STUDIES ON AEROSOL PARTICLE EMISSION, SAMPLING AND AUTHENTICITY

Sampo Ylätalo

STUK Radiation and Nuclear Safety Authority

ACADEMIC DISSERTATION

To be presented with the permission of the Faculty of Science of the
University of Helsinki, for public criticism in Department of Physical Sciences,
Gustaf Hällströmin katu 2, Auditorium D101 (Physicum),
on 19th May 2006 at 12 o'clock noon

University of Helsinki
Department of Physical Sciences

Report Series in Physics HU-P-D129

ISBN 952-10-2113-6

ISSN 0356-0961

ISBN 952-10-2114-4 (pdf)

Abstract

Emissions of coal combustion fly ash through real scale **E**lectro**S**tatic **P**recipitators (ESP) were studied in different coal combustion and operation conditions. Sub-micron fly-ash aerosol emission from a power plant boiler and the ESP were determined and consequently the aerosol penetration, as based on electrical mobility measurements, thus giving thereby an indication for an estimate on the size and the maximum extent that the small particles can escape. The experimentals indicate a maximum penetration of 4% to 20 % of the small particles, as counted on number basis instead of the normally used mass basis, while simultaneously the ESP is operating at a nearly 100% collection efficiency on mass basis. Although the size range as such seems to appear independent of the coal, of the boiler or even of the device used for the emission control, the maximum penetration level on the number basis depends on the ESP operating parameters. The measured emissions were stable during stable boiler operation for a fired coal, and the emissions seemed each to be different indicating that the sub-micron size distribution of the fly-ash could be used as a specific characteristics for recognition, for instance for authenticity, provided with an indication of known stable operation.

Consequently, the results on the emissions suggest an optimum particle size range for environmental monitoring in respect to the probability of finding traces from the samples. The current work embodies also an authentication system for aerosol samples for post-inspection from any macroscopic sample piece. The system can comprise newly introduced new devices, for mutually independent use, or, for use in a combination with each other, as arranged in order to promote the sampling operation length and/or the tag selection diversity. The tag for the samples can be based on naturally occurring measures and/or added measures of authenticity in a suitable combination. The method involves not only military related applications but those in civil industries as well. Alternatively to the samples, the system can be applied to inks for note printing or other monetary valued papers, but also in a filter manufacturing for marking fibrous filters.

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CONTENTS

“*Did Cartesius have any independent empirical proof on his thoughts?”*

(Sampo Ylätalo: 22th December 2005)

ACRONYMS

AAAA	An Aerosol Authentication Apparatus
AC	Alternating Current
AFM	Atom Force Microscopy, or -Microscope
CINDERELLA.STUK	A specific type of automated high-volume sampler at STUK
CPC	Condensation Particle Counter
DC	Direct Current
DMA	Differential Mobility Analyzer
EDB	Electro Dynamic Balance
EDP	Electro Dynamic Precipitator
ESP	Electro Static Precipitator
GSD	Geometric Standard Deviation
ID	Identity
IITA	Impactor Inlet Through-put Array
IT	Information Technology
LATEX	A Suspension of Particle Material
MICRON	A data inversion algorithm with constraints
SCA	Specific Collection Area (of an ESP)
SEM	Scanning Electron Microscopy or -Microscope
SIHTI	A roof-organization for a research project of an ensemble of projects including aerosol research
STUK	Finnish acronym for Radiation and Nuclear Safety Authority (Säteilyturvakeskus in Finnish)
TEM	Transmission Electron Microscopy or -Microscope
TSI	Manufacturer of Aerosol Measurement devices, met also with the device model number
UTHSCA	A graphical tool algorithm for particle post analysis from images
VIE-11 cm	A DMA-type designed in Vienna with the indicated analyser specific length
VTT	Finnish acronym for Technical Research Center of Finland (Valtion Teknillinen Tutkimuskeskus in Finnish)
WAES	Wide Area Environmental Sampling
XRF	X-Ray Fluorescence

I INTRODUCTION

In the dominating short time-scale economy of the industry, developing countries cannot afford new technologies. A strong temptation may lead to ignoring the environment, keeping the countries outside the related treaties and driving them to repeat the mistakes of the industrialised world. Also, the industrialized world may ignore their own share in the scenario to reduce the emissions. Whatever the scenario, modern burners, filters and gas cleaning devices are badly needed, not only in the developing countries, which are rich-populated, about to industrialize right now, and whose electricity increase will consequently dramatically increase the emissions associated with the increasing living standards, but in the developed countries all over the world as well.

The suspended mass, used in fly-ash emission estimation, is not sufficient when estimating the emission in respect to health. Small particles between 0.1 µm and 3 µm in size contribute to the suspended mass only within few per cent, but the number fraction is much larger, and can easily exceed 90 % of the total number of the particles. Their residence time in the atmosphere is longer than within the large sizes, thus also contributing to remote area transport and deposition on the background areas. Due to the shape and mechanical properties, these small particles can deposit very deep in the human respiratory system and stay there as attached exposing the tissues for a long time, even for years. Because of the formation mechanism of these small particles, on their surfaces, they contain water-soluble substances, which may be poisonous to any life forms, including the human life form. While accumulating and concentrating, the chemicals in the particles are released in moist conditions inside the lungs. The consequences vary between allergy and even much worse. It is namely shown that small particle concentration among other pollutants correlates with mortality (Dockery et al. 1993). It is thus important to observe the emissions in order to develop suitable counter measures and/or controlling techniques, whether or not the surveillance is occurring either on-site or in the remote environment. New techniques are needed for the measurement and for tuning the existing devices for their optimum operation.

As an alternative to coal, nuclear power has been considered. Aspects of weapon manufacturing control, and the emission risk in disaster and/or in a terrorist act, make the international surveillance necessary for the nuclear material and related emissions in wide areas. In such environmental monitoring, radioactive substances can be monitored by high-volume samplers. In dust- or sand-laden conditions piling sands can be a problem. As a solution, rejecting the irrelevant matter can extend the sampling time and thus smaller concentrations of the monitored matter can be revealed. In close relation to military applications, sample authenticity is an important aspect.

The know-how of producing nuclear weapons and/or biological war material has been lately become well-known world wide, including unstable developing countries. International supervision requires reliable samples for reporting and thus means to indicate the authentic sample origin. Even any potential thread on forgery of such delicate samples can create crises but also motivate developing of systems to establish a link between the on-site-collected sample and the observations of its constituents reported on the paper. Mistrust can be avoided by measures that are sufficiently reliable and arranged to be present in the sample.

In case of doubt, for restoring the confidence such measures need to be taken that can be relied upon when the chain from the report to the sample is verified backwards. The last link of the chain, the sample itself, plays a very important role and thus its authenticity should be secured. It is fatal for international relations, for instance, if an environmental sample were changed - even accidentally - with another one so indicating false nuclear manufacturing activities.

However, authentic origin of man made substances is not a common question only in various fields of delicate laboratory analysis in the military world but also in chemical technology or closely related commercial civil industrial fields as well. Industrial business, having the focus on preventing illegal copying or patent infringement, has used to some extent the existing impurities of the precursors used in a process to prove an authentic product made in the process. But the impurity composition can vary in the raw materials. Thus, the new method of this work can help not only in authenticating for instance inks for note printing or other monetary valued papers, but it may also be of value in a filter manufacturing for marking fibrous filters.

II OBJECTIVES OF THIS WORK

In general terms, the aim of this work relates to emission determination techniques via aerosol particle sampling and also to sample authenticity. The scope of this work comprises a summary of several emission and fly ash aerosol studies made in real-scale power plants. Within the scope, new devices are introduced for sampling and to insure sample authenticity. Thus, the objectives of this thesis relate not only to questions of determination of particulate emissions from coal fired power plants, but also to means to indicate the sample authenticity and to development of devices that can be used in corresponding duties. The thesis is primarily based on publications I-IX, which are attached as appendices, relating to the following objectives specified in more detail below.

The first objective of this work is to apply electrical mobility based measurement techniques in power plant environments for determination of emission of small particle fly-ash size distributions and, consequently, to characterize electrostatic precipitator (ESP) performance in different conditions of coal combustion and operating parameters of the ESPs that have an influence on the penetration of the fly-ash aerosol by the indication in what size and to what extent the particles can escape from a commonly used gas cleaning unit.

The second objective of this work is to determine the emission from a commonly used gas cleaning device and thus give an indication of the size of the particles that will most likely escape the controlling ESPs, and thus give an indication/confirm what kind of particles are advantageous to search for instance for the purposes of environmental monitoring.

The third objective of this work is to introduce new devices that can be used independently or in combination with each other in the authentication techniques to promote the sampling operation length and the diversity of the tag selection and also to facilitate automatic post-analysis for particles on an authenticated substrate.

The fourth objective of this work is to embody a system to authenticate environmental aerosol samples and to facilitate inspection of the authenticity of the sample reliably from any macroscopic piece of the sample, even after several years or decades from the event of the sampling and without disturbing the actual sample content.

II.1. PAPERS AND THE OBJECTIVES

Papers I-III relate to the first and the second objectives and demonstrate accurate size distribution determination in coal fired power plant conditions, as based on electrical mobility. Common to Papers I-III is the sampling system that was used to take out aerosol samples of small particles from the flue gas stack in a high temperature, to dilute the aerosol to the ambient temperature and to analyze the particle size as based on the electrical mobility. The small particle size distributions were determined in three different power plants in order to discover the electrostatic precipitator's collection efficiencies. For this purpose, not only emission from the boiler to the ESP inlet was determined, but also the emission at the outlet of the ESP. Such technique to measure size distribution as based on electrical mobility has been available in laboratories, but it has so far not been used with similar instruments in industrial conditions.

In the Paper B, the results of Paper I were evaluated in respect of the data-processing algorithm. The differential mobility analyser manufacturer’s simple algorithm and an algorithm with constraints, MICRON (Wolfgenbarger and Seinfeld, 1988) were used in data reduction and the results were compared with each other. In paper B, the comparison indicated good agreement between the two algorithms and further that the findings of Paper I were not constraint-driven and are thus independent on the algorithm used in data reduction.

In Papers I-III, the power plants used different types of coals. They had different conditions of firing and a different kind of electrostatic precipitator operation. In Paper I, the power plant A (Fig.1, Table 1) was operated within a 2/3-level of full power and the last block of the electrostatic precipitator was not fully switched on duty during the measurements. Paper III describes measurements for the electrostatic precipitator performance in the same power plant as Paper I. However, in Paper III the measurements were made mainly as a repetition to those described in Paper I, and once again a year later in conditions as identical as possible (Tables 1, 2 and 3 respectively for the sub-micron peaks, coals and ashes for the power plant A). All the blocks of the electrostatic precipitator were switched on, but an opportunity was available for the measuring with the last block switched off and was utilized for obtaining the data shown in Paper III.

Paper II describes measurements made in two different power plants, B and C (see Fig.1, Table 1) and with two types of coals. Details of the measurements, the boiler operation as well as the electric fields are accounted for in Papers II and III, and even more in detail in the Licentiate thesis of Sampo Ylätalo (1996) (Paper A). The sampling locations in the power plants are shown in Figure 1.

In Papers I-III, as relating to the second objective, the penetration of the fly ash occurred always within the same size range and almost independently of the fired coal or the precipitator, although the penetration level was depending on the operation parameters. It also turned out that each power plant had its own small-particle size distribution in sub-micron as a result from the coal conversion process. In suitable conditions, the small particle size distribution could thus be used as a coal conversion process-specific trace.

In a similar manner, sub-micron size distributions can also be measured from other stacks, i.e. in ambient temperature and from high volume sampler's sampling line, but even in a more simple manner and thus preferably without the dilution. The sample size-distribution can be attached to the authenticity information as a natural measure of authenticity, where applicable (Table 6, Figures 2-7). Using the DMA (Differential Mobility Analyzer) facilitates also sampling onto a substrate via the device of Paper VI for post-counting analysis from the substrate such as TEM-grid.

Paper IV relates to the third and the fourth objectives and introduces a new device that can be used with a sampler at a sampling site in order to disperse an aerosol tag into the sample filter and the textures of it automatically so that any macroscopic piece of the sample can be authenticated. The authentication is based on observations of the tag properties found on the sample filter and on a comparison thereof with certain known features of the tag. Paper IV demonstrates the use of synthetic measures of authenticity by utilization of aerosol particles that are selectable for their size.

Paper V relates to the third and the fourth objectives and introduces a pre-filtration unit designed for collecting particles by impaction as such but applied in a new way so that it can be used in front of a sampler and thus restrict the entry of particles into the stack of the high volume sampler for environmental sampling. Such a unit was embodied as an impactor with a cut-size facilitating even more diverse tag combinations. Thus, the pre-filtration unit in Paper V supports not only extending the sampling duration length but also the promotion of the tag diversity by cutting out of the inlet size distribution the particles larger in size than the cut-size of the pre-filtration unit. The pre-filtration unit in this work was designed also for integration into the case of the sampler itself.

Papers VI (**and VII,** the translation of paper VI into English), relate not only to the third and the fourth objectives, but also to the second objective, as introducing a new device, an EDP-device (electrodynamic precipitator) that can be used for sampling in combination with a differential mobility analyzer (DMA), as demonstrated later, for providing a particle sample on a substrate for microscopic analysis. By the substrate selection, the sample can be photographed and addressed to automatic post-counting. The fibrous filter as such, demonstrated as authenticated in accordance with Paper IV, hides all kinds of particles into the textures. Post-counting of the particles, including the sub-micron, is extremely difficult to be performed from such a sample. Substrates other than filters can also be authenticated according to the principles adopted from the Paper IV. The EDP-device was patented in Paper VI, which is the patent application as allowed.

