

# AEROSOL SAMPLING METHODS FOR WIDE AREA ENVIRONMENTAL SAMPLING (WAES)

Finnish support to IAEA

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## Abstract

Enrichment of uranium or reprocessing of nuclear fuel are expected to produce releases of non-natural radionuclides, that are carried away in air attached in airborne particles. Wide Area Environmental Sampling (WAES)-method utilises air samplers distributed in the monitored area to detect the possible releases. For reduction of expenses, the samplers located in remote areas should be able to operate long periods of time unattended. In this work, a high-volume (flow rate 150 m<sup>3</sup>/h) air sampler with an automatic filter changing system, Hunter MKII, was developed for WAES. The sampler can collect six one-week samples until it needs to be visited for unloading the used filters and loading the new ones. The device sends real-time state-of-health information to headquarters so that long-time loss of sampling can be avoided in case of malfunction. The state-of-health data also includes indication to prevent inconspicuous tampering of the unattended sampling process.

Organic filter materials are used to collect particles due to their applicability to radio-chemical analysis. Four filter materials were tested for collection efficiency and pressure drop. The material selected for current use (Petrianov FPP-15-1.5 used as two-layers one upon the other) can collect more than 90% of the 0.2 µm particles throughout the sampling period. If there is a large concentration of coarse particles in air (as is typically the case in desert conditions), the filter clogging rate can be significantly decreased by preceding it with a low-pressure-drop pre-filter that collects the coarse particles. The filter pressure drop is low enough to easily allow one-week sampling time in typical sampling conditions (a pre-filter may be needed in heavily dust laden desert air).

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# 1 Introduction

The international Wide Area Environmental Sampling (WAES) working group considers aerosol sampling as a potential method for detecting undeclared nuclear activity [1]. Enrichment of uranium and reprocessing of nuclear fuel are expected to produce releases of non-natural radionuclides, that are carried away by air flows. The radionuclides, with an exception of noble gases and the gaseous fraction of iodine, are attached in airborne particles.

Airborne particles can be collected by an air sampler that aspirates air through a filter. Particles are retained on the filter. The air flow rate must be large in nuclear applications, because the detection limit with many analytical methods, such as gamma spectrometry, is the better the larger the sample is. Usually the air flow rate when detecting radionuclides in air is of order of 100–1000 m<sup>3</sup>/h. The period between filter changing typically varies from 1 to 7 days.

WAES-method utilises a large number of samplers located in an area of hundreds of square kilometers. Fine aerosol particles (diameter smaller than one micrometer) can be carried in air thousands of kilometers from the source. However, it is important to locate samplers close to the source, since the concentration of the non-natural nuclides is decreased as the distance from the source is increased. A sufficiently dense network of samplers is important especially for the localisation of the source. For reduction of expenses, the samplers located in remote areas should be operated long periods of time unattended. Therefore, devices that carry out filter changing automatically are preferred.

An automatic air sampler, Hunter MKII, developed for WAES is described in this report. The

device is equipped with 6 filters. Each filter collects a one-week sample, thus the sampler needs to be visited once every six weeks. The device sends real-time state-of-health data to the headquarters. Long-time losses of sampling due to malfunction or interruption in sampler use can be avoided by real-time information of the sampler status, as an inspector can be sent to the site when needed. Also, the state-of-health data includes indication to prevent inconspicuous tampering of the unattended sampling process.

The Finnish Nuclear Verification organisation (FINUVE) performed an aerosol sampling campaign in Iraq during the autumn 1998. One of the findings was that the large concentration of airborne dust in the Baghdad area resulted in filter clogging in a few days. During that time, some 10000 m<sup>3</sup> of air was aspirated through a 500 cm<sup>2</sup> filter, or 20 m<sup>3</sup> of air through 1 cm<sup>2</sup> of filter. In development of the new device, a special attention has been given to the filter sampling process to enable longer sampling periods. The filter material has been changed, filter collection area has been doubled, and a pre-filtration technique has been implemented to reduce the filter clogging-rate by collecting the coarsest particles (that are usually not interesting) separately.

The device is described in section 2. The criteria and the tests for filter material selection are presented in section 3. The optional pre-filtration technique for extending the filter collection-time in desert conditions is discussed and test results shown in section 4. Conclusions from the testing are drawn in section 5. Development of filter sample analysis methods has been carried out in connection with the work described here. Results are reported in reference [2].

## 2 Description of Hunter Automatic MKII

The new air sampler, Hunter Automatic MKII, was developed based on the experience from using a standard-model Hunter and Hunter MKI (Senya Ltd.) in Iraq 1998. The design specifications for the Hunter Automatic MKII were:

- Six filter samples can be collected automatically. Automatic data storing and transmission.
- Air flow rate at least 150 m<sup>3</sup>/h (expressed in Standard Temperature and Pressure, STP), when the filter is clean.
- Sampling time 3 or 7 days per filter, total unattended operation time 18 or 42 days.
- Filter composition adequate for chemical analysis. The filter must be easily commercially available.
- Particle collection efficiency: Sampler should collect ≥ 80% of particles with a diameter 0.2 μm, and ≥ 60% of particles with a diameter 10 μm. The former requirement constricts the properties of the filter, and the latter the properties of the rain shield upstream of the filter.
- Automatic data handling: Data logger with a sufficient capacity to calculate and store data.
- Can be operated in a temperature range of -30°C...+50°C.
- Power supply 230V / 50Hz, power less than 1.5 kW, employs single phase electricity.

### 2.1 Air sampling

Air is aspirated by a gas ring vacuum pump. The air flow rate,  $Q$ , depends on the pressure drop caused by the filter,  $\Delta p$ , approximately as

$$Q = 203 - 0.008 \Delta p, \quad (1)$$

where  $Q$  is in m<sup>3</sup>/h, and  $\Delta p$  is in Pa. A flow rate larger than 150 m<sup>3</sup>/h is maintained as far as the filter pressure drop does not exceed 6600 Pa. Filter blocking results in blower overheating. The primary reason for sampler stoppage as the filter is

clogging up is an intentional stoppage for overheat protection. If the ambient temperature is very high, then the overheat protection stoppage may take place at a lower filter dust loading level than in cool climate.

Hunter Automatic MKII is shown in Figure 1. The rotating drum has positions for the six filters. During operation, the drum is automatically turned 60° once a week, exposing a new filter to the air flow. The filter change position is on the front. The filter collection area is 42 × 24 cm, or 0.1 m<sup>2</sup>. The face velocity, that is the flow rate divided by the filter collection area, or the velocity of the air just before being aspirated through the filter, is 0.4 m/s when the flow rate is 150 m<sup>3</sup>/h. The total filter dimensions, including the margins for attachment, are 46 × 27.5 cm. Filter is put in its place by a fixing frame, that is closed without a need for a screwdriver or any other tool. In case of flexible filter material, the filter is attached in advance to a cardboard frame to obtain the firmness needed for easy filter placement and handling.

