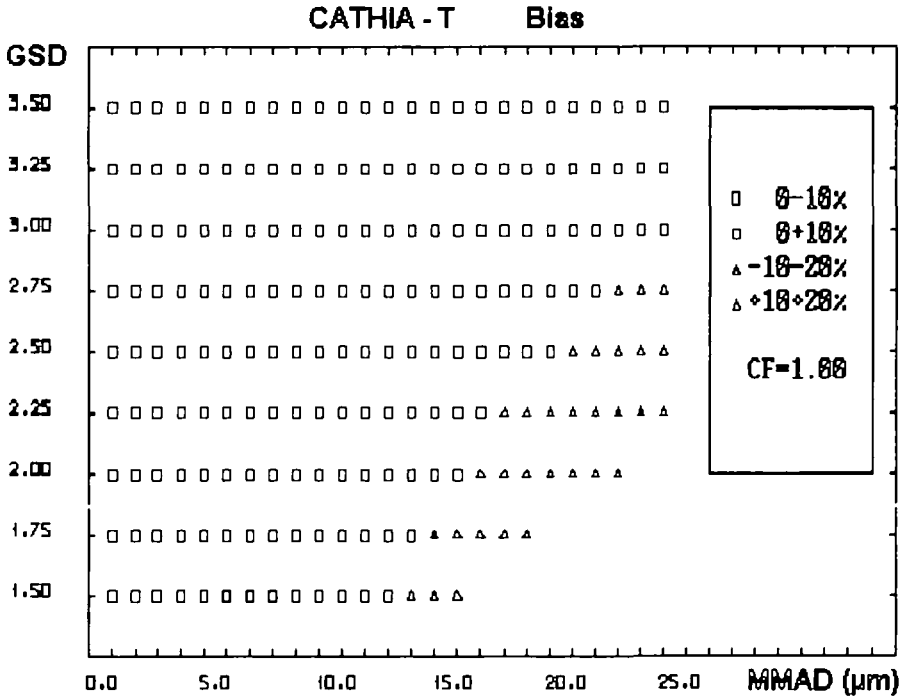

Fig. 8 Bias map of the CATHIA-T sampler at 7.01 min^{-1}. The bias values are expressed as percentages. Negative values indicate an underestimation of the conventional concentration, positive values indicate an overestimation No correction factors (CF = 1) are used

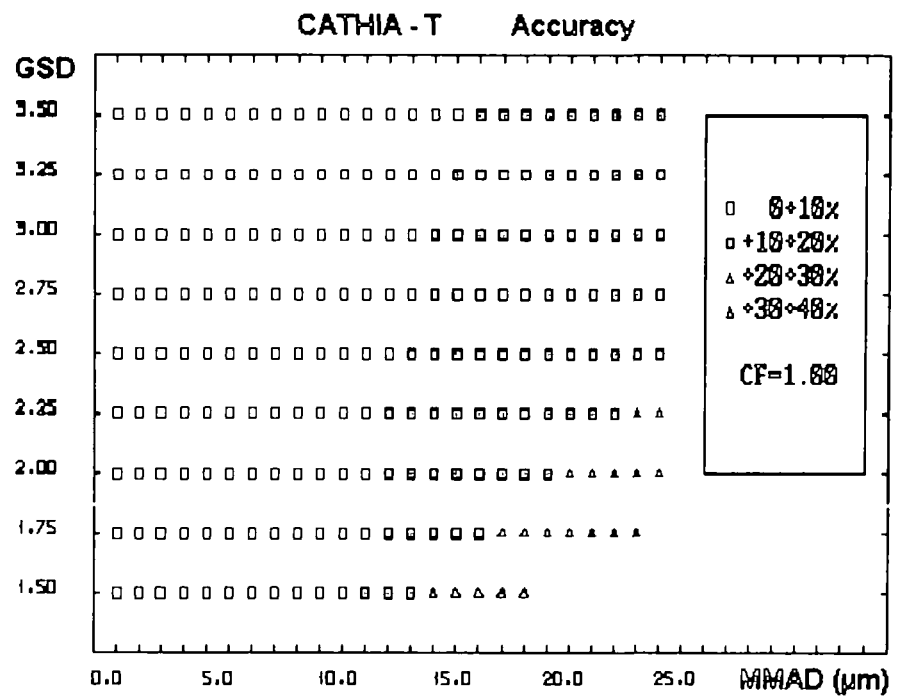

Fig 9 Accuracy map of the CATHIA-T sampler at 7.01 min^{-1}. Each point of the map indicates the accuracy range in percentages Accuracy is calculated as indicated in annex No correction factors (CF = 1) are used

example to 4 $\mu m \leqslant$ MMAD $\leqslant$ 14 μm for GSD = 2. For the same geometric standard deviation, the accuracy is still below 20% for 15 $\mu m \leqslant$ MMAD $\leqslant$ 21 μm. For the CATHIA-T sampler, the accuracy map can be roughly divided into two parts. The accuracy remains less than 10% for MMAD $\leqslant$ 12 μm, and it lies between 10 and 20% for coarser aerosols, with 13 $\mu m \leqslant$ MMAD $\leqslant$ 20 μm, provided GSD $\geqslant$ 2.