Paper VIII, in relation to the objectives of Paper VI, describes preliminary test runs and results achieved in laboratory conditions, for the purpose of facilitating a new way to collect particles and also for the purpose of post-counting, for which purpose the EDP-device was designed and built as embodied in Paper VI. The EDP-prototype was operated on one hand for determination of particle removal efficiency and on the other hand for the collection efficiency on the substrate to be taken out for a post-count analysis in a microscope.

In addition, the EDP-device was used for Kernel determination for the DMA, but unfortunately the TEM-microscope turned out to yield too small particle sizes. In order to distinguish the difference as well as to indicate the correct samples, large particles of 100 nm were used for the purpose as a tag, and they demonstrate a nano-scaled authentication. The EDP-device's particle removal efficiency was studied in several test runs, which showed that the removal efficiency was dependent on the particle's electrical mobility. The DMA appeared ideal as a particle-size selector because of the particle charge number was well defined in the nano-scale operation. An analytic formula between the observed removal efficiency and the fit by the formula was found and was shown in Paper VIII.

Paper IX actually embodies several individual devices to meet all the above mentioned objectives 1 to 4 by introducing a modular system to be used in fly-ash studies. Paper IX show that in a one collection a series of samples for a parameter can be studied in the same conditions. It has been problematic in the lab-scale ESPs (Papers D, E) to collect sufficiently many samples in identical conditions. Especially, when phenomena that are dependent on the ash resistivity are under study, the small trace levels of impurities can have a great influence on the ash, also to the collected and thus to the current-voltage characteristics and, consequently, to the particle charging and/or further to the collection. The ESP system of Paper IX comprises several identical ESPs that can be fine tuned for the wall and/or wire characteristics in the ash related studies. Depending on the flow, ESPs can be operated in turbulent, laminar or near laminar flow conditions which can be achieved by the embodied two-stage design. As Paper IX indicates, the system therein can be used also in environmental monitoring as a sampler that is provided with an electrostatic pre-filtration unit and with a facility to authentication of the samples.

III COAL FIRED POWER PLANTS, FLY ASH AND ELECTROSTATIC PRECIPITATORS

III.1 OVERVIEW ON THE COAL FIRED POWER PLANTS

The present and the other sub-paragraphs of paragraph III cite Paper A (Licentiate thesis of Sampo Ylätalo, 1996) for summarizing in a comparative manner the emission studies of Papers I-III in general at the coal fired power plants A, B and C, which are illustrated structurally schematically in Figure 1. The main features of the processes are shown in Table 1. The main focus for one aspect of this work for the Papers I-III was on the electrostatic precipitator (ESP) performance and especially in sub micron fly ash particle penetration.

The pulverised systems at each power plant had a different design and geometry for the coal conversion, which features are accounted for in general terms in Table 1. The boiler loads were based on the input coal heat values. The nominal boiler load for power plant A was 315 MW and the power plants B and C, respectively, 630 MW and 550 MW. For further details of the flue gas compositions for the power plants A and C, reference is made to Papers II and III, as well as to Paper A. The process details were not recorded at power plant B and therefore they are not presented here, neither in Paper II nor in Paper A, but the process observed at the site was as stable as the other two processes.

In each power plant, bituminous-type coal was fired. Polish bituminous coal was fired at the power plant A. In the two larger power plants B and C South African Klein Kopije coal was fired, and at the power plant C additionally also Eldorado coal from Columbia. The coals were analysed. Table 2 shows from the coal analysis the heating values but also, the ash and the volatile content, the moisture, and the carbon content (cf. also for the Papers II and III). The heating values are approximately similar, so is the volatile content.

III.2 THE HOPPER ASHES

The ashes (Table 3), as hopper ashes, from the three coals (Table 2) were expected to be moderate resistivity ashes and thus considered easy to precipitate, except for the Klein Kopije coal that can form high resistivity ashes and thus has in some cases been considered as a problematic coal. The ash compositions are compared in Table 3. The ash content variations can be seen as follows: In power plant C the fired Klein Kopije had almost double ash content to Polish bituminous in Power plant A in 1991. Eldorado coal seems to be similar to Polish bituminous for except the moisture content.

The main constituents of the ashes in each coal are aluminium oxide and quartz. Klein Kopije has 50% more aluminium oxide than Polish bituminous or Eldorado coal ashes. CaO is present over 50% higher level in Klein Kopije ashes than in the other ashes, whereas iron concentration level is only 50% of that in Eldorado or Polish bituminous ashes.

Alkali's concentrations are lower in Klein Kopije than in the other ashes, except for lithium. The absence of the alkalies is mitigated by a slightly higher SO_3 content in the

Klein Kopije than in the others. In power plant B fired Klein Kopije had almost decade higher resistivity than the very same coal fired in power plant C.

For power plant B, table 20 in Paper A indicates a downstream concentration trend of the main charge carrier species. Sodium and Lithium as well as sulphur are enriched into the last blocks whereas quartz as a good insulator, quartz seems to be enriching into the early blocks. This supports the findings on the species selective penetration prior to resistivity.

The same table 20 (in Paper A) suggests that ionic conduction related species (Sodium) are enriched to the downstream blocks whereas good insulators such as SiO_2 behave inversely. Species, which are related to electron conduction enhancement via multivalent-ion-presence-related process, such as Fe_2O_3 for instance, are present at the minimum concentration at the middle block, as well as Ca, which is related to the blocking effects of alkali ion migration in ashes, and Mg, being contributing to conduction in the ashes when no alkalies are present. Ti, as a transition metal, as comprised would contribute to multivalent ion enhanced conduction but is behaving as the alkalies. Phosphate pentoxide and sulphur trioxide are enriching to the late blocks as well as the alkali species, from which they both are known as surface conduction enhancing agents, while some free alkalies ions are present in trace levels in the ashes (Bickelhaupt, 1981) and sulphur trioxide is known as ash resistivity decreasing agent (Cook, 1975). The enrichment phenomena of alkalies into late blocks is not on a profound statistical basis, but consistent trends are existing, at least for these combustion conditions, and for both measurement days at the power plant B. Al_2O_3 is having maximum concentration at the middle block. Alumina is a compound of ternary porcelain, such as corderite, being a high resistivity insulator it pure and undoped.

III.3 THE ESPS

The ESPs were different in the power plants in the size and the capacity (Table 4) but only in a minor way. All units were operated with a specific collection area (SCA) between 83-97, The indicated lower SCAs in Table 4 were occurring during switching off of some of the fields. The ESPs were operated so that the one at the power plant A was operated in a DC-mode and the ones at the power plants B and C were in a pulsed mode. Pulsed mode is advantageous within high resistivity ashes. All units were single stage sectionalized electrostatic precipitators, whose spacings were 0.3 m and 0.4 m. The ESPs as well as their further details are shown in paper A. The experiments made by the ESPs in real scale as described in Papers I-III are shown in Table 5. At power plant A, load variation tests were made to study sub-micron penetration dependences on the loading. A coal sieve test was made at power plant A. Block switch-off tests were performed with the last block at power plant A and also with the second last block at power plant C. Charging ratio tests were made at power plants B and C.

III.4 THE EMISSIONS IN SUB-MICRON

Paper II contains detailed information on the emissions in the results shown therein. Table 6 summarizes sub-micron fly ash emissions. The results are presented separately for each power plant. In this work, having the link to the second aspect of this work on authenticity, only sub-micron size distributions are considered. All data is not shown, only the most relevant and the most typical portion of it.

Figures 2, 4 and 6 show the emission from the respective boilers at power plants A, B and C, as measured at the inlet of the corresponding ESP. The fly-ash size distributions in the figures in sub-micron are shown for the shown experiments indicated in Table 5 with the distribution curve characteristics shown in Table 6. The Figures 3, 5 and 7 show the corresponding emission at outlets of the ESPs, respectively. The corresponding power, special condition, coal and/or the measurement date are indicated.

III.4.1 *POWER PLANT A*

The number size distributions in all conditions appeared bimodal. The sub micron distributions at the ESP inlet and outlet for the small particle modes were at about 0.05 µm and 0.08 µm, respectively. The coarse particle mode occurred at 0.3 - 0.4 µm in both measurement locations, at the inlet and outlet. The peaked particle sizes as well as maximum concentrations and peak broadness seem to depend on the boiler load at the inlet and at the outlet of the ESP as the higher boiler load, the higher emission applying also to the penetrations.

Comparing the size distributions at the ESP inlet (Figure 2) among 234 MW-operating conditions does not indicate a great mutual difference. The size distributions curve-shapes are similar within all the conditions. The number size distribution in the largest boiler load-case (263 MW) has exceptionally broad sub-micron peaks which rises approximately to the same maximum concentration level as those in 234 MW operation conditions. Between these two boiler-loading conditions the difference in the size distributions in the range from 0.07 µm to 0.15 µm is significant. The fine particle sub micron peak is broader during operation condition 263 MW than during 234 MW, applying to the coarse peak, too. The coarse particle mode during the 263 MW load peaks into larger size than that in the rest of the size distributions. The broader shapes are suspected to originate from the extra burners being on duty for loads larger than 260 MW as related to the feed rate effect observed later within drop tube experiments. The peak below 0.1 µm are thinner at the outlet than those measured at the ESP inlet, whereas the coarse particle peaks show uniform maximum concentrations and widths, in which the operating condition 263 MW makes an exception. It can be seen from the size distributions that during higher power loading produced ashes the coarse peak size correlates with the load at inlet but does not do so at the outlet.

When comparing the distributions of conditions "234 MW" and "234 MW 3rd block off", the fine particle modes differ less than the coarse particle modes (logarithmic scale effect excluded), indicating that the last field may selectively collect more particles from the fine peak than from the coarse peak. The milling condition change was dramatically seen after the ESP: The operating conditions of "234 MW" and "234 MW with Coarse Coal Particles" are essentially similar as well as the size distribution curve shapes, but both sub micron peaks were sifted to the finer particle sizes during the coarse coal particle conditions, the sift being from 50 nm to 40 nm.

III.4.2 *POWER PLANT B*

Similarly to power plant A, size distribution features were found from the power plant B data as well. The number size distributions appeared skewed to the coarse side, which includes a possibility of a second mode that however may be not so clear in statistical terms. Table 6 presents also these hypothetical peak’s characteristics. Particles smaller than 0.1 µm were observed to agglomerate as shown in the SEM photographs in Paper G, and the 0.3 µm or larger particles appeared as spheres, which is referring to different formation mechanism and different particle density, resistivity and properties in charging or adhesion chemistry.

The number size distributions appear within a range of 20% also indicating typically a stable combustion process for such a large boiler. Similar observation could be seen earlier at the power plant A measurements and during all combustion conditions as well as later at power plant C. Due to the agglomerated structure of the particles in ultra-fine regime (below 0.1 µm) the density would be different than that of spherical particles, which could influence on the penetration data evaluation.

At the ESP outlet the number size distributions are similar to the inlet size distributions but peak into a coarser location, which is supported by the same observation at power plant A and from the data collected therein. The ESP was operated in pulsed mode with different charging ratios applied during two measurement days. The influence of different operation of the ESP on the size distributions is visible, but it can be seen more clearly in the penetration curves. This observation was supported by power plant C data, too.

III.4.3 *POWER PLANT C*

At power plant C two different coals were fired during the measurements into the furnace. Klein Kopije coal was the same as in Power plant B, but the other coal that was used was Colombian Eldorado. The boiler was different than the one used in power plant B. The ESPs (Table 4) were different, semi pulse operation of the ESP was common to both power plants B and in C for both coals.

At the ESP inlet the number size distributions were similar to those determined within Klein Kopije coal in power plant B. However, the inlet and the outlet distribution curve shapes are less skewed in Power plant C than in Power plant B, indicating just one sub-micron mode, and different particle formation conditions and precursors. Within both coals the size distributions peaked into a coarser particle size in Power plant C than in Power plant B. The observation was applying to both the ESP inlet and the ESP outlet. The fly-ash particle number concentration is significantly smaller in Power plant C than in Power plant B. The two coals' fly-ashes are different also in ultra fine range, D_p<0.05 µm, where Klein Kopije coal ashes additionally peaked at 0.03 µm but those of Eldorado coal did not do so in similar conditions. The mutual differences between the size distributions during each measurement day were minor thus indicating a stable combustion process, typical for large boiler as in Power plant A and Power plant B.

At ESP outlet the high peak in 8.5.-94 in the ultra-fine particle size was traced to be originating to charging ratio experiment. Normally the ESP was operated in pulsed mode with certain ESP-specific charging ratios. In this experiment the charging ratios were set to 1/1/1/1/1 for all the five fields. However, the 30 nm peak was not seen within Eldorado coal. Paper II deals with the mechanism and explains that as a formation of ammonium bi-sulphate referring to the figure 8 in Paper II.

The charging ratios as well as SCA were varied when one or two blocks were switched off before and during the sampling. By following one DMA's mobility channel and corresponding singlet size of 0.08 µm at the outlet peak maximum, the ESP stability were monitored during the second last block switch off. Rapping could not be observed contribute to that.