The filter air collection position is at the bottom of the device, thus the air is aspirated upwards through the filter. Particles with a diameter larger than 80 μm (assuming particle density of 2.5 g/cm<sup>3</sup>) are not sampled, as they are settled in air faster than 0.4 m/s. On the other hand, 10 μm particles easily follow the flow, as their downwards settling velocity is only 0.003 m/s [3]. The rejected coarse particles include sand, occasional garbage, and rain droplets. Upwards sampling carries out the duty of coarse-dust-rejection conveniently, without a need for a leak-tight joint between a special immobile sampling inlet and a moving filter drum.

Appendix presents the tasks that an inspector needs to carry out when visiting the sampler after six weeks of sampling.

## 2.2 State-of-health data transmission

To avoid major interrupts in sampling, and to control the status of sampling network, the following state-of-health (S-O-H) signals should be transmitted:

- External power supply (on/off).
- Sampler blower (air flow rate).
- Temperature inside the sampler.
- Tamper indication. It can include data from a door switch, motion sensor and electronic seal.

The sampler is equipped with a datalogger, which stores sampling data from various sensors. The S-O-H data is also collected by the datalogger. The sampler uses the data for air flow calculation. The datalogger will also be used for storing data from optional meteorological station. The program in the datalogger automatically sends the data out via RS-232 serial port. The data is received and stored by an industrial PC, which is run under NT operating system. The data sent by the datalogger is read by the PC, encrypted and sent to the headquarters. The data transmission will be carried out via Inmarsat-C due to its global and reliable

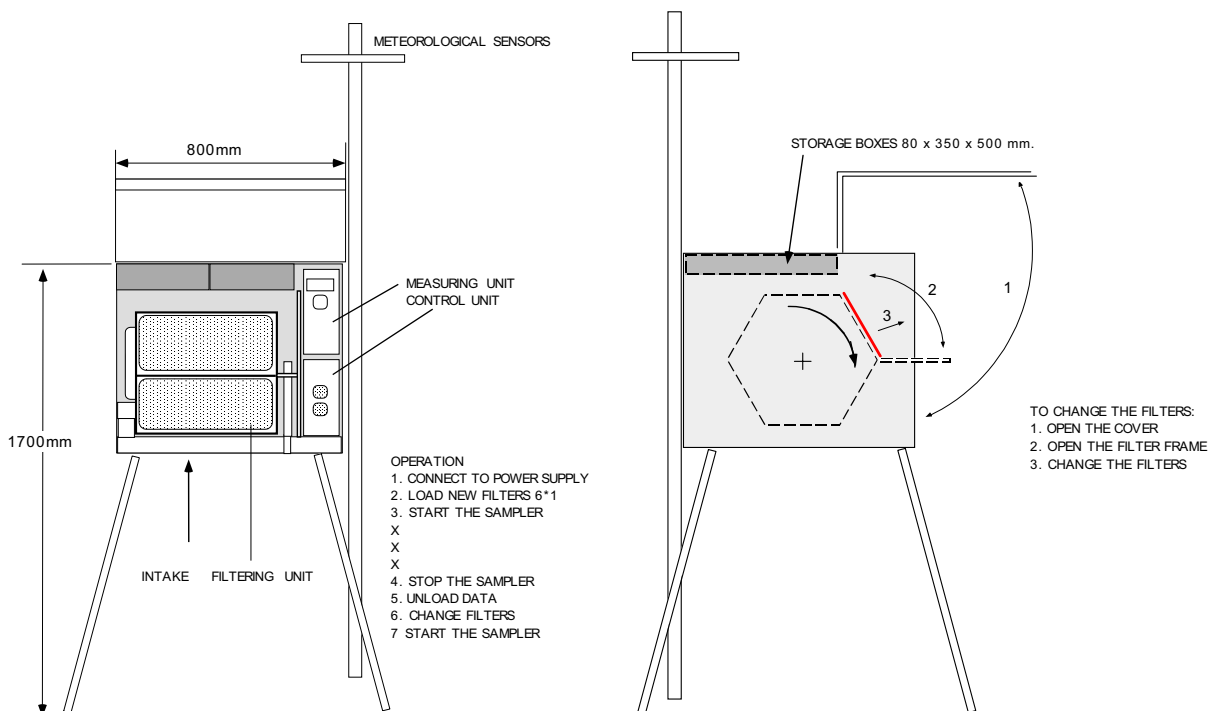
coverage. The data can be e-mailed or faxed to specific address.

Data reading and transmission is controlled by the industrial PC. It performs the following tasks:

- The S-O-H data is monitored in very short intervals.
- All received data is stored in the PC.
- In a case of alarm or failure the alarm message is sent in real time.
- The PC sends the status information to the headquarters at predefined intervals

Encryption method is One-time pad (OTP), which can not be decrypted without knowing the original encryption key [4]. OTP is useful in low bandwidth applications. Each key is used only once. The keys are produced in the headquarters and the inspector loads them into the sampler during his visit, so the risk for unauthorised access to the key is minimal. All information is encrypted before sending. Tampering the equipment is not possible without leaving any evidence behind. In case of tampering, the PC destroys the original encryption keys.

### HUNTER MK2 AUTOMATIC



**Figure 1.** Hunter Automatic MKII air sampler. Air is aspirated upwards through the filter located on the bottom position.

### 3 Choosing the filter material

#### 3.1 Criteria

Usual criteria for filter material include capability to collect particles with a sufficient efficiency, resistance to clogging, convenient to handle, and mechanical strength (does not break easily). The material should have a low concentration of U and Pu, if these elements are to be analysed. Mostly the problem here is uranium, as it is, unlike plutonium, present in nature to a significant extent. The filter material should be appropriate for preconditioning methods used in radiochemistry. Organic filter materials, such as polypropylene or polyvinylchloride, are suitable as they are ashable at low temperatures, leachable to weak acids such as HNO<sub>3</sub>, and do not contain significant amounts of uranium.