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APPENDIX

Any particle-size selective aerosol sampler, characterised by its particle sampling efficiency $E(D_{ae})$, placed in an aerosol of concentration C_0 and mass particle-size distribution $f_M(D_{ae})$, measures a concentration C given by:

$$C = C_0 \int_0^{\infty} f_M(D_{ae}) E(D_{ae}) \mathrm{d} \ln D_{ae} \qquad \text{(A1)}$$

The concentration corresponding to the target convention C^* can be calculated by simply replacing E by the conventional function $E^*(D_{ae})$:

$$C^* = C_0 \int_0^{\infty} f_M(D_{ae}) E^*(D_{ae}) \mathrm{d} \ln D_{ae} \qquad \text{(A2)}$$

In this work the efficiency functions which are considered for either sampler are the following:

$$\text{for CATHIA.} \quad E(D_{ae}) = 1 - F(D_{ae}) \tag{A3}$$

where F is the cumulative log-normal function defined by:

$$F(D_{ae}) = \frac{1}{\sigma\sqrt{2\pi}} \int_0^{D_{ae}} \exp\left[-\frac{(\ln D - \ln D_{50})^2}{2\sigma^2} \right] d\ln D$$

with two parameters: D_{50} (median aerodynamic diameter) and σ (or σ_g, geometric standard deviation defined by $\sigma = \ln\sigma_g$)

For the CIP10-T sampler the efficiency is simply the product of two cumulative log-normal functions:

$$E(D_{ae}) = [1 - F_1(D_{ae})]F_2(D_{ae}) \tag{A4}$$

where index 1 is applied to the penetration efficiency through the particle-size selector (equivalent to the sampling efficiency of the CATHIA-T sampler) and index 2 to the collection efficiency of the rotating cup.

The bias Δ between the measured concentration and the conventional concentration is defined by:

$$\Delta = (C - C^*)/C^* \tag{A5}$$

The bias map is obtained by calculating Δ for several log-normal particle-size distributions $f_M(D_{ae})$ from Eqs (A1)–(A2) and (A5). The mass median aerodynamic diameter MMAD was varied from 1 to 25 μm in steps of 1 μm, and the geometric standard deviation GSD from 1.5 to 3.5 in steps of 0.25 μm. This range of aerosol distributions covers most situations which can be encountered in occupational hygiene (Görner *et al*, 1995). This calculation implies the knowledge of the sampling efficiency function $E(D_{ae})$ of the sampler. The procedure consisting in finding a suitable model of representation of the experimental data $E_i^E(D_{ae,i})$ was adopted in this work. The model used for data fitting contains p adjustable parameters $P_j (j = 1, p)$. Their optimal values were calculated by minimising the weighted least-squares function χ^2 defined by:

$$\chi^2 = \sum_{i=1}^{n} w_i (E_i^E - E_i^C)^2 \tag{A6}$$

with

$$w_i = 1/\sigma^2(E_i^E) \tag{A7}$$

$\sigma^2(E_i^E)$ is the estimated value of the variance of E_i^E, obtained by applying the standard error propagation rules to each experimental value of sampling efficiency E_i^E, taking into account the errors in all measured quantities. E_i^C means the value calculated from the model (exponent C) and the ith experimental value of particle aerodynamic diameter $D_{ae,i}$. The error in $D_{ae,i}$ was not taken into account. To do so, it would be necessary to calculate the variance of the residual $[E_i^E - E_i^C(D_{ae,i}, \mathbf{P})]$ for each point and to replace $\sigma^2(E_i^E)$ by $\sigma^2[E_i^E - E_i^C(D_{ae,i}, \mathbf{P})]$ in Eq (A7). A Newton-Gauss algorithm was used to minimise χ^2 with respect to parameters P_j. It can be shown that the procedure allows to determine not only the optimal values of parameters, but also to estimate their variance-covariance matrix V (Bevington, 1969). The diagonal terms of V are the estimated variances of parameters $\sigma^2(P_j)$, and the extradiagonal terms are the covariances $\text{cov}(P_j, P_k)$ $(j \neq k)$. V is obtained from the following equation:

$$\mathbf{V} = (1/2)\mathbf{U}^{-1} \tag{A8}$$

where the terms U_{jk} of the curvature matrix U of the χ^2 function are the second derivatives of χ^2 with respect to parameters:

$$U_{jk} = \partial^2\chi^2/\partial P_j \partial P_k \quad j = 1, p; k = 1, p \tag{A9}$$