III.5 PENETRATIONS

III.5.1 *POWER PLANT A*

All the power plant A measurements were performed with the ESP in a DC-current mode (Table 4). The sub micron fly ash penetration in operating conditions 208, 234 and 263 MW in Figure 8 have similarly bi-modally shaped curves occurring at the same particle sizes at the penetration window. Comparing the electrical conditions of the ESP in Table 5 for 234 MW cases indicates that the bimodal penetration was occurring during such an electrical set up where the 3rd block was turned off (in 1992), but also during the low current level in a block (208 MW, 1991). So the bi-modality could be related to the electrical operation conditions or to a reduced SCA. When inspecting the peaks of the distributions at the penetration window, the coarser particle penetration peaks are uniform in cases ”208 MW, 1991” and "234 MW 3rd block off", whereas the finer peaks in the same set-ups are clearly sifted to coarser sizes, especially on the left side of that peak. Milling of the coal has influence on the fly ash penetration level. A sudden switch from one coal grind to another changes the combustion and therefore the particles' properties as individuals or as aerosol in their firing. When burning conditions were essentially the same as in installations “234 MW” and “235 MW, COARSER COAL”, the milling sieve rotation speed was decreased by about 23%, which apparently increased the sub micron penetration level. Due to very low mass concentration levels, the penetration curves differ from those determined by DMA below 0.5 µm and are therefore not presented in the size range below 0.5 µm (Ylätalo, 1996).

III.5.2 *POWER PLANT B*

The sub micron penetrations were measured during semi-pulse operation within different charging ratios in the two measurement days (Figure 9). However, all the penetration curves peaked into the size range 0.1-1.0 µm, like in power plant A, but with higher penetration maximum and narrower peak-curve morphology. The peaks were not as clearly bimodal as in Power plant A. The intermittent energization contributes possibly on the penetration curve shape and level. The penetrations were measured with reduced average and with increased charging ratios in the range of 0.3-1.0. The observation is supported by power plant C data. The penetration curves determined with BLPI were calculated with different densities for super micron and ultra-fine particles due to the different particle morphology, judged by the SEM photographs, and to support the DMA-data based results.

III.5.3 *POWER PLANT C*

The lowest penetration levels in sub micron were measured with non-disturbed ESP during at the very first days. The penetration with the Klein Kopije ashes was similar to that in Power plant B, also the penetration window shape and height were similar (Figures 9 and 10). However the penetration was on a lower level during Eldorado coal combustion, being only half of the maximum seen with Klein Kopije coal ashes. Playing with the charging ratio had dramatic effect on the penetration in the ultra-fine region of 0.02-0.06 µm: the penetration exceeded over 100%, by several magnitudes, which could be explained with no terms of normal ESP operation, but by new particle formation via ion induced nucleation from the stack gases assisted by the large ion number. There was an ammonia slip of few ppms from the catalyst which caused additional particle formation (Dismukes, 1975). The phenomenon was observed only with Klein Kopije coal ashes. Switching off the last blocks increased the penetration only by 2-3 % units, which was only less than half of the effect in the power plant A experiments. The charging ratio was observed to influence on the penetration level in the particle size range of 0.2 µm - 0.9 µm at the ashes originating from Klein Kopije coal. During few measurements the charging ratio was changed in the middle of sampling, and this caused the penetration curve to explode in both ends of the sub micron range (D_p<40 nm and D_p>800 nm).

The penetration in the coarse sub micron range was studied by measuring with DMA and with long averaging time in the few last channels in sizes 0.5-1.0 µm. These results indicated the penetration to be on same level as in normal sizing methods within Eldorado coal, but on smaller level within the Klein Kopije coal.

III.6 SUMMARIZING THE PENETRATIONS IN SUB-MICRON

As citing to the literature survey in Paper A on the previous works, the penetration of fly ash through the ESP has been known to be from few per cents up to tens of per cents in the size range of 0.1 µm to 5 µm, called penetration window. By using a superior apparatus and dilution methods the penetration window existence was quantitatively confirmed in this work. In the past, the connection between the firing process and the ESP performance was not combined thoroughly, although separate substances like SO_2 and water have for long been known to be able to affect on to the ESP collection efficiency. Combustion conditions are influencing significantly on the ash properties with regard to the ash chemistry and size distribution. The sulphur and the alkalies concentration of the ashes are important parameters having direct influence on the ash resistivity. Chemical bulk composition of ceramic or glass type particulates is determined in the combustion from the aluminosilicate impurity inclusions in the coal and their capabilities to react with the present chemicals and to form the ceramic particles in reductive conditions. The alkalies impurities dope these ceramic particles when gaseous species are condensing and forming particles and later in the cooler environment deposit on the particle surfaces layers. The collection efficiency of an ESP has been observed to be material dependent prior to bulk resistivity of a particulate material (Lind, 1995) in a narrow super-micron size range, which results are supported by the findings concerning electrostatic separation as an enrichment method of coal for example (Inculeti et al. 1975, 1981). The particle resistivity can be influenced in many ways by additive selection in pre-or post combustion agents (Raask 1985, Talmon and Tidy 1975, Dismukes 1972, and Cook, 1975). In addition to electrostatic forces also adhesion and cohesion chemistry of individual particles influence on the collectability of a dust and re-entrainment probability (Pontius, 1991). The charging of the particles is clearly a key factor, and relates to the bulk properties of the ash cake on a plate and to the individual particle's properties via corona discharge and ion formation, which are also dependent on the impurity species in the stack (Marlow, 1978, Okada and Sakata, 1994). Small particles, when being largely suspended in the stack gas, form a space charge in the inter-electrode volume when the small particles are charged. The particles move slowly when compared to the ions. This particulate space charge can reduce the corona current significantly and therefore influence on the ion formation,and it can also reduce charging and the collection efficiency. Ion wind as such is not a problem for large scale ESPs due to their low current density operation mode. However, pulsing in extreme conditions might enhance vortical movement in an ESP, contributing to re-entrainment of the already collected particles. Within very high resistivity ashes, these phenomena may be important, as well as in back corona conditions when the current density increases significantly and thus contributes to ion wind formation.

Turbulence intensity in the gas flow is enhanced by the corona discharge. However, the turbulence minimisation would yield the best collection efficiency, near the laminar conditions (Leonard et al, 1983). In conservative Deutschian models the particle mixing is a widely used assumption, but Williams and Jackson (1962) have pointed out that the eddies in the flow cannot provide the complete mixing for valid Deutsch model utilisation. There are many models, which take into consideration imperfections in the gas distribution, sneakage, rapping, secondary flow, etc. but the penetration of the aerosol particles in the sub-micron size range as a function of particle size is different than what has been measured.

The very first measurements were performed in a campaign by McCain et al. (1976), conducted to detect the sub-micron penetration through the ESP. The results were different than in theory. The conflict of the theory and the experiments was observed later in the studies by Ensor, Carr, McElroy et al. (1979-1982) in several power plants, as well as in the latest measurements described in this work. Figure 5 of the Paper III presents penetrations for power plant A as determined in current measurements compared with earlier measurements. The comparison of these results has agreement for the penetration window.

To analyse and explain why or what components are involved for such increase in migration velocity (and consequently decrease in penetration) in ultra-fine size range throughout the power plants involved for the experiments. The over-simplified Deutsch model does not explicitly account for any mechanisms responsible for the enhanced collection efficiency in the ultra-fine size range. Therefore, a calculation was made based on the ESP as a wind tunnel with the appropriate dimensions without electricity. Thermophoresis and deposition were studied and they would yield no such effect. In the experiments, the combustion conditions were definitely not rising the migration velocity due to stableness of the process. However, the turbulence enhancement by electric field would result a small penetration increase trend, but only on a level of a magnitude and a half too low.

This work confirms the previously measured penetration window and the varying shapes of the curves. Experiments and theories agree satisfactorily as indicated in the sensitivity analysis of Paper A, but theories can not describe accurately the aerosol penetration through an ESP as measured in real industrial scale. Within well-defined aerosols species and in controlled laboratory conditions theories and experiments have synergy for fly ash penetration (c.f. Zhibin and Guoquan 1994, versus Riehle and Löffler 1990), but not in industrial coal combustion fly ash aerosol. Problems of describing quantities are complicated and the affecting phenomena are dependent on many details in a feedback type loop. A realistic model should describe the dynamic situation where particle charging and collection are simultaneously occurring in an environment where interactions with the neighbouring particles and ions determine the faith of individual particles to be or not to be collected. Chemical composition of ash particles as well as that of the gas influence on the corona current utilised for the particle charging.

The experiments seem to be more advantageous to be performed in a smaller scale, as in a laboratory, to better be able to control the conditions than in a real scale power plant. This is the development aimed at with the techniques used in Papers E and D. Practical experimenting with a laboratory scaled device can be difficult for a sufficient number of repeatable experiments, which can be made for instance by an embodiment of the device of Paper D. For a better regulation of the conditions than those available in a real scale power plant, a laboratory scaled ESP with a drop-tube furnace was designed and built. The Lab-scaled ESP has been described in SIHTI report (1996) (Paper E), and the relating results on the operation and the measurements of the lab-scaled ESP were shown in a conference held in Toronto (1995) (Paper D). The Papers D and E show a solution to avoid reproducibility problems encountered in real-scale, but such a device is also dependent on the ash chemistry and the so-related temperature problems. Consequently, Paper IX shows a design of an ESP-sampler that promotes sample diversity in a one set of measurement conditions. Paper IX shows an ESP-sampler for lab-use, but embodies variations suitable for high-volume sampling, too. Paper IX was first addressed nationally to patent examination, and later also to an international PCT-patent examination, including Finland. The patent application has been submitted for the national examination of the priority application, and thus the application is pending.

IV ON THE ENVIRONMENTAL SAMPLING

In Finland STUK- Finnish Radiation Safety Authority- monitors continuously airborne radioactivity (Pöllänen et al. 1999). Monitoring is based i.e. on filtration techniques of the outdoor air. The monitoring network comprises in addition to dose rate measurement stations also several manually operated stations, but also an automated station, CINDERELLA.STUK in which there is a facility for an on-line nuclei analysis from the collected particles. In a nuclear disaster, such sampling stations are an important part of the radiation-monitoring network.

For a WAES-program, Wide-Area Environmental Sampling, the facilities for a sampling network were developed for the purpose of revealing undeclared nuclear activities, enrichment or reprocessing as based on automated samplers (Valmari et al., 2002). A similar kind of a study on the feasibility of aerosol sampling in rough field conditions was made as a field test in Kazakhstan at a former nuclear weapons test site for developing methods and test equipments for use in special conditions (Tarvainen et al., 2001).

Pre-filtration was also studied for promoting the sampling time in desert conditions or in other loading conditions of heavy coarse particles. A pre-filtration unit IITA (Paper V) was designed for the purpose, to stop entrance of large mineral particles into the sampler and thus into the filter for clogging it.

International supervision by environmental sampling requires, however, reliable samples for their purposes and thus means to indicate and verify the authentic sample origin. Even any potential thread on forgery of delicate samples of the above type can create crises, but it can also motivate developing of systems to establish a link between the sample collected on a site and the observations of the sample constituents shown in the study report.

In case of doubt, for restoring the confidence such measures need to be taken that can be relied upon when the chain from the report to the sample is verified backwards. The last link of the chain, the sample itself, plays a very important role and thus its authenticity should be able to be secured. It is fatal for international relations, for instance, if an environmental sample were changed - even accidentally - with another one so indicating false nuclear manufacturing activities, although not performed in reality. Mistrust can be avoided by measures that are sufficiently, and arranged to be present in the samples. It is namely so that international crises may potentially be built up or ceased along with such samples and their authenticity. Similarly, also civil engineering would benefit of a sharp indication of the sample authenticity, detectable in a simple but reliable way.

The studies of the penetrating aerosols originating to a coal-fired power plant indicate that sub-micron particles can escape even the filtering and so contribute to the emission, and thus can travel long distances with the winds. Thus, certain particle sizes can be used also in the environmental monitoring for surveillance of the activities.

IV.1 HIGH-VOLUME SAMPLERS WITH A PRE-FILTRATION UNIT

It is advantageous to perform the monitoring with several redundant samplers, taking into account the variations in wind conditions. The samplers (Toivonen et al. 1998, Valmari et al. 2002) can be automated for the filter change and/or for acquiring the spectra of the radioactive substances. The suitable time between the packet collections of the collected filters for a further analysis depends on the filter cogging probability, but can be once a week or even a much longer period. The filters can advantageously be pre-analyzed on-line in a similar way as in the CINDERELLA.STUK, which is a fully automated high-volume sampler provided with an inbound detector on the site at the STUK, (Radiation and Nuclear Safety Authority). Thus the filters are analysed on-line before the fine-analysis in the laboratory for the substances is carried out.

The sampler type as such is not so critical. For practical reasons the automated sampling principles according to the CINDERELLA.STUK (Toivonen et al. 1998) seem to suit well and are applicable and thus they were used in the experiments of Paper IV. In field conditions, also other implementations can be used for redundancy, for instance such as those indicated in Figure 2 of Paper IV or as those by Tarvainen et al. 2001. Also electrostatic precipitator type sampler as shown in Paper IX can be used. The second sampler type can be similar to the first one, but it would be preferable it it were better equipped for desert conditions in order to avoid massive sand occurrence in the samples due to winds and storms (Pre-filtration, Paper V). For authenticating purposes, each sampler can be provided with An Aerosol Authentication Apparatus (AAAA) similar to the one of Paper IV.

IV.2 ON THE MEASURES OF AUTHENTICITY IN GENERAL

The measures of authenticity are featured in the following by a few common principles that should be obeyed for having the sample sufficiently reliably authenticated for the purpose that the sample was taken so that any arbitrary macroscopic location of the sample could indicate the authenticity. When these features are applied according to the defence-in-depth principle, falsifying of authenticated samples will be extremely difficult, if not entirely impossible.

1) Measures of authenticity can comprise physical and/or chemical features apparent to the size distribution of the aerosol particles themselves. The particle size and/or morphology, radio-chemical composition, optical properties or magnetism can be used.
2) The measures of authenticity can comprise original features of the aerosol per se and/or added features.
3) In either case, a single feature as such may be inadequate for the recognition of definite authenticity at a low risk level. Several features available in pre-defined mutual harmony lowers the risk level for making mistakes.
4) An important feature of the authentication of aerosol samples is the principle of equal or at least known adequate dispersion of the material, which is used as a measure of authenticity, i.e. in the sample for instance, and preferably so that the removal thereof afterwards will be impossible without leaving behind any traces of tampering.
5) The measure of authentication that is used should not interfere with the sample or the information that is under the study for the interest, i.e. an environmental sample on a filter, for instance. That aspect is a very important principle that should be acknowledged scientifically especially not only in military work but in patent

infringement cases as well.