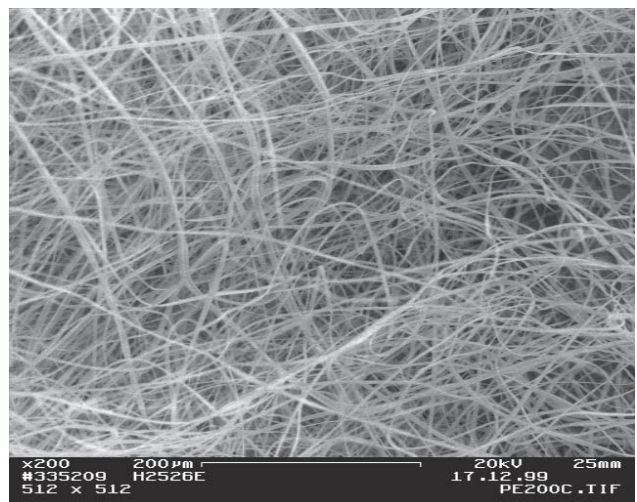
Collection efficiency is defined as a fraction of particles collected on the filter of the particles in the air entering the filter. Collection efficiency depends on the filter structure, the particle size and face velocity [3]. It is typically the lowest for particles of about 0.1–0.3 μm in diameter. Particles in this size range are most easily able to follow the air flow through the porous filter matrix (an example of filter structure is shown in Figure 2). Super-micrometer particles have too much inertia to follow the curved air flow, and are deposited on the filter fibers due to impaction. Particles finer than 0.1 μm experience substantial Brownian motion, as gas molecules collide with them. The wiggling Brownian motion increases the probability of particles to collide on filter fibers. The collection efficiency in the particle size range below 0.1 μm is therefore the higher the smaller the particle is. Generally, the filters with a good collection efficiency also produce a high flow resistance. Collection efficiency  $E$  can be increased by using two consecutive filters, but that also doubles the pressure drop caused by the filter,  $\Delta p$ . A quantity called quality factor,  $Q_f$ , can be used to

compare the collection characteristics of filters with different pressure drops [3],

$$Q_f = \frac{\ln(1/(1-E))}{\Delta p}. \quad (2)$$

Quality factor describes how large pressure drop is needed to collect the particles with a given efficiency. Here it is assumed that a two-stage filter consisting of two similar layers placed one upon the other have the same quality factor than one filter alone. Thus if the first layer collects 70% of the 0.2 μm particles, then the second layer is assumed to collect 70% of those 0.2 μm particles that were not collected by the first layer (that is 21% of all the 0.2 μm particles). The overall efficiency of this hypothetical two-stage filter would be 91% for 0.2 μm particles.

Collection efficiency is likely to change during sample collection, as the particles collected change the structure of the filter matrix. The phenomena is the most dominant for electret-type filters. They utilise fibers made of charged and insulating material so that one side is positively, and the other



**Figure 2.** A scanning electron microscope photograph of a fibrous filter. The scale is shown under the photograph.

negatively charged, resulting a zero net charge. Electrostatic attraction enhances collection efficiency, especially of 0.1–1  $\mu\text{m}$  particles that are the most difficult to collect otherwise. However, particles collected on the filter may mask the charge and reduce collection efficiency [3,5]. Also, the fiber charging can be lost when electret filters are exposed to, e.g., high humidity or organic liquid aerosols.

The filter with the lowest initial pressure drop is not necessarily the most resistant to clogging. The pressure drop increase rate during air sampling depends on filter properties, such as fiber diameter and the fraction of the filter volume that is covered by fibers. The larger the empty space between the fibers, the lower the clogging rate usually is. The properties of the collected particles, such as particle size distribution and particle surface properties, also affect the clogging rate.

### 3.2 Concentration and size distribution of the atmospheric aerosol

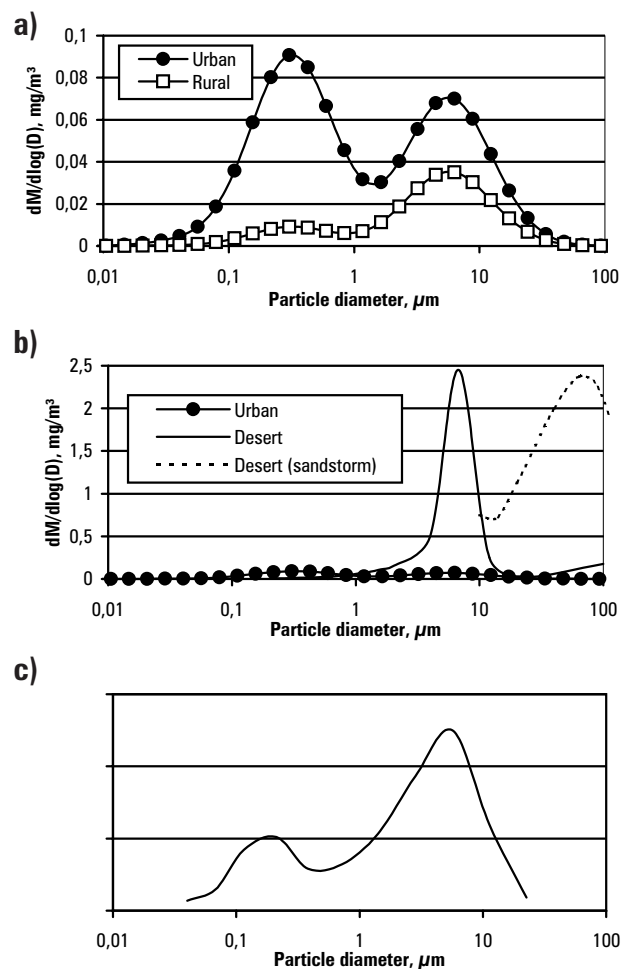
The mass concentration and size distribution of airborne particles, and therefore also the filter clogging rate, varies depending on the environment. There is two airborne particle modes with a different formation mechanism, and consequently a different particle size. Fine particles (diameter below 1  $\mu\text{m}$ ) are formed as gases and vapours are condensed. Atmospheric fine particles are mainly formed in various combustion processes, such as power plants and motor engines, where the substances are volatilised in high temperature. In addition, gases released in combustion processes form fine particles in atmosphere, as they are converted to condensed species by chemical reactions (such as conversion of gaseous  $\text{SO}_2$  to condensed  $\text{H}_2\text{SO}_4$ ). Coarse particles (diameter larger than 1  $\mu\text{m}$ ) include wind-blown soil, dust and other particles released to air mechanically, that is without volatilisation. In marine conditions coarse particles are predominantly sea salt that are released to air mechanically as sea-water droplets, followed by evaporation of water.

Urban aerosol is dominated by the fine particle mode. Particle concentration, especially that of the fine mode, is lower in rural continental areas as compared to urban areas (Figure 3.a). There is large spatial variation in aerosol mass concentration. It is typically more than 0.1  $\text{mg}/\text{m}^3$  in large

cities, and around 0.04  $\text{mg}/\text{m}^3$  in rural areas. Desert aerosol is characterised by a high concentration (may exceed 1  $\text{mg}/\text{m}^3$ ) of coarse wind-blown crustal material (Figure 3.b). Wind velocity strongly affects the mass concentration and particle size in desert, as high velocity wind raises very coarse (> 10  $\mu\text{m}$ ) particles from the ground. Particles in the size range of 10–100  $\mu\text{m}$  have been found to dominate the total airborne mass concentration during sandstorm in Sahara [6].