If all observed deviations $(E_i^E - E_i^C)/\sqrt{w_i}$ are reduced normal deviations, which can be expected at the end of the iterative procedure, then χ^2 should follow a standard chi-square distribution with ν degrees of freedom, where ν is calculated from the number of experimental data n and the number of adjusted parameters p:

$$\nu = n - p \tag{A10}$$

The sum S^2 defined by:

$$S^2 = \chi^2/\nu \tag{A11}$$

should be close to unity. The chi-square test applied to χ^2 gives an indication about the goodness of the fit. A more realistic estimation of matrix V can be obtained by the following equation, when S^2 is larger than unity:

$$\mathbf{V} = (1/2)S^2\mathbf{U}^{-1} \tag{A12}$$

Matrix V allows the estimation of the variance of the bias $\sigma^2(\Delta)$:

$$\sigma(\Delta) = \sigma(C/C_0)/(C^*/C_0) \tag{A13}$$

with

$$\sigma^2(C/C_0) = \mathbf{T}^t \mathbf{V} \mathbf{T} \tag{A14}$$

The jth element of vector T is simply the first derivative of the concentration ratio C/C_0 with respect to parameter P_j:

$$T_j = \partial(C/C_0)/\partial P_j = \int_0^{\infty} f_M(D_{ae}) \partial E^C(D_{ae}, P_1, \ldots, P_p)/\partial P_j \, d\ln D_{ae} \tag{A15}$$

$\mathbf{T}^t$ means the transposed vector of vector T.

The CEN draft document (Comité Européen de Normalisation, 1995) proposes then to calculate the upper limit of the absolute bias at some specified confidence level. The single-sided $(1-\alpha_1).100\%$ upper confidence limit estimate of the absolute bias is:

$$\Delta_{upper} = |\Delta| + t\sigma(\Delta) \tag{A16}$$

where t is the $(1-2\alpha_1).100\%$ percentile of Student's—t distribution with ν degrees of freedom (Kenny and Bartley, 1995). It corresponds to the probability α_1 that the true value of the absolute bias exceeds Δ_{upper}.

The accuracy of aerosol concentration measured by the sampler has several components arising from various origins. The variance of Δ calculated by Eq. (A13) originates from the errors in the measured sampling efficiency values. Inter-specimen variations, related to manufacture differences between several samplers of the same type, or variations due to differences in the flow rate setting, may also affect imprecision of measured concentration. This imprecision can be expressed as the total variance of C/C^*, RSD_t^2:

$$RSD_t^2 = RSD_s^2 + RSD_{flow}^2 \tag{A17}$$

where RSD_s^2 means the variance of the concentration ratio C/C^* due to inter-specimen variability, and RSD_{flow}^2 the variance corresponding to the flow rate setting. Taking into account all these contributions, it is also possible to determine the upper limit RSD_{upper} of RSD_t at some $(1-\alpha_2)$ 100% confidence level. It was considered in this work that the variance due to inter-specimen variations was sufficiently low to be neglected ($RSD_s = 0$), and that RSD_{flow} could be estimated and equalled to the fixed value 0.02 without any further information, as it was suggested in the CEN draft (1995).

Knowing both upper limits for the bias and the total

sampler imprecision at the specific risk levels α_1 and α_2 respectively, the sampler accuracy was estimated according to the Bonferroni approach, discussed in the literature (Bartley and Fischbach, 1993; Bartley *et al*, 1994). The accuracy is defined as the quantity X, so that the interval $(1 \pm X)\ C^*$ around the 'true' concentration value C^* contains some fraction (1-α) of all concentration estimates C. It can be shown that X is obtained by solving the following equation:

$$\Phi[(\Delta_{upper} + X)/RSD_{upper}] - \Phi[(\Delta_{upper} - X)/RSD_{upper}] = 1 - \alpha \quad \text{(A18)}$$

where $\alpha = \alpha_1 + \alpha_2$, and Φ is the cumulative reduced normal distribution function. In our calculations, α_1 was fixed at the value 0.05, whilst α_2 was set equal to 0 as RSDt was taken as a constant.

In the last version of the CEN draft (1997), the accuracy X is more simply taken equal to Δ_{upper}.