6) In a preferred embodiment of the authentication method, the measures of authenticity can be roughly categorized into two main groups. The first category comprises common measures, which correspond to a public key or an ID in an IT-field. The second category comprises hidden measures, corresponding to the private key or the signature in the IT-field. Either categories can comprise several features or just a single one. At least in principle, it is advantageous that the features can be varied in accordance with a time aspect to hinder the tamperer's investigations.

IV.2.1 *ON NATURAL MEASURES OF AUTHENTICITY*

One aspect of the studies of this work continues from the previous work described in the Licentiate Thesis of Sampo Ylätalo (1996) Paper A, which comprises a very thorough study on electrostatic precipitator performance in coal combustion, as cited also in this work in chapter III. Therefore, the work by Ylätalo (1996) covers in its experimental part pretty much the aspects shown in Papers I-III.

The work in Paper A reviews ash formation related aspects from a point of view, which is actually relating to particle removal. In that work coal, its combustion and fly ash, as well as the fly ash formation studies are reviewed for a long and comprehensive introduction into the behaviour, composition and the properties of the material that the electrostatic precipitators are collecting in coal fired power plants. Additionally, size distribution measurements are dealt with as performed in real coal fired power plants and the electrostatic precipitators in various conditions.

In addition to applying the number-size distribution determination to emission and ESP performance, one can realize from Papers I-III that the same measurement techniques actually provide means to measure any size distribution applicable in a more device-friendly environmental sampling. Thus, the technology is available for a long term monitoring with or without diluting (Papers I-III) and can be applied also for the verification of the sample authenticity. Indeed, for studies of aerosols in forests as well as in an urban areas, long-term monitoring studies have been made by collecting environmental data in several projects carried out in the University of Helsinki (Hussein et al. 2003, 2005).

Papers I, II and III show as experimental results and observations on the mere ESP performance and the aspects influencing thereupon. However, the utilization of size distribution data for authentication has previously not been dealt with at all. Therefore, Papers I, II and III are treated in the following as they are, but also as relating to the non-published authentication aspects of this work and as a continuation to the earlier work, and they are used to demonstrate the aerosol size distributions in different conditions and are thus applicable for the scenarios as a measure of authenticity.

IV.2.1.1 SIZE DISTRIBUTION AND MOBILITY ANALYSIS

In the above mentioned works by Ylätalo et al. Differential Mobility Analyzers (DMAs) provided with a dilution system have been used, for taking the sample out of the stack, as explained in the Papers I-III and Paper A. Taking a sample out of the stack is technically speaking a more challenging task than bare monitoring in the ambient air at the fence of a factory, although the techniques are straight forwardly applicable.

In Papers I-III, a differential mobility analyzers (3071) of TSI-type were used in combination with condensation nucleation counters. Data inversion algorithm influence on real data was studied and two algorithms were compared (Paper B) yielding essentially the same size distributions. In Paper I, the results were based on the utilization of MICRON-algorithm (Wolfgenbarger and Seinfeld, 1988) modification in the data reduction but in Papers II and III with the TSI's own simpler algorithm. However, in the studies that were conducted in Paper VI, Vienna-type DMAs were used as classifiers to produce monodisperse aerosols to be collected by the EDP-device of Paper VI. The operation of a DMA has been described in various Papers, from which for instance Knutson and Whitby 1, Knutson and Whitby 2 (1975 both) and Reischl (1991) are among the commonly cited ones.

For any measurement with a DMA that deals with polydisperse aerosol, the pre-impactor has a key function to cut off larger particles from entering the DMA. Double or even higher state charged particles (Paper A, Figure 34) can influence on the number of counts in a mobility channel. An unknown number of multiple charges can destroy the mobility measurement. Thus, it is safe to restrict the entrance of over-sized particles by a pre-impactor and to deal with charge distribution that allows reliable estimation of the doublet and triplet etc. shares in a sub-micron in a singlet channel. The classified particles from a mobility channel can be counted by a particle counter (TSI 3020, or

3025 for instance), or collected on a substrate for size distribution determination by programmatic thresholding, as demonstrated with the EDP-device (Papers VI, VII and VIII). For the DMA operation as well as for the authentication purposes, a pre-impactor plays an important role also in preventing the large particles as considered large differently in each case with the diameter of for instance over 0.5 µm and 12 µm respectively, from entering into the analysing/sampling device.

In case the coal is known and the conversion process in control, the size distributions of the penetrating aerosols are suitable for use as fingerprints of the power plant, or any combustion originated particle source that produces a constant sub-micron mode. This is so especially in cases demonstrated effectively in Paper I (Figures 1 and 2 therein) with the two-mode distribution at the penetration window range at the inlet and outlet, but also between the uni-modal size distributions of Paper II, because the main number size peak in sub-micron seems to vary from boiler to boiler (Ylätalo, 1996), and also from coal to coal in a boiler, but according to the boiler load as well. So, when the environmental monitoring is made near a power plant, or inside a power plant, the sub-micron size distribution applies to one natural measure.

Nevertheless, the number size distribution formed in coal combustion can be a good measure as it is and thus not easy to falsify without leaving traces, it were impossible to determine from a fibrous filter. Therefore, samples on fibrous filters as such are not at all applicable to number-size distribution determination purposes by scanning electron microscope (SEM) (cf. Paper IV, Figures 4 and 5). However, for instance TEM-grid and/or pre-surface-tensioned silicon substrates are sufficiently smooth to collect particles for TEM or AFM-analysis (Atom Force Microscopy) for photographing and programmatic post counting and classification with a collector, as the one embodied in Paper VI.

IV.2.1.2 RADIONUCLEI AND COMPOSITION

Radioactive substance became known as a tag from the Chernobyl accident. The accident released a huge amount of radio nuclei into the atmosphere. The reactor borne, artificial radioactive matter was dispersed all over Europe by winds influenced by other meteorological conditions in the route of the winds. The fall-out dropped onto the ground and constituted a radioactive tag into the geological soil-layers, now existing as a part of the radioactivity of the nature, wanted or not. As a consequence, trees and vegetation at the heavy fall-out areas were tagged. Therefore, the accident did not only provide means for future forest studies but, it simultaneously provided means to indicate the origin of a certain timber in a limited scope.

Since the half life of the Caesium and the long lived isotopes, still present in the nature, the wood collected and used today may contain the radioactive time stamp for several hundred years to come in a detectable form for the archaeologists of the future so that they will be able to verify and indicate the authenticity of today's master pieces made of such timber.

In the field of nuclear technology, reactor burn-up is related to elemental composition of the fuel inside the reactor at the moment of an accident. Thus the core composition provides an effective tag to be used. It could be useful if fall-out substances originating to Chernobyl for instance were discriminated from the natural sources and/or other man-made sources. When the Chernobyl accident took place, there was at least a first amount of a first nuclide and at least a second amount of a second nuclide from the plurality of the nuclei present in the reactor of the Chernobyl, having the quantities of which their relative composition is known as a time series and thus dynamically scalable to any moment as long as the radio-nuclei are detectable in the samples. In another reactor, the core composition is not very likely the same and thus the nuclide ratio will be distinguishable if the substances are dispersed in an accident. The isotopes are present in the nature and the long-lived ones shine as a man made tag for a long time. Such a tag based on the nuclei ratio is so distinguishable for a long time. Work in that field of radioactivity studies has recently been carried out in Finland in STUK, the Radiation and Nuclear Safety Authority.

In a smaller scale than the Chernobyl, a similar kind of utilization of a radioactive tag, well known but made on purpose, was distributed into the nature in Studsvik. A relatively large amount of Caesium isotope was vaporized in a furnace and thus dispersed into the nature as a Studsvik signal. However, the amount of radionuclide was considered small and irrelevant for the health aspects, at least according to the knowledge available at the moment.

IV.2.2 *ON ADDED MEASURES OF AUTHENTICITY*

To sum it all up, any measures should follow Principle 5 so that they would, not to deform the desired information of the sample. Thus, in the sampling of airborne radioactivity inactive tags should be preferred as added measures. Radioactive tags are not excluded from utilisation as far as Principle 5 sustains. For provisions of a good tag, reference is made to paragraph IV and especially to point 1), to state that the added measures can be basically similar to the natural measures. The added measures advantageously comprise features that are unnatural to the collected sample. Some features can be made open for inspection, but at least a few of them should be hidden to secure the system. For environmental sampling in the scenarios of airborne radioactivity, an example of a complex tag is considered in the following.

IV.2.3 *A TAG AND ENVIRONMENTAL SAMPLING*

Securing the sample authenticity in sampling incidences against accidental and/or tampering made on purpose, it is assumed that an authentication tag is used. The tag comprises at least three added features (Table 1). For the first, unnatural particles comprising LATEX, but stained by a fluorescent agent as added into the precursors to yield the fluorescence of the particles. So, a quick distinction can be gained from the other spherical mineral particles, such as the cenospheres for example. Paper IV demonstrates how to distinguish stained LATEX particles on a glass-fibre filter.

The size selection of the tag particles is basically free, but those with the diameter of 14 µm (Paper IV) are suitable. If the pre-filtration unit of Paper V is used in the duty, even larger particles can be selected and so two valuable additional measures are gained, i.e. the lack of the large particles from the original sample in combination with the presence of the large tag particles in the filter. Optionally, or in addition, particles of 7 µm could be used (they were not used in Paper IV), and as a hidden feature, particles of 2 µm could be used in a similar way as in Paper IV. On the contrary to the tag in Paper IV, any one of the tag particles could have been doped with a radioactive label, or even provided with a Cs137 and/or Cs134 to comprise a certain ratio of activity or other nuclei with the same. The dopants in a tag could also be stable substances such as Yb and/or Tb and so provide a stable concentration ratio.

Collecting outdoors, especially in desert conditions (Tarvainen et al 2001), can lead to sand piling into the sample. Because of the expected heavy loads of sands carried into the samples made by filtrating ambient air, the sampling time can be reduced accordingly. Therefore, an inlet head in front of the sampler should stop the intake of massive loads of sand. Thus, it is advantageous to use a pre-filtration unit as the inlet head, such as the one shown in Paper V (IITA, Impactor Inlet-flow Throughput Array). If several systems are used (Valmari et al 2002, WAES), they could each be provided with their own authentication system (Paper IV) with a common or an individual tag.

Pre-filtration (IITA Paper V) reduces sand intake and thus lengthens sampling time so that even higher cumulative concentrations of fresh nuclei on the filter can be obtained, whereas the influences of naturally occurring minerals comprising the natural nuclei of the sands are reduced for the background activity.

Figure 3 in Paper V demonstrates the influence of a single stage of IITA on coarse sands with the D(50%) of 12 µm (equ. 3 Paper V). Cascaded stages can also be used, although they were not demonstrated in Paper V. Successive stages in series can mitigate bouncing effects in sever dust conditions, although the collection efficiency curve of an impactor stage may be considered quite sharp at the D(50%).

The S-shaped collection efficiency curve of a single IITA stage will guarantee that some sands will still reach the filter. Thus the radio nuclei on the filter should reflect the radionuclide composition of the sands at the sampling site. The natural isotopes and daughters of Th and U should indicate that the sample is fresh, when acquired at the site, and the decay series in comparison to the initial activity should indicate in a laboratory that the minerals originate to the correct sampling site.

If a tampering attempt was suspected with the local sands and/or if coal combustion originating ashes were arranged onto a false filter in order to tamper the sample taken at a sampling site, the missing tag applied onto the filter in the sampler with the AAAA (Automated Aerosol Authentication Apparatus, Paper IV) would reveal a tampering attempt immediately. Even if 14 um particles were arranged into a false sample, it would not be obvious at all for an outsider tamperer weather the particles were stained by a fluorescent agent or not. Nor would a potential tamperer take any notice on the smaller particles used as the hidden feature.

It is thus important that the method itself is secured and the hidden feature particles are properly marked so that they can be distinguished from any ordinary fly ash particles, for example. In Paper IV, the hidden feature was provided by the smaller particles. Even if their presence was detected by potential tamperers, they would not become aware of the isotopes potentially carried by the hidden feature particles or the fluorescence properties. The fluorescence may also be arranged by certain non-local minerals as earlier discussed, to be revealed in an XRF-analysis (X-ray Fluorescence) only. For instance, doping of the large particles additionally by Yb makes it possible to prove that it is the large particle where the Yb-signal in the filter is coming from and thus to exclude other potential Yb sources. It is also possible to vary the authentication scheme from day to day. For example, we could use particles with the same appearance, but Yb-doped particles on even days and Tb-doped particles on odd days. In such a case, two apparatus of AAAA might be needed. Several tags can be used to make the tampering as difficult as possible.

The order in which the particles are collected onto the filter can also be used also for authentication purposes. Figure 5 in Paper VIII demonstrates such a valuable piece of knowledge, where the large particles appear to be first on the TEM-grid (Transmission Electron Microscopy) and the smaller silver particles are partly on the surfaces of the large particles, which feature indicates to the inspectors of the sample that the one they are looking at is actually the original one.