### 3.3 Filter tests

Four filters were tested for collection efficiency and pressure drop during exposure to aerosol that contained 25 weight-% of diesel fume generated by a motor boat engine, and 75 weight-% of SAE fine dust (Figure 3.c). Diesel fume simulates combustion-originated sub-micrometer airborne particles,

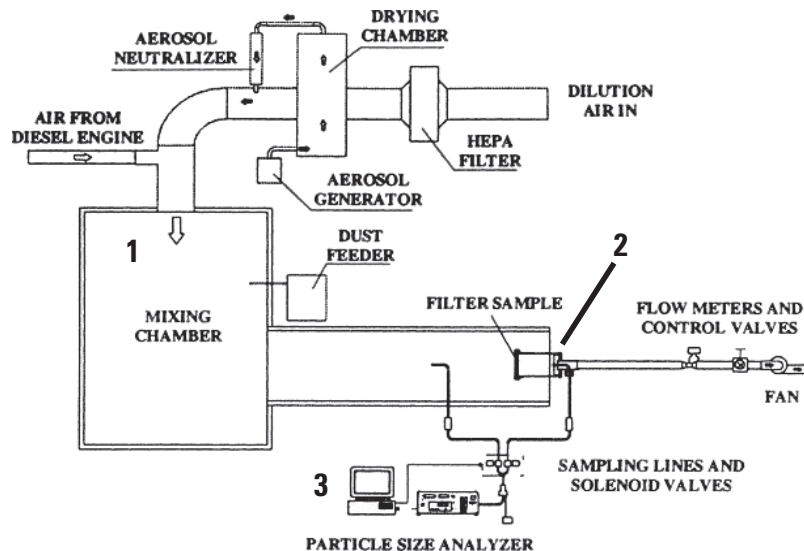
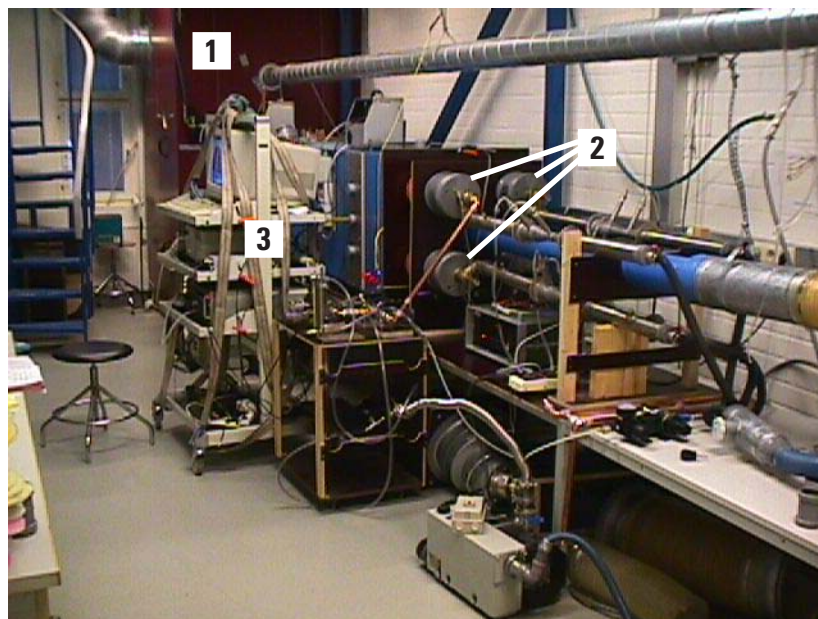


**Figure 3.** *a) and b) Typical aerosol particle mass size distributions (based on references [3,6 and 7]. Note the different scales in ordinate axis. c) The mass size distribution of the aerosol used for filter exposure in our tests.*

and SAE fine dust is a typical test dust for coarse wind-blown dust. Our test aerosol resembles continental rural area aerosol, whereas in urban areas fine particle mode is more dominant. If marine aerosol was to be simulated, the coarse particle contribution to total mass concentration should be about 95%, and the coarse particles should be salt instead of crustal dust used here.

The experimental set-up is shown in Figure 4. Collection efficiency was determined by measuring size distribution of di-ethyl-hexyl sebacate (DEHS) aerosol with an optical particle size analyser (PMS LAS X) alternately upstream and downstream of the filter.

Collection efficiency was measured for unexposed filters and after exposure to  $2.5 \text{ mg/cm}^2$  and  $5 \text{ mg/cm}^2$  of aerosol (Figure 5). Here exposure is defined as a mass of particles in air volume that is drawn through a unit area of filter. The values  $2.5 \text{ mg/cm}^2$  and  $5 \text{ mg/cm}^2$  correspond to a one-week operation of Hunter MKII (flow rate  $150 \text{ m}^3/\text{h}$  and filter collection area  $0.1 \text{ m}^2$ ) in an environment where the particle mass concentration is  $0.1 \text{ mg/m}^3$  and  $0.2 \text{ mg/m}^3$ , respectively. The measured collection efficiencies are shown in Figure 6, the pressure drops in Figure 7 and the quality factors calculated from equation (2) to  $0.2 \mu\text{m}$  particles in Figure 8.