V AN AUTHENTICATING SCENARIO

Relevance of the sample authenticity can be demonstrated by a hypothetical example. Let us assume a monitoring scheme in desert conditions (Tarvainen et al., 2001), but where radio nuclei are sampled from a radio-chemical lab or a plant or where they are searched via the emissions that occur in the power generation (Valmari et al., 2002) according to a WAES-scheme. The searched radio nuclei may be collected by high volume sampling systems arranged to collect particles and/or gases on filters that each constitute such a sample. The samples shall be analysed for the filter-borne nuclear material. As the samples in the hypothetical scenarios can carry very delicate information, the sample authenticity should be able to be secured as reliably as possible. The environmental sampling and thus the samples could be a tool involved with disarmament or other political international topics to demonstrate the binding of the sample radioactivity on the filter, nuclear technology policy of a nation in respect to an international organization. Thus, it is in the sampler's interest to prevent any tampering of the samples, made accidentally or on a purpose, by any party involved, irrespectively of the interest of the party's own to switch the filters to different kinds of filters or tamper them for any reasons to adjust or to falsify the results to the party's own purposes. For illustrative purposes, let us assume demonstrative scenarios, in which the authentication by aerosol is used in the spirit of Papers I-VIII to demonstrate the securing of the sample authenticity. However, we should keenly keep in mind that any resemblance to any political detail in non-physical context is just a co-incidence, if occurring at all.

In Scenario I, location of emission source of air borne radioactivity is searched by a plurality of environmental samplers, in a similar fashion as in the WAES (Valmari et al. 2002). The source of the radioactive emissions is originally unknown, but it could be a plant for chemical and/or technical processing of nuclear material.

In Scenario II, the source of the radio nuclei is known and under surveillance by environmental sampling practiced by an international organization, possibly with a continuous size distribution determination in sub-micron to detect emission changes originating to the thermal parameters. In Scenario II, the sub-micron size distribution can be used as a measure of authenticity. A stable coal conversion process can provide means to use the size distribution as a measurable quantity for on-line counting. Similar DMA apparatus as described in Papers I-III for on-site measurements in coal-fired power plants can be used for a long-term monitoring of on-line size distribution measurement. In Paper II two power plants were compared. The size distributions at the inlet but also at the outlet (Figures 3 and 5 therein, as well as Table 6) show that the two power plants in the sub-micron size can be distinguished and, similarly, could be used for the authentication of the samples in Scenario II.

In Scenario III, especially small nano-particles are searched and sampled in a different kind of scheme, in which the device of Paper VI has been used for sampling through a DMA from a high-volume sampler's sampling line. Sub-micron particles were used to demonstrate the authentication. The collected sample was arranged for an automatic number-size-post counting. The sample was authenticated by large sub-micron particles on the substrate distributed in advance. For a further demonstration, in Figure 4 of Paper VIII there is shown a small-particle sample. The TEM-grid used for the substrate presents a normal sample collected in non-critical or non-delicate conditions in a laboratory (Figure 1, Table 2 in Paper VIII). In the Figure 4 of Paper VIII, there is shown a smaller icon also for the Laue-pattern to confirm that the ammonium sulphate particles are mainly ammonium sulphate particles, which can be judged from the analysis of the parameters of the Figure 4 in Paper VIII compared to the yield of the appropriate theory.

However, weather we are convinced or not on that the particles are really of ammonium sulphate, we are not able to state, if they have really been collected on the correct substrate. If we assume, that a similar sample should have been taken at a monitoring target, we simply cannot judge from Figure 4 of the paper VIII if the sample was really taken from the monitoring target or if it is just a skillfull forgery sampled correctly to yield the ammonium sulphate, maybe even the composition supported by the Laue-pattern but made elsewhere or in a laboratory where the sample in Figure 4 of Paper VIII was collected in reality (Figure 1, Table 2, Paper VIII).

Contrary to Figure 4 in Paper VIII, the large particles of the size of about 100 nm in Figure 5 in Paper VIII confirm that the used substrate is of the correct type, which adds on a further measure of authenticity by revealing that the small silver particles of Figure 5 in Paper VIII are even on the larger particles, thus confirming that the larger particles were first on the substrate, as was assumed in the pre-collection authentication scheme of the set-up for particles in Figure 5 in Paper VIII. So, because of the large 100 nm particles as pre-dispersed on the substrate, the analysing person knows from Figure 5 in Paper VIII that the small silver particles collected on a TEM-substrate are the correct silver particles.

In each scenario, a pre-filtration unit can be used for limiting the sand intake into the filter structures (Paper V). All the samplers in the scenarios can be equipped with an authentication and/or remote monitoring system (Paper IV and Toivonen et al. 1997). Thus, the samplers cannot be entered without a notice. In these circumstances, the only possibility to replace a hot filter (a filter with a clear indication of the substances in the interest of search) with a cold one is to switch them outside the sampling site.

VI CONCLUSIONS AND DISCUSSION

Papers I-III indicate that the sub-micron fly ash size distribution depends on combustion conditions. The boiler operation seems to dominate the size distribution of the coal in the studied boilers in the range of the small particles at the inlet, at least in the studied extent. The size distribution of the sub-micron fly ash emission can have one or two peaks at the sub-micron size, reduced in height, but as shifted towards the coarse sizes due to the electrostatic precipitator performance. The aerosol penetration curve for the electrostatic precipitator can be two-modal at the size range from 0.1-3 µm for a normally operating electrostatic precipitator having a collection efficiency of nearly 100% in mass bases. The maximum penetration level on the number basis depends on the ESP operating parameters. According to Paper III, a properly operated ESP can let 4% of the small particles go through within the size range, but in slightly disturbed conditions even 20% of the particles can escape within the same size range. However, the size range seems to be essentially the same independently on the coal, the boiler or the device used for the emission control. This can be indicated by comparing the results of some previous studies to the findings of the work shown in Paper III (Figure 5 in Paper III). Consequently, for the operators practicing environmental monitoring, these results suggest to concentrate on the very same range, where the particle emission on the number basis is the largest for traces and thus the potential to find substances of interest to be monitored is at its highest.

The emissions indicate that the size distribution of the sub-micron fly ash can be used as a measure of authenticity provided with an indication of stable operation. Environmental sampling requires reliable marking of the samples in a tamper-proof way and is a challenging task provided that the tag can be formed and dispersed on the sample sufficiently evenly and in a manner that does not disturb the analysis. As a disadvantage, a complicated tag can increase the amount of work for stating individual measures and the consequential authenticity. For this purpose, natural measures as well as artificial measures as added measures can be used. The method can be secured by itself (Paper IV) by using a visible measure or several such measures, but in addition, also invisible or nearly invisible measures the presence of which can be stated only in certain conditions. Alternation of the tag in use and out of use for marking as well as keeping the potential tamperer unaware of in what parts of the measure features are used will make it impossible for the tamperer to succeed in his attempts.

It is also possible to bind several sampling methods into one sample family by using certain codes to indicate the authenticity features of the family members only via one member. Therefore, in order to increase the diversity to mark and to indicate the unity of a sample family, it is possible to use a rare-earth element or an equivalent in the tag particles or in the filter material, or several of them in a certain composition ratio in order to indicate that several other sampling systems are used in parallel with the filtration unit. For instance, if in such a concept Yb is used for instance in a parallel DMA sample authentication, the filter fibres may have Yb or certain tag particles may have Yb as doped or mixed.

If in such a concept one impactor or several impactors are used as a third sampling device in a further parallel device to sample in parallel with the filtration unit, this can be indicated for instance by Tb in certain sized tag particles on the filter, or in the filter fibres. Even the tag particle size can be indicated and the place where to search the tag element. If for instance the Yb is in 14 µm particles on the fibrous filter, the operator or inspector could search the 14 µm or nm particles. The tag could also be in a cascade impactor stage having the cutting size of 14 µm depending, however, on how the particular coding was made. Therefore, it is possible to provide a net of features to bind the authenticity of the DMA samples, impactor samples and fibrous filters together and confirm in this way that they belong to the same series. If a rare element present in the fibres is used as a measure of authenticity, it may preferably be marked already in the manufacturing process. Nevertheless, woven filter fibres may be impregnated to some extent afterwards to comprise the tag element in the fibre. In a similar way, also silicon substrate for AFM analysis can be authenticated.

It is submitted indeed that the method serves in several kinds of applications as well. For instance, ink to be used in note printing or other monetary valued papers can be marked within a similar kind of a tag. The method as such is not limited to fluids or fibrous filters. Even the material, for instance any material used in the devices of Papers IV-VIII, could be marked by certain particles or grains arranged into structures of a certain size or distribution indicating the original manufacturer.

So, for instance, even the impactor of Paper V could be indicated to be authentic by inspection from a cross-section material and the particles therein could be detectable by an ordinary SEM (Scanning Electron Microscope), whereas an illegal copy might not have those particles with the same features, so facilitating the manufacturer to start a patent infringement process, to take a civil example. Or, to take a military example, an international organization using such a unique piece could easily certify that the instrument is really the same that they originally assigned into the duty instead of a tampered version.

The author believes that it is possible to make economical applications for the indicated methods. For instance, marking of fibrous filters can be achieved by a suitable selection of waste material, such as optical fibres for instance, to be manufactured for a commercial value.

So, Papers I-III as well as Papers A and B show studies on emission through electrostatic precipitator in real-scale in three industrial-size electrostatic precipitators. When knowing the observed size-range of the small-particles penetrating the ESP, the knowledge including that on the ash chemistry indicated in Paper A can be actually used for optimizing environmental sampling in respect of escaping particles formed most likely in a combustion or alike process. As Paper V indicates, in severe dust laden conditions, coarse particles and the matter contained in them can be removed by a pre-filtration unit in front of a high-volume sampler. Thus, the collection of one sample can be extended and, consequently, escaping particles can be collected on to the filter of the sampler in higher concentration than without such a pre-filtration unit. In addition, the findings on the samples would comprise fresh aerosols and substances comprised therein, because of the residence in air-borne state and the long distance transportation from the emission, indicating current activities of the monitored target, in a Wide Area Environmental Sampling scheme for example. Paper IV provides means for making the samples authentic against any reasonable tampering attempt. Such a system comprising a high volume sampler, a filtration based or an electrostatic one according to Paper IX, and the device of Paper IV can be used of course without the pre-filtration unit of Paper V, but the tag diversity to indicate the sample authenticity can be then more limited, although any appropriately sized coarse particles were not naturally occurring at all at the sampling site.

Papers I-III show that the properties of the size distributions of the particle emission can be used as measures of authenticity, provided that the particle origin can be identified to a stable process, and that the particles are countable, preferably in an automated process. Thus, a sampler to utilise a TEM-substrate instead of fibrous filter was designed in Paper VI, and its preliminary features were characterized in Paper VIII. A DMA arrange to determination of small particle distribution from a sampler stack for instance can be used for the size distribution monitoring or as a classifier to monitor certain sized particles, which can be collected onto a TEM-grid in the device of Paper VI, as shown in the test runs of Paper VIIII.

Paper IX embodies actually a group of new ESP-devices for use in environmental sampling, even with authentication and/or pre-filtration facilities, but also for use in ESP-related optimization studies as embodied in Paper IX. Thus embodies Paper IX even alone a system in which the sample can be authenticated, but in a more compact way than the system discussed previously.

VII ACKNOWLEDGEMENTS

The author would like to express this deepest gratitude to all the persons and institutions that have been supporting this work. The author would like to acknowledge the University of Helsinki and the department and faculty of Natural Sciences for their valuable contribution in facilitating this work and making it possible. Dean Heikki Saarinen is thanked. Head of the Department of Physical Sciences Professor Juhani Keinonen is thanked. The author thanks Professor Erik Swietlicki from the Lund University and Professor Kaarle Hämeri from the Helsinki University for the pre-examination of this work. This work has been made in several institutes and departments the open minded support from the personnel, administration and managements as well as the heads of each laboratory or department of which is acknowledged and considered valuable, likewise the Department of Physical Sciences in Helsinki University, Aerosol Technology Group in VTT, STUK and Berggren's patent agency. The Director General Erkki Leppävuori from VTT is thanked. Anneli Hase is thanked, but also many persons who wish to remain unnamed are greatly thanked for their contribution to the present work. The Director General Jukka Laaksonen and Research Director Sisko Salomaa in STUK are acknowledged as well as Lab Chief Harri Toivonen in STUK. Leif Norden, Irina Tallberg as well as Juhani Kupiainen but also Markus Levlin from Berggren's are acknowledged and thanked for their support. Also the SIHTI project and the management are greatly acknowledged for their support in the compilation of Papers I-III. The author is greatly thankful for the priceless contribution of the co-authors for their valuable opinions and discussions in progressing the papers. Special thanks are addressed to Dr. Harri Toivonen from STUK for especially witted far-sight contribution in progressing the work. The author also thanks the co-authors, especially Dr. Martin Mohr, Dr. Jukka Hautanen and Dr. Tuomas Valmari (STUK). Senya Oy and there especially Manager Jukka Lehtinen are thanked for the machinery and device-related valuable support and practical guidance. Also Asko Turunen from STUK is thanked for solving machinery related problems. The late director Mr. Tillander from Amet Oy is acknowledged together with the present Amet personnel. Dr. Matti Tarvainen (STUK), Group Manager Rolf Rosenberg (VTT) and Chief Research Scientist Matti Lehtimäki (VTT) are thanked and acknowledged for their contribution in providing back-ground support. Further, the author thanks Professor Folke Stenman from the Department of Physical Sciences at the University of Helsinki. The author would like to thank the Foundation of Finnish Inventions and the workshop therein, respectively, for their monetary support and fine machinery for facilitating the prototype building. Special appreciations are addressed to Project Manager Erkki Väisänen and Timo Kilpeläinen for administration and building the prototype, respectively. The author further thanks the Department of Physical Sciences in the University of Helsinki. Special kind of thanks are personally addressed to Professor Markku Kulmala, Professor Kaarle Hämeri and Dr. Pasi Aalto, and especially to Docent Jyrki Mäkelä, who is now working in the Aerosol Physical Laboratory in the Tampere University of Technology. All the above persons are appreciated for their indispensable contribution on the laboratory facilities and valuable guidance therein. Soile Huikku and “Benny” Berndt Träskman are acknowledged for their kind and indispensable linguistic support in the English and Swedish languages, at Berggren's. And last but not least, the author thanks Mr. Jyrki Juhanoja from the University of Helsinki, Department of Electron Microscopy, for his valuable and indispensable support in microscopy studies for this work. The author thanks his friends for their support and his family for their patience and the buffering of the up and downs of the author with the work. The warmest and most sincere thanks are addressed to Pirjo, Jaakko, Johanna and Liisa for their loving care in the background without which this work would never have been possible.

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IX REVIEW OF THE PAPERS

Paper I describes shortly the first studies based on size distribution determination via electrical mobility made in industrial scale within high precision instruments, such as two cross-checked DMAs (Differential Mobility Analysers) used for sub-micron fly ash analysis and tuned up for size distribution measurements on an industrial power station site. The Paper was presented by the Author in European Aerosol Conference in 1992 in Oxford, showing measurement results on the size distributions at the ESP inlet and outlet, as utilised for aerosol penetration determination and as calculated pair-wise channel by channel of the DMA from the experimental inverted data. The size spectra yielded the resolution of 30 size-channels into the small particle diameter, ranging from 20 nm to 800 nm. The whole series of eight penetration curves were measured and shown as based on the electrical mobility derived size distributions. They comprised both types of distributions as well as penetration curves: those that were simultaneously measured and those that were determined within 1½ hours time difference determined, appearing consistently and supporting each other for the mutual penetration level and aerosol penetration curve morphologies between the conditions. MICRON algorithm was used as the data reduction mechanism. The Paper also describes size distribution measurement techniques that can be applied in the perspective of this work to any stack or sampling channel of a high volume sampler, either with the dilution system or without such, for small particle size distribution determination.

Paper II shows the results of a measurement campaign similar to the one described in Papers I and III, but in different coal and combustion conditions as well as using a different ESP energization method and a different flue gas chemistry. The Paper uses data from two power plants, which both power plants fired the same coal and one of the two was using also a different additional coal. The size distributions, as determined in Papers I and III, have different modal values for the peaks in the sub-micron and provide an indication of the sub-micron peaks to be utilised as a characteristic feature of the coal conversion process, contributing to the scope of this work.

Paper III summarizes the results of Paper I into an average penetration of one specific boiler loading, which data is checked by a repeated series of experiments in similar imitating conditions. The size distributions were measured with a special interest in the sub-micron range, analysed by the two-DMA systems being cross-checked again and as provided with improved operation in power-plant conditions. The aerosol penetration data is compared according to the boiler loading and testing while one field was shut down. The experiment causes similar morphology for the penetration curve as discovered in Paper I and indicates that the sub-micron peaks can vary according to the coal conversion process and thus a provision should be taken if the sub-micron peaks are used in accordance to the perspective of this work. Papers I-III demonstrate the mobility measurements for the size distribution but the stable size distributions also can be considered as an indication for size distributions to be used on natural measures of authenticity.

Paper IV describes an authentication method and device that can be used against tampering of samples, which samples are collected on a filter by an automated sampler for analysis of radioactive substances. A defence-in-depth principle is suggested for the authentication by aerosol to be applied and demonstrated for the sample protection. The device is totally new and has been tested in long-term performance tests. The performance of the tag dispersing into the filter structures is demonstrated to be observable remotely via Internet, for example.

Paper V describes a design of a new high-volume sampler pre-filtration unit as based on the removal of coarse particles by impaction from the sampling inflow. The pre-filtration head was designed for modifying the size distribution of the raw input sample at the coarse end of the distribution, so that the filtration of the coarse sands for instance could be removed from the inlet flow before the actual sampling of such fresh particles that carry airborne radioactivity onto the filter. The impaction phenomena was utilized, having the device operating in laminar flow conditions but in a high-volume flow rate facilitated by the parallel impactor stages each operating at laminar flow conditions. As a benefit of the removal of coarse particles, also sands or particles or alike could be removed, and thus the clogging of the sample filter could be avoided. Also the background activity originating to the natural activity and the related isotopes in the coarse particles or sands could be reduced for the analysis of the actual sample on the filter. An important aspect for the perspective of this work is the cut size of the sampled aerosol particles, so facilitating further means to use also the larger particles inside the sampling track after the pre-filtration unit.

Paper VI (and VII) describes a design of a totally new EDP sampler (Electro-Dynamic Precipitator) for small particles at a low sampling flow. The operation of the EDP-sampler is based on charged particles in a DC field perpendicular to an AC field, which together and in co-operation yield the particle sample on a sampling substrate. As a sampling substrate for instance a TEM-grid (Transmission Electron Microscope) can be used. One benefit of using TEM-grids is that they suit well for microscopy and direct counting of the particles by software means, which can be performed without a human counter. The EDP-device can be used with a DMA for the sampling of sub-micron particles, which was demonstrated in this work with the authenticated sample. The authentication was made by pre-dispersed large particles on the TEM-grid before the sampling.

Paper VIII describes preliminary tests and results made in laboratory conditions, for the purpose of facilitating a new way to collect particles, also for a post-count, for which the EDP-device was designed and built as embodied in Paper VI. The EDP-prototype was operated on one hand for determination of particle removal efficiency and on another hand for the collection efficiency on the substrate to be taken out for a post-count analysis in a microscope.

Paper IX introduces a modular system to be used in fly-ash studies so that in a one collection a series of samples for a parameter can be studied in the same conditions. It has been problematic in the lab-scale ESPs (Papers D, E) to collect sufficiently many samples in same conditions. Especially, when phenomena that are dependent on the ash resistivity are under study, the small trace levels of impurities can remarkably influence on the ash, also to the collected and thus to the current-voltage characteristics and consequently to the particle charging and/or further to the collection. The ESP system in the Paper IX comprises several identical ESPs that can be fine tuned for the wall and/or wire characteristics for the ash related studies. Depending on the flow, ESPs can be operated in turbulent, laminar or near laminar flow conditions, which can be achieved by the embodied two-stage design. As the Paper IX indicates, the system therein can be used in also for environmental monitoring, as a sampler, which is provided with an electrostatic pre-filtration unit, with a facility to authentication of the samples.

X ERRATA:

Paper I: The dimensions of the concentrations at inlet and outlet (before the ESP and after the ESP, respectively) as shown indicate the concentrations without dilution fraction, being constant throughout the sub-micron as assumed.

Paper IV: Table 2 makes a reference to Hurford in context of radioactive label at last line, which is a copy paste error from above line.

XI APPENDIX I

PAPER I

XII APPENDIX II

PAPER II

XIII APPENDIX III

PAPER III

XIV APPENDIX IV

PAPER IV

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XVII APPENDIX VII

TRANSLATION OF PAPER VI

XVIII APPENDIX VIII

PAPER VIII

XIX APPENDIX IX

PAPER IX

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TABLES AND FIGURES

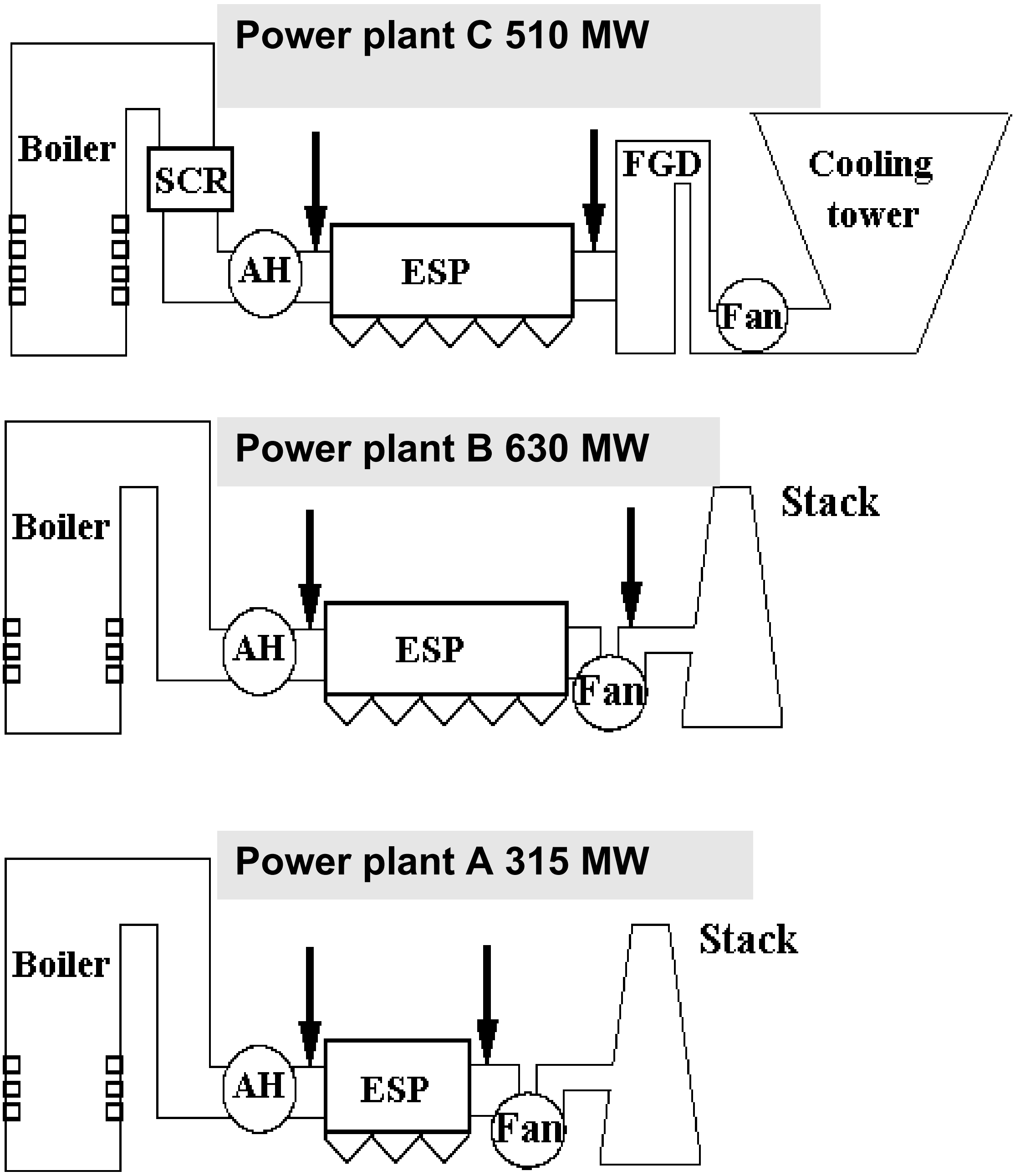

Figure 1. *Boilers, ESPs and stacks. Measurement points at the stack are indicated by the arrows (Paper II, Ylätalo, 1996).*

POWER PLANT A 315 MW

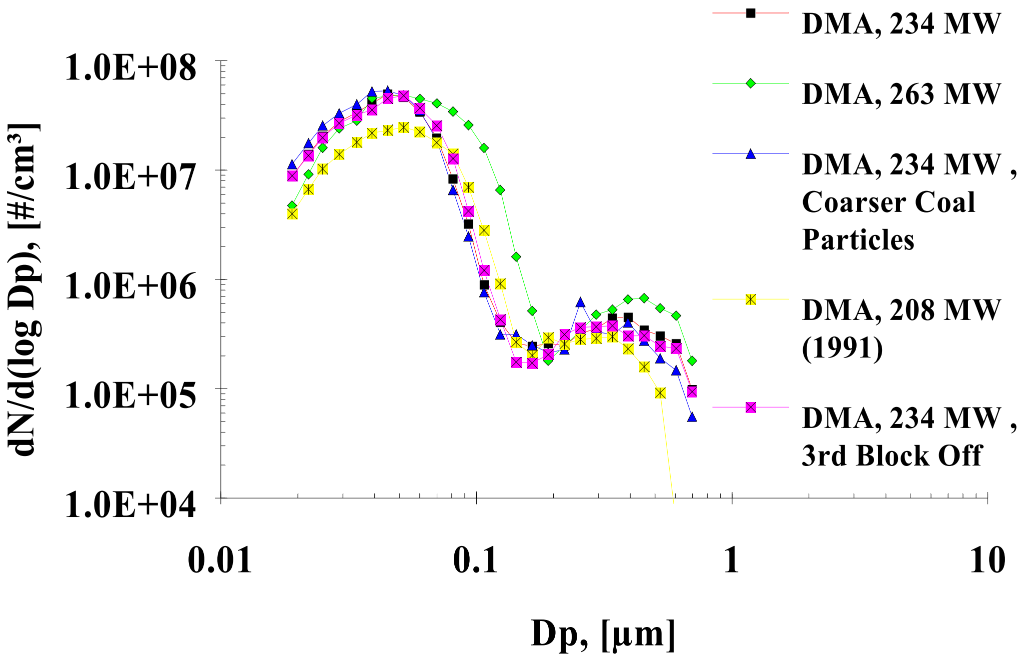

Figure 2. *Number size distributions at the Power plant A ESP inlet (Ylätalo, 1996).*