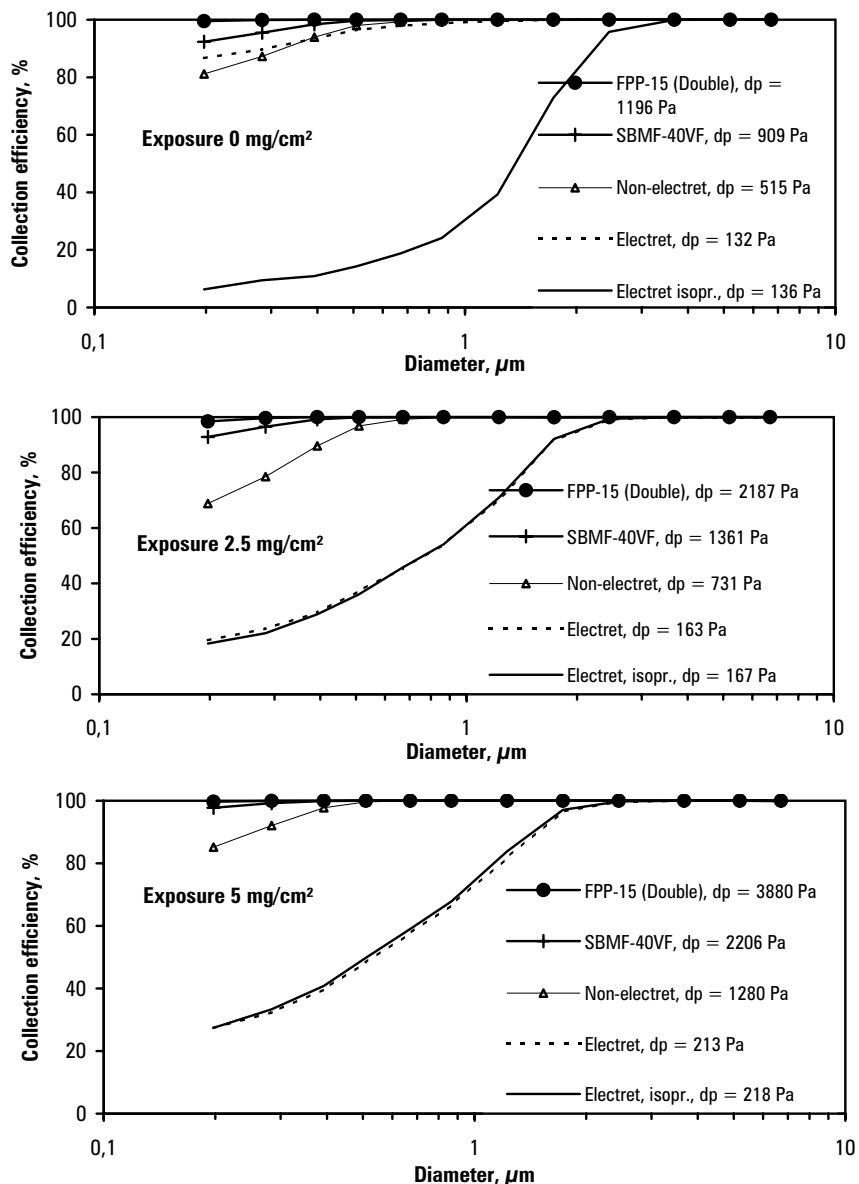


**Figure 4.** The experimental set-up for collection efficiency measurements. **1** = Mixing chamber, **2** = Air outlets downstream of the filters (four filters can be measured simultaneously, one is shown in the drawing), **3** = Particle size analyzer and computer.

One of the filters tested was of electret-type. It had a superior quality factor as unexposed. The good initial performance was shown to be based on filter charging, as 24 hours treatment with isopropyl alcohol damaged its capability to collect sub-micron particles. Even an untreated piece had lost the gain from fiber charging by the time exposure was  $2.5 \text{ mg/cm}^2$ , when it collected  $0.2 \text{ }\mu\text{m}$  particles only with an efficiency of 19%. Further exposure to  $5 \text{ mg/cm}^2$  slightly increased the collection efficiency, apparently due to the enhancement of non-electrical collection mechanisms. There was no significant difference between the isopropyl-treated and untreated sample at the exposures of  $2.5 \text{ mg/cm}^2$  and  $5 \text{ mg/cm}^2$ .



**Figure 5.** A filter exposed to  $5 \text{ mg/cm}^2$  of aerosol containing 25 wt-% of diesel fume particles and 75 wt-% of SAE fine dust. Diameter of the darkened area, where the particles are collected, is 15 cm.



**Figure 6.** The dependence of collection efficiency on filter exposure. Pressure drops are shown in legend. Isopr. = 24 h treatment in isopropyl alcohol.

Two of the filters, Petrianov FPP-15 (Double) and SBMF-40VF, were able to collect 0.2  $\mu\text{m}$  particles with an efficiency better than 90% at all the three exposure levels. Petrianov FPP-15-1.5 was tested as double-layer configuration, two filters one upon the other. The advantage as compared to the normal single configuration is the increased collection efficiency, and the disadvantage is the higher pressure drop. For easy handling, the nominal collection sides of the two FPP-15 filters were against each other (as delivered) so that the filter on the top was upside down. Thus the collection on FPP-15 took place on the side other than the one recommended by manufacturer.

The decision between FPP-15 (Double) and SBMF-40VF depends on the application. FPP-15 (Double) is better if the collection efficiency is to be maximised, but SBMF-40VF produces a lower pressure drop. Apparently FPP-15 (Double) is bet-

ter if sampling time is so short, that clogging is not a problem. The difference in collection efficiencies becomes less significant when the exposure is increased, as the collection efficiency of both filters approaches 100%. Thus the collection efficiency becomes less significant selection criteria, if the collection time is extended. SBMF-40VF appears to be better if the collection time should be as long as possible, because the pressure drop of FPP-15 (Double) is increased faster than that of SBMF-40VF. It should be noted, that the conclusions drawn here are limited to the filters when used as configurations similar to those tested here. For instance, single-layer FPP-15 produces a lower pressure drop, and may therefore be more preferable to some applications than the FPP-15 (Double) tested here. The fact that we collected particles on the bottom side of FPP-15 may also have affected the results.

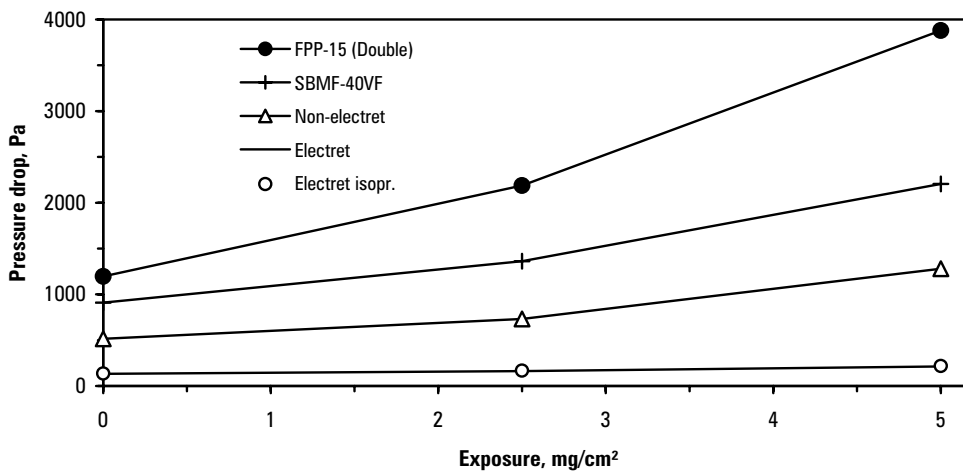


Figure 7. Pressure drops as functions of exposure (the same test runs as in Figure 6).

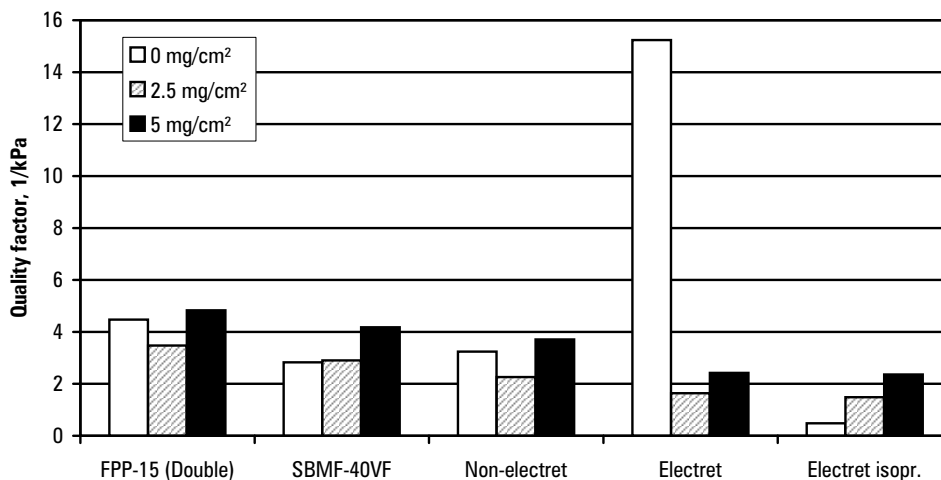


Figure 8. Quality factors for 0.2  $\mu\text{m}$  particles when filters were exposed to 0–5 mg/cm<sup>2</sup> of aerosol (the same test runs as in Figure 6).

## 4 Pre-filtration of coarse particles

### 4.1 Motivation

Natural uranium in air is present in coarse particle mode associated with wind-blown dust. Pre-filtration of coarse particles may increase the signal to background ratio, since the freshly released uranium and plutonium are presumably enriched in the fine particle mode. This is because the volatilised species, such as  $UF_6$ , are more easily accidentally released than the condensed ones. Species that have volatilised, followed by condensation, are enriched in the fine mode [3]. Besides, the species attached to coarse particles can be found only if the sampler is located near the source, unless the species were released to a significant height (Table I).

Collection of coarse particles from air separately upstream of the sampling filter serves also another purpose. The filter clogging rate is decreased, if clogging is caused predominantly by coarse particles. The benefit from pre-filtration is presumably largest in desert aerosol conditions (especially during sandstorm), where airborne particles are dominated by coarse crustal particles.

### 4.2 Methods

Pre-separation of coarse particle is usually carried out with devices utilising impaction. The sampled air flow makes abrupt curves that the coarse par-

ticles are not able to follow due to their inertia, but are deposited on the collection surface. For instance, sampling inlets to meet PM-10 criteria collect particles with aerodynamic diameter larger than  $10\ \mu\text{m}$  [3,8]. This kind of sampling inlets are meant to be permanently leak-tightly assembled upstream of the filter cassette. A sampler with automatic filter changing can not easily apply a permanently assembled inlet, because there is a need for a sealing between two parts (inlet and filter cassette) that move relative to each other. Instead, an alternative system consisting of two consecutive filters, with the first one acting as a pre-filter, was applied here. The pre-filter is significantly more porous than the collection filter, and thus both the clogging rate and the collection efficiency are lower for the pre-filter. In our configuration, the pre-filter and the collection filter are placed directly one upon the other.

### 4.3 Tests

Four different filter configurations were tested;

- i) a single collection filter (Petrianov RFM-1.7),
- ii) two consecutive collection filters,
- iii) a collection filter preceded by a non-electret pre-filter and
- iv) a collection filter preceded by an electret pre-filter.

**Table I.** Distance that a particle travels with the air flow during the time it settles down from a given height. Values are for spherical particles with a density of  $1\ \text{g}/\text{cm}^3$ . Gravitational settling is assumed to be the only mechanism causing vertical particle movement (no vertical advection).

Diameter, $\mu\text{m}$	Deposition velocity, m/s	Time to settle down from a height of		Horizontal distance travelled during settling (wind velocity 1–10 m/s)	
		10 m	100 m	10 m	100 m
10	0.003	54 min	9 h	3–30 km	30–300 km
100	0.25	40 s	7 min	40–400 m	0.4–4 km

Configuration i) is the conventional case, with no pre-filtration. Configuration ii) is similar to i), except that it has a back-up filter to increase the collection efficiency. A separate analysis of the back-up filter gives a sample with a very high enrichment of sub-micron particles.

Configurations iii) and iv) provide a pre-filter. The electret pre-filter collects fine particles more efficiently than the non-electret one, at least during the early stage of sampling. The electret pre-filter is meant to be used when the filter clogging rate reduction is the main motivation for pre-filtering, and the non-electret pre-filter when a sharp size-classification between the fine and coarse modes is more important than the clogging prevention.

Filters were exposed to three different kinds of aerosol. The aerosol used in collection efficiency tests (mixture of diesel fume and SAE test dust, Figure 3.c) was also used here. In addition, diesel fume alone was used to represent sub-micron particles, and SAE test dust alone to represent coarse particles.

The pressure drop during exposure is shown in Figure 9. Pre-filter decreases clogging rate, especially when particles are coarse. If particles are fine, only a small fraction of them is collected on the pre-filter, and consequently the clogging rate

is not remarkably decreased. Figure 10 shows, that the collection filter located downstream of the pre-filter is almost clean when particles were coarse, but heavily loaded when particles were fine.

The pressure drop of the two-stage filter configuration ii) is initially two times that of the single filter i), but the difference is not increased during exposure. In the two-stage system, most of the mass is collected on the first layer, and consequently the additional pressure drop developed during exposure is that developed on the first layer alone. Thus, additional filter layers do not increase clogging rate significantly, as far as the initial pressure drop with the additional layers remains low as compared to the maximum acceptable pressure drop.