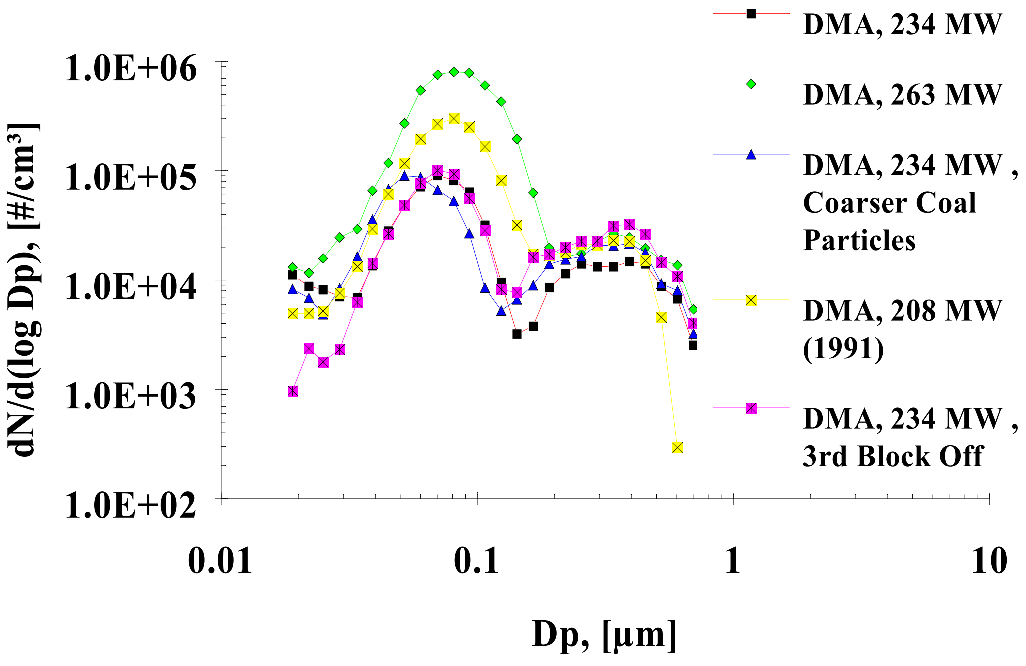

Figure 3. *Number size distributions at the Power plant A ESP outlet (Ylätalo, 1996).*

POWER PLANT B 630 MW

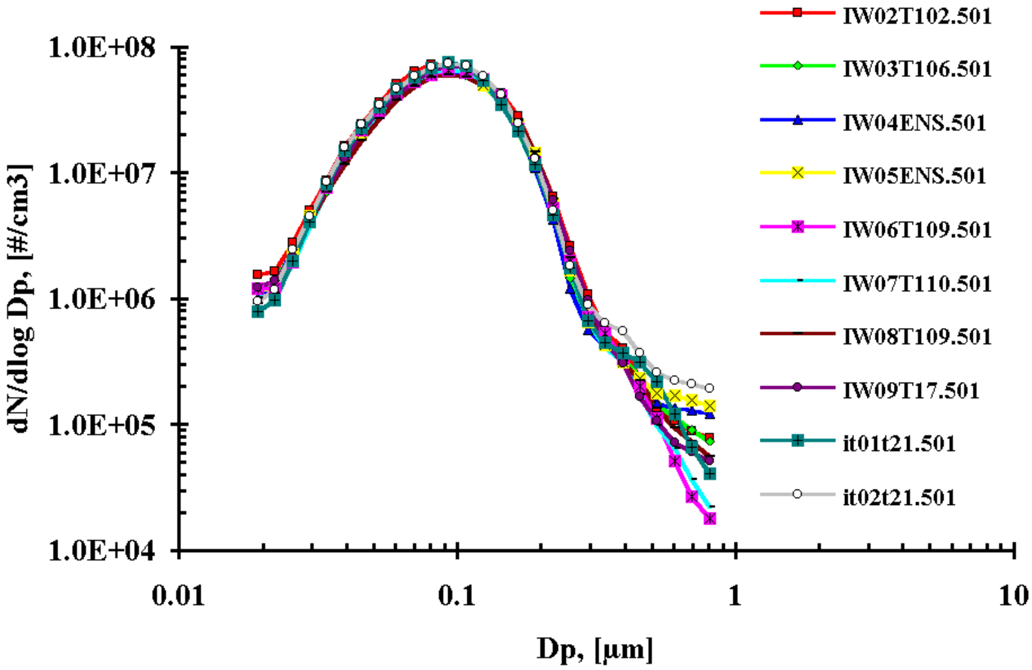

Figure 4. *Inlet number size distributions at Power plant B (Ylätalo, 1996)*.

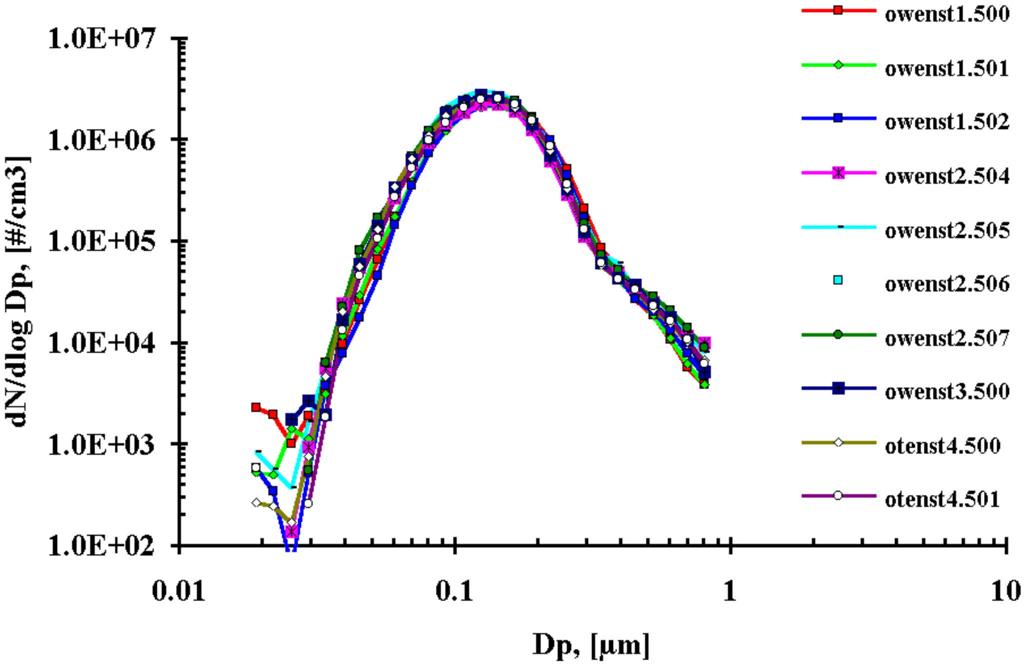

Figure 5. *Outlet number size distributions at Power plant B (Ylätalo, 1996).*

POWER PLANT C 550 MW

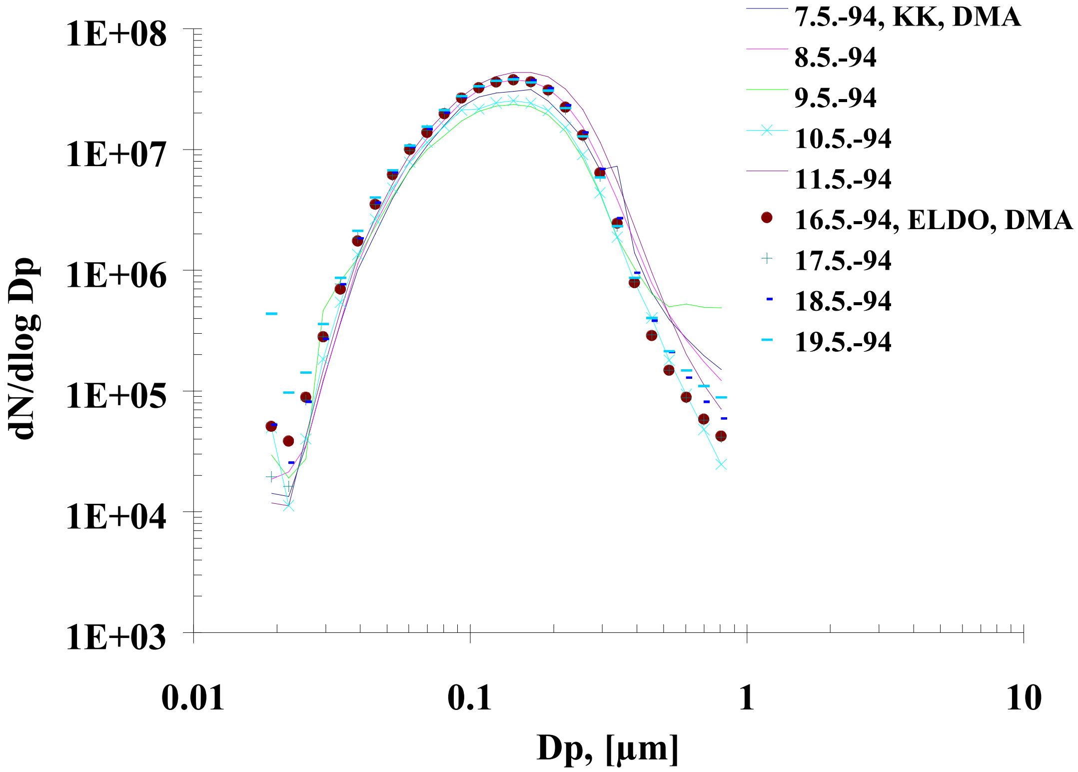

Figure 6. *Inlet number size distributions at Power plant C (Ylätalo, 1996).*

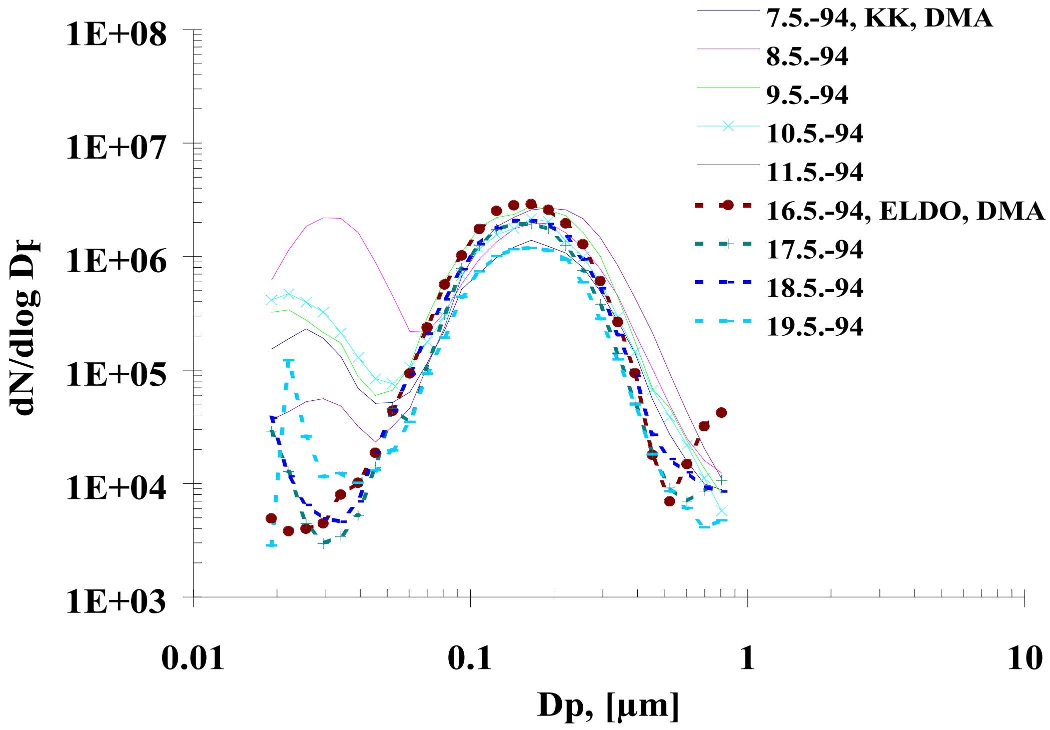

Figure 7. *Outlet number size distributions at Power plant C (Ylätalo, 1996).*

PENETRATION AT POWER PLANT A 315 MW

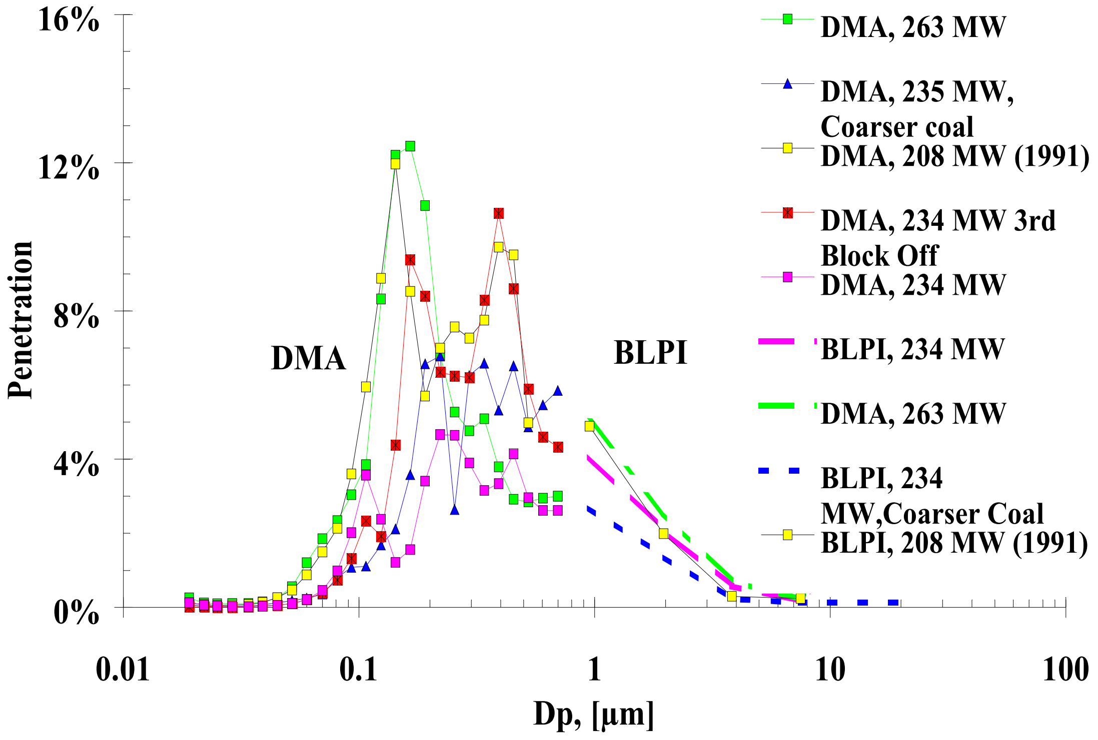

Figure 8. *Polish bituminous coal fly ash aerosol penetrations as determined with DMA and BLPI. Conditions as inTable 1 (Ylätalo, 1996).*