#### 4.4 Simulation of air sampler flow rate

The air flow rate maintained by a sampling pump is the lower the higher the pressure drop over the filter is. Filter clogging finally results in a situation where the flow rate is too low to be acceptable, or the pump has to be stopped to avoid overheating. Figure 11 shows the simulated flow rate for Hunter MKII during sample collection with the following assumptions:

- Flow rate maintained by the pump depends on

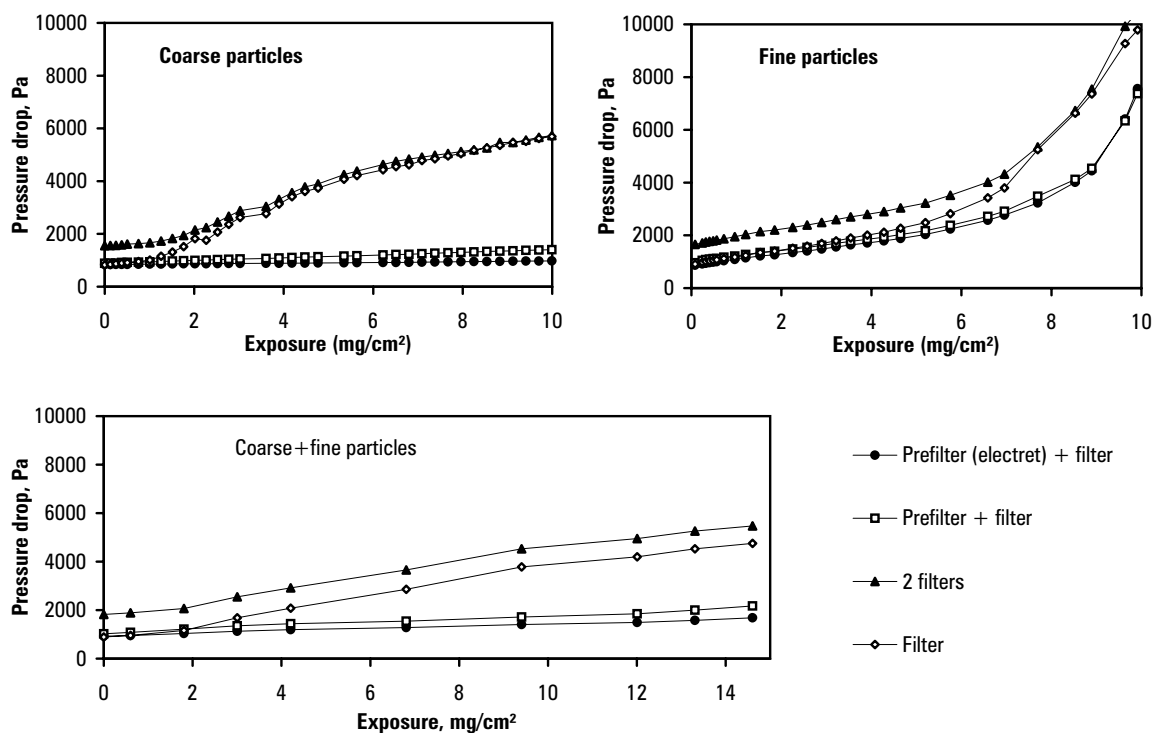


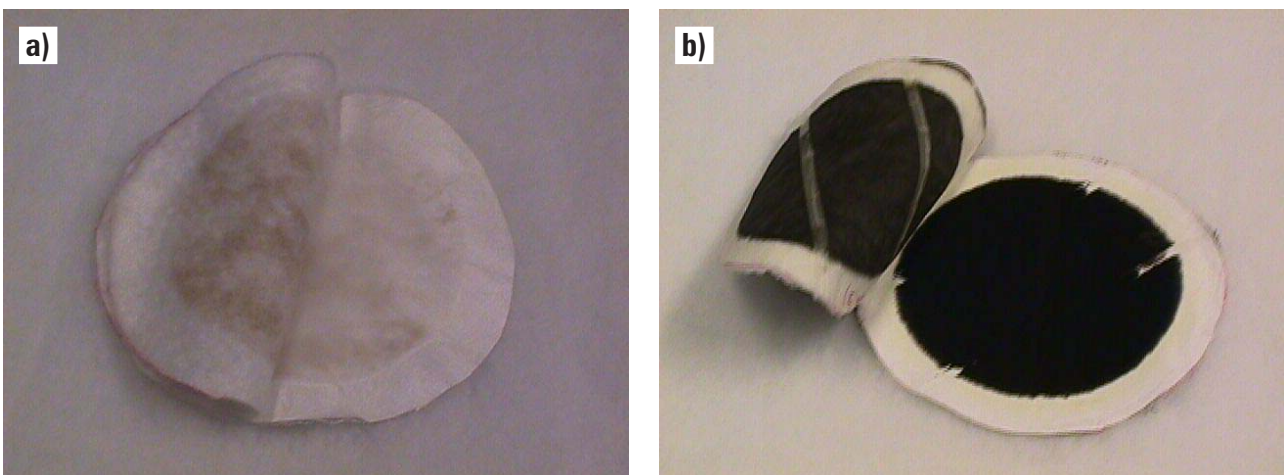
Figure 9. Pressure drop during exposure at face velocity of 0.4 m/s.

the pressure drop as described by equation (1)

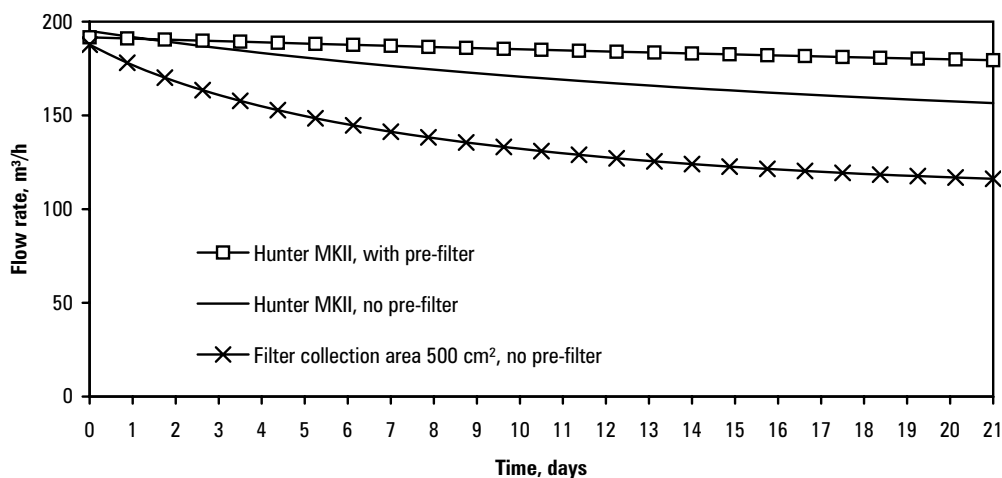
- Pressure drop at a face velocity of 0.4 m/s depends on exposure as in Figure 9 (coarse+fine particles). In other words, the properties of the airborne particles and of the filter are similar to those during our exposure tests.
- Pressure drop is directly proportional to the face velocity.
- Mass concentration in air is  $0.2 \text{ mg/m}^3$ , which is a typical value in urban areas. Having other parameters unchanged, the time for the flow

rate to drop to a given value is directly proportional to the mass concentration.

Figure 11 shows, that doubling the filter area from 500 to  $1000 \text{ cm}^2$  increased collection time significantly. Also pre-filtration extends the collection time in this example, because there is a high concentration of coarse particles in air. If there is a network of air samplers being built, simulation can be helpful in the process of applying practical experience from the first samplers to design modifications to the later samplers.



**Figure 10.** The pre-filter + collection filter system after exposure to  $5 \text{ mg/cm}^2$  of **a)** coarse particles and **b)** fine diesel fume particles. Pre-filter is turned away, and the upside of the collection filter is shown.



**Figure 11.** Simulated flow rate of Hunter Automatic MKII (filter collection area  $1000 \text{ cm}^2$ ) assuming that the mass concentration of airborne particles is  $0.2 \text{ mg/m}^3$  and that they are similar to the mixture of diesel fume and coarse dust used in our tests. Flow rate is also shown for a sampler with a half-size collection area.

## 4.5 Practical experience from field trial in Kazakhstan

Pre-filtration was tested during field trial in Kazakhstan [9,10]. Two Hunter MKII samplers were utilised next to each other. The electret-type pre-filter was used in one of the samplers during six separate weeks, otherwise there was a single collection filter only. The other sampler was equipped with a permanently assembled impactor-type filtering unit to remove particles larger than 13  $\mu\text{m}$  [11]. The sampling period was usually one week, longer sampling periods up to 4 weeks were also tested. Mass concentration was 0.1–0.25  $\text{mg}/\text{m}^3$ . Filter dust loading did not cause clogging-up problems to any of the filter configurations. The analysis results are presented in [12,13]. The following practical observations were made during the field trial:

- The preparation of two-stage filters before field campaign is an additional work task. However, working time can be spared if the coarse particle fraction is also of interest, as it can be conveniently recovered with the two-stage filter.
- The upside of the two-stage filter must be clearly indicated to avoid upside-down loading during sample collection. This is not so acute with single filters. Collection on the wrong side of a single filter does not create a serious problem with most filter types, even if also single filters usually have a preferred upside.
- The physical separation of the pre-filter from the collection filter at laboratory did not involve difficulties. Particles were collected inside the pre-filter matrix, and were not detached to a significant extent during the filter handling process. Detachment could be a problem, if particle loading was so high, that the particle deposit layer begins to grow on the surface of the pre-filter. If there is loose particulate matter present in the transport package, it presumably originates predominantly from the pre-filter.

## 5 Conclusions

An automatic air sampler for WAES, Hunter Automatic MKII, was designed and built. The device enables unattended operation for 6 weeks, during the time six weekly samples are collected. Several modifications were made to the original samplers. To avoid filter clogging, the filter collection area was doubled, filter material was changed, and an optional pre-filtration method was designed to remove coarse particles. Due to their negligible uranium concentration and ashability in low temperatures, the presently used organic filter materials are more suitable for chemical analysis methods than the previously used glass-fiber ones.

The filter exposure tests and simulation results show, that with an applicable filter (possibly preceded by a pre-filter), the sampler is easily able to operate for one week without clogging, if the airborne particle mass concentration is that typical in desert conditions. This can be done without compromising the specification for collection efficiency (must collect more than 80% of the 0.2  $\mu\text{m}$  particles). However, the functioning in real-conditions can be verified only on the spot, as we do not know beforehand if the conditions on site can be considered "typical". Especially sand storms set a

challenge to any air sampler. Very high ambient temperature may also decrease the operation time before the sampler is stopped for overheat protection. Simulation method demonstrated in this report provides a tool for sampler modifications during sampler-network build-up, if the first samplers taken into operation show clogging, or other need for modifications.

The pre-filtration of coarse particles has two advantages. It decreases the filter clogging rate, if the airborne particles contain large fraction of coarse particles, as is the case in desert conditions. In typical urban aerosol conditions pre-filtration is less beneficial. Secondly, the absence of coarse particles is expected to improve detection limit in chemical analysis, as the artificial radionuclides are most likely enriched in the fine particles, and the natural uranium in the coarse particles. The downside is, that there is no absolute certainty on the absence of artificial nuclides in the coarse fraction, and separate analysis of both fractions doubles the number of samples. The decision, whether pre-filtering is used or not, is to be made based on the precise objectives of the campaign in question.

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## APPENDIX

### EXAMPLE OF THE SAMPLER OPERATION PROCEDURE

Each sampling station collects weekly samples. An inspector visits each of the samplers every six weeks, unless the state-of-health data indicates problems.

During the normal filter-changing visit to a sampler, the inspector should carry the following items with him/her:

- Six new filters stored in plastic bags.
- Two pairs of clean disposable rubber gloves.
- Watch and ball pen.
- Six empty large plastic bags.
- Logbook.
- Organic solvent, e.g. alcohol, to clean filter holder frames.
- Empty bag for waste.
- A data sheet for each of the six new filters, filled in with the following information; Filter number, Sampler ID, Scheduled sampling week, Scheduled filter position at sampler (position 1 for first filter of the six weeks cycle, etc), Filter loading date, Name of the inspector.

During the visit to a sampler, the inspector carries out the following operations:

#### Removal of used filters from sampler

- Open sampler front cover and lay down new filters on **left** top shelf of sampler.
- Write down in logbook the following information; Is power on?, Which filter is in sampling position (number six down indicates that automatic filter changing is completed)?; Condition of sampler based on visual observation (e.g. indication of tampering, dust, power is on, etc.); Number of records in the data terminal; Date and time when the sampler was stopped.

- Stop sampler, Disconnect data terminal, take it into laboratory and start data unloading to computer (unloading takes time!).
- For the filter in front of you, select the proper data sheet (check filter number) and fill in Condition of filter prior to removing it (is it whole, well in place, evenly loaded, etc), Actual filter position at sampler, Date and Your name.
- Remove used filter from sampler using clean disposable rubber gloves, insert the used filter straight into large plastic bag and seal. Check filter number against number on the data sheet.
- Lay used filter in plastic bag on **right** top shelf of sampler.
- Repeat the previous 3 steps to remove other filters one by one.
- Clean air sampler frames from dust, using organic solvent, if needed.

#### Loading new filters into sampler

- Put on new clean disposal rubber gloves
- Take a new filter (from **left** top shelf), and the relevant pre-filled data sheet. Check filter number against the number in the data sheet.
- Confirm that the filter position of sampler (position 1–6) is correct.
- Take a new filter from plastic bag, set it in place in sampler and close the fixing frame.
- Repeat the previous 3 steps to load other new filters.
- Rotate sampler holder, as needed, and leave position number 1 down (that is, on sampling position).
- Connect data terminal to sampler after completing data unloading and deleting old files.
- Start sampling and close the sampler cover.
- Fill in Logbook.