PENETRATION AT POWER PLANT B 630 MW

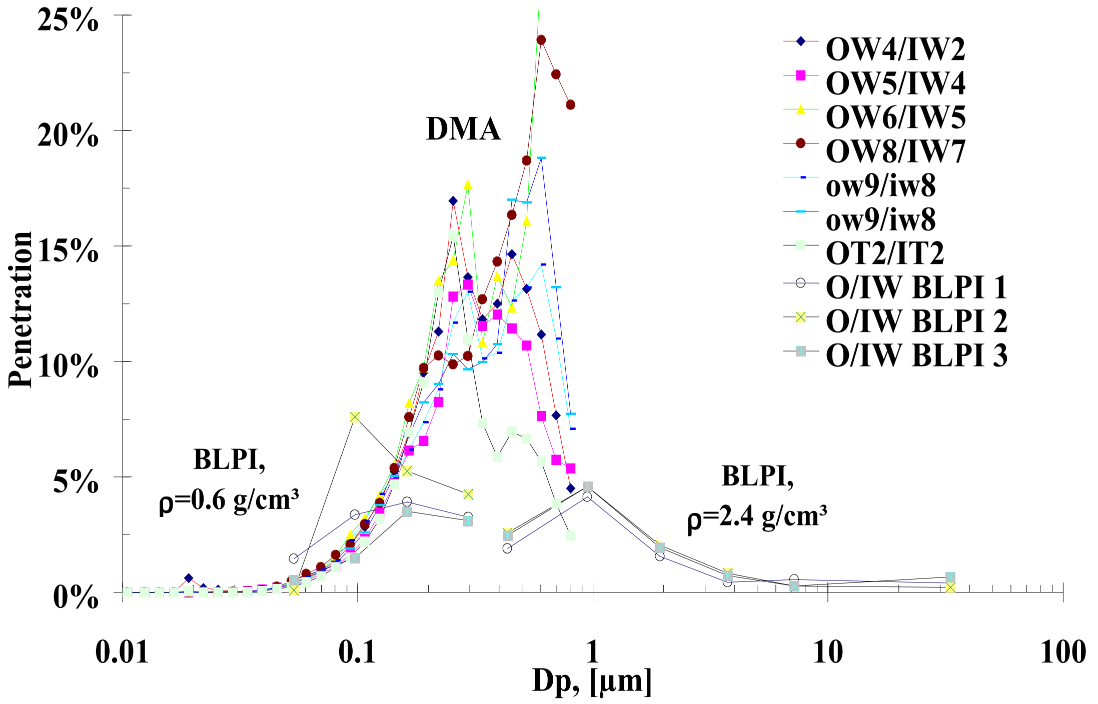

Figure 9. *Klein Kopije coal fly ash aerosol penetrations as determined with DMA and BLPI. Measured data in two days with different charging ratios (Ylätalo, 1996)*

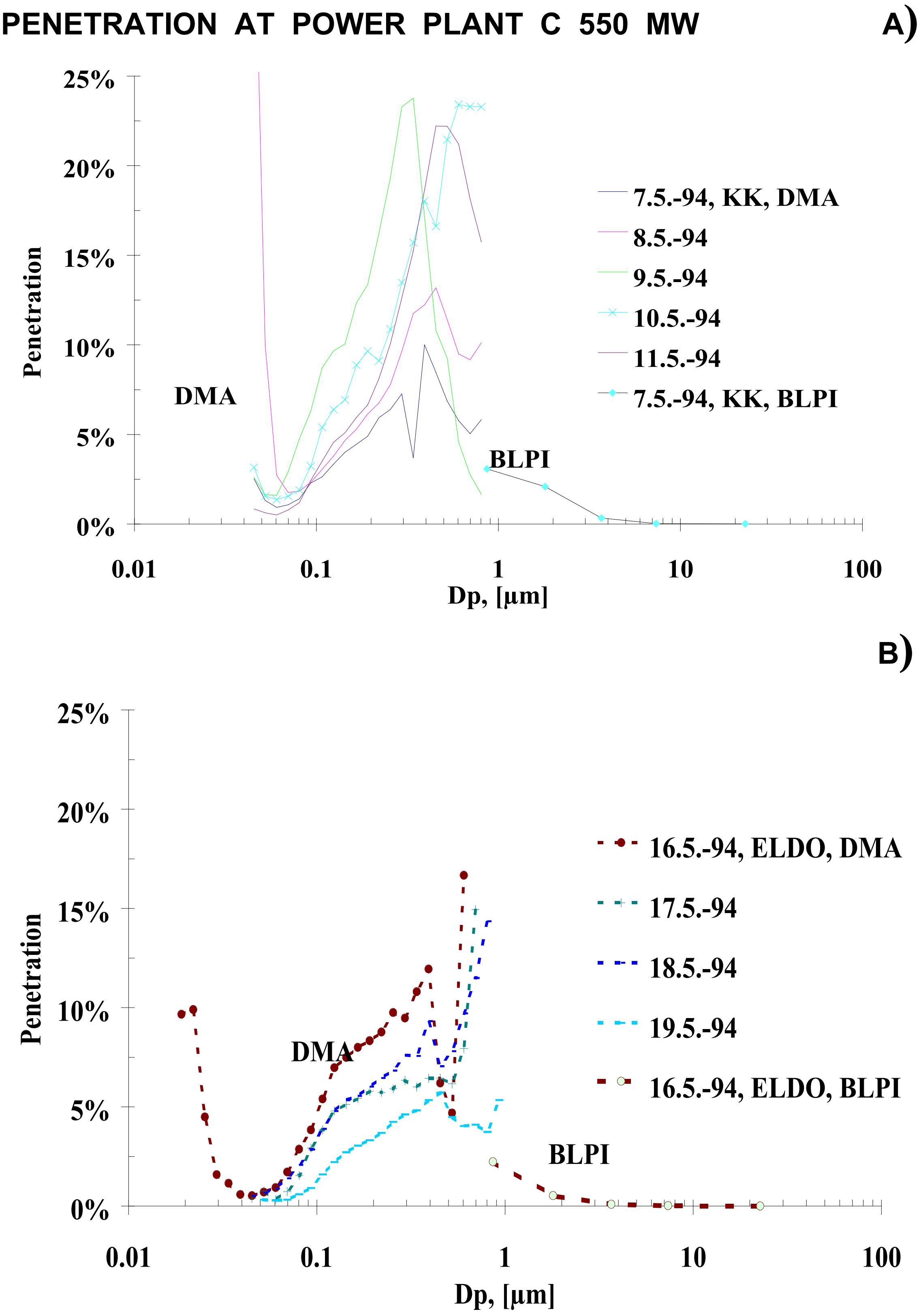

Figure 10. *South African Klein Kopije* ***A)*** *and Colombian Eldorado* ***B)*** *coals originating fly ash penetrations as determined with DMA and BLPI. The anomaly in curve 8.5. -94 at 0.05 µm occurs due to an ion induced nucleation in the ESP within a full charging ratio and ammonia (injected as a catalyst upstreams of the ESP) (Ylätalo, 1996).*

Table 1. *Features of the processes of the boilers A,B and C (Ylätalo, 1996).*

Nominal ratings	Load,	Burners	at Levels	Peaks			
				super micron	sub micron	inlet	outlet
Location, [MW]	[MW], coal	[#]		[#]	[#]	[10^7 #/cm^3]	[10^7 #/cm^3]
A, 315	208 PB	8	3	1	2	3.2	0.3
	234-263 PB	8/12	3/4	2	2	4-5	
B, 630	615 KK	36	3	1	1	3.2-4.6	1.3-2.6
C, 550	540 KK	16	4	1	1	3.7-3.9	1.3-2.9
	540 EL	16	4	1	1	6.7-7.4	2.5

Table 2. *Analysis of the coals in power plant A, B and C (Ylätalo, 1996).*

Coal	Polish bituminous, PB		Klein Kopije, KK South Africa		Eldorado, EL Kolumbia
Power plant, year Quantity (down)	A, 1991	A, 1992	B, 1993	C, 1994	C, 1994
Heating value, MJ/kg	30.5	27.8	27.2	27.4	30.0
Moisture, wt-%	1.0	2.3	7.4	9.9	13.5
Volatiles, wt-%	29.6	33.6	22.9	20.7	35.5
Ash, wt-%	9.5	12.8	15.9	17.1	10.2
Carbon, wt-%	79.4	71.7	68.1.	70.3	73.4
Hydrogen, wt-%	4.5	4.6	1.5	3.4	4.6
Oxygen, wt-%	(missing)	(missing)	10.3	7	9.4
Nitrogen, wt-%	1.2	1.3	3.5	1.7	1.5
Sulphur, wt-%	0.72	0.74	0.50	0.43	0.81

Table 3. *Analysis of the ashes in powerplants A, B and C (Ylätalo, 1996).*

Ash	Polish bituminous, PB		Klein Kopije, KK		Eldorado, EL
			South Africa		Kolumbia
Power plant, year	A, 1991	A, 1992	B, 1993	C, 1994	C, 1994
Mineral (down)					
Al_2O_3	24.0	24.5	33.2	31.4	21.0
CaO	2.4	3.9	6.2	7.3	4.1
Fe_2O_3	8.5	8.7	3.11	3.5	7.3
K_2O	2.3	2.4	0.65	1.0	2.1
MgO	2.4	2.6	1.36	1.7	2.1
Na_2O	1.0	0.75	0.17	0.3	0.6
SiO_2	44.6	45.3	47.7	45.3	55.7
Li_2O	(missing)	(missing)	0.07	0.0	0.0
TiO_2	1.3	1.2	1.73	1.6	0.9
P_2O_5	<0.10	0.91	1.20	1.2	0.3
SO_3	4.65	3.12	4.77	5.4	3.1

Table 4. *Comparision of the ESPs at the power plants A, B and C (Ylätalo, 1996).*

Quantity (down)	ESP at the Power Plant		
	A	B	C
Field number	3	5	5
Sections	1	4	4
Field height, [m]	13.5	12.5	12.5
Field length, [m]	3.75	2.8	3.75
Field width, [m]	10.8	14.1	10.8
Spacing, (wire to plate), [m]	0.15	0.3	0.4
SCA, $[m^2/(m^3/s)]$	83-97, 60	83	52,70,87
Electrical mode of operation	DC	Semipulse	Semipulse

Table 5. *Experiments with real scale electrostatic precipitators at the power plants A, B and C (Ylätalo, 1996).*

Experiment to influence on Penetration	Power plant			
	A, 1991	A, 1992	B, 1993	C, 1994
Load variation	208	263/234	630	550
Coal variation	PB	PB	KK	*KK/EL
Last block off	-	Yes	-	Yes
2nd last block off	-	-	-	*Yes
CR variation	-	-	Yes	Yes
Sieve rotation speed change	-	Yes	-	-
Number size range DMA [μm]	(0.017-0.8)	(0.017-0.8)	(0.017-0.8)	(0.017-0.8)
Mass size distributions	BLPI (0.01-20 μm)	BLPI	BLPI	BLPI
Ashes for CCSEM	Bottom, blocks 1,2,3	-	Bottom, blocks 1,3,5	Bottom, blocks 1,3,5

Table 6. *The number size distribution summary for the ashes (PB = Polish bituminous coal ashes, KK = Klein Kopije coal ashes, EL = Eldorado coal ashes) (Ylätalo, 1996).*

Power Plant/Coal	A/PB	B/KK	C/KK	C/EL
Number of sub micron modes	2	1/(2)	1	1
Inlet Peak location, μm	0.050/0.3	0.095/0.15	0.150	0.150
Outlet Peak location, μm	0.080/0.35	0.130/0.5	0.160	0.160
Inlet Particle Conc. 10^6#/cm³	25	70/0.4	39	38
Outlet Particle Conc. 10^6#/cm³	0.28	2.5/0.02	2	2

Table 7. *Tag categories and the scenarios for an example of radioactive source search.*

Feature	***Scenario***			***Added Measures***
	1	2	3	
Potential Target Substances	Uranium, enriched	Uranium, enriched	Ammonium compounds	
	Daughters	Daughters	Sulphates	
	Cobalt	Cobalt	Silver	
Random Occurrences	Fires, Maintenance of the cleaners	Fires, Maintenance of the cleaners	Explosions, Maintenance of the cleaners	
Natural Measures	Coarse sands	Coarse sands		Lack of Coarse certain sands
	Minerals	Minerals		Fluorescence of the tag particles
	Radio chem of the minerals	Radio chem of the minerals		Isotope tag in the added particles
	Particle size distribution	Fly ash size distribution	Particle size distribution	Unnatural particle sizes in the sample