MONITORING PM 10, PM 2.5 AND PM 1.0 EMISSIONS FROM INDUSTRIAL SOURCES AND DOMESTIC STOVES

Dr. Chr. Ehrlich, Dr. G. Noll and Dipl.-Ing. W.-D. Kalkoff, Halle (Saale)
Landesamt für Umweltschutz (LAU), Sachsen-Anhalt, Germany

1 ABSTRACT

Recent epidemiological studies have shown that fine particles in ambient air are more harmful than it was supposed to be. Therefore the latest legislation of the EC [1] includes PM 10 and PM 2.5 limit values at a low level. In Germany and in other European countries there are gaps in keeping these demanding goals. For the gap closure it is necessary to carry out research in fine particle emission data. Emission measurement investigations have been carried out into various industrial sources like cement kiln (industry), secondary copper smelter, glass industry, wood combustion, different coal fired power plants and into a small scale domestic heating unit with the help of cascade impactors (8-stage and 6-stage). In total 202 emission measurements at 34 industrial plants and 4 domestic stoves have been evaluated.
The results show a high amount of fine particles in the waste gases, dependent on the kind of industrial plant and on the used fuel. Approximately 75 % of the examined installations have at least an average PM 10-proportion > 90 %. Higher proportions of PM 2.5 und PM 1.0 were found at firing installations with coarser structured solid fuel in contrary to installations using finer structured pulverized fuel.
A distribution analysis of the dust constituents, like Pb, Cd, Cr, Ni, Zn of the examined fine dust fractions was carried out. In some cases the proportions of the dust constituents related to the mass of the particles for each examined impactor stage show a considerable enrichment of heavy metals in the finer particle fractions (grate cooler system). For other systems examined so far this strong enrichment effect could not be proven.

2 INTRODUCTION

On international and on European Union level [2], [3], [4], [5], [6] the health relevance of fine particles has been re-evaluated. The suggested limit values will be much stricter than the former standards. This will lead to limit value exceeding in certain areas not only in Germany [1].
The strict limit values imply the necessity for abatement measures of fine particle emissions. Emission abatement can only be achieved, however, if the emission structure of the sources of the fine particles and of the mass flow is known.
Since the former legislation contained only determinations of the total suspended matter emissions, the level of knowledge of fine dust emissions is still incomplete nowadays. The results of comprehensive measurement programmes in Germany, which are presented in this paper, shall help to clear up these problems.
The programmes contain measurements at industrial sources with a high proportion of the total dust emission in several German states, like Saxony-Anhalt [18], Bavaria [15] and Baden-Wuerttemberg [16]. Further measurements were carried out at installations, whose dust emissions include health-relevant substances like heavy metals and metalloids.
Investigations were taken in particular into installations of the cement industry, the copper metallurgy, glass industry, wood combustion and different coal fired power plants. First results have already been presented [7], [16].
In the new German States and in future EC member states like the Czech Republic, Poland and others brown coal briquettes are still frequently used for the heating of domestic stoves. Therefore fine dust emissions were measured at an iron stove using different briquette sorts. Examination of wood burning in domestic stoves and small scale combustion units was also included in the measuring programme.

3 EXPERIMENTAL

3.1 Basic experimental procedures

For the indication of particle dispersion in gases the aerodynamic diameter d_{ae} is used. This is defined as diameter of a sphere made of a material with the density of 1 g/cm³ with the same settling rate in gases as the regarded particles.
In [1] the reference method for the sampling and the measurement of the PM 10-concentration (as well as a provisional procedure for the sampling and measurement of the PM 2.5-concentration) in ambient air is mentioned

(appendix IX). The procedure is based on the separation of the PM 10 particle fraction according to the inertia principle and the following gravimetric measurement.
Analogous to this immission-related determination of PM 10 (and PM 2.5) fractionated dust emission measurements with cascade impactors shall be carried out according to an agreement [8]. Thus a measuring procedure is used which is based on the separation of particles following the inertia principle.
Moreover, the procedure combines the advantages of the relatively simple management and the in situ sampling without modification of the particle size distribution by agglomeration effects compared with other applicable particle size-selective procedures.
The basis of the sampling of particle size-dependent dust fractions by means of impactors is the utilisation of the different inertia of particles. A cascade impactor contains several impactor stages, which basically consist of a nozzle and an impact plate. Particles with sufficient inertia of the particle fraction, accelerated in the nozzle, strike the impact plate and are collected there. Particles of smaller inertia are separated at one of the following stages, so that fractions of equal particle size are collected at each stage. The not separated particles are collected on a backup filter, which is arranged behind the stages. The mass of the particles, separated on one stage, is determined after the sampling by difference weighing and can be analysed for dust constituents. If there is a larger proportion of coarser particles, the application of a preseparator is necessary.
Measurement planning and sampling with impactors must be carried out similar to measurements of the total dust emissions, i. e. grid measurement with isokinetic sampling. However, the gas flux is no longer freely selectable after having defined the probe diameter, but must be kept constant for all measuring points of the grid measurement in contrary to the dust emission measurements without impactors. Therefore only measuring points with approximately the same gas velocity (permissible variation: max. ± 30%) can be sampled in an individual measurement. Otherwise several measuring processes are necessary. The sampling period should be chosen to ensure a collection of a sufficient mass of dust per impactor stage to permit weighing with sufficient accuracy and to prevent overloading of the stage.

3.2 Sampling technique

The sampling was carried out with the help of 8-stage or 6-stage Anderson impactors type Mark III (material: stainless steel) and/or with the help of a 6-stage impactor of the Stroehlein company type STF 1 (material: titanium). Preliminary tests have shown that the particle size distribution, detected with these two impactors, coincides under the same sampling conditions.
Both impactors are heatproof up to 850 °C. For the particle sampling perforated sampling plates and backup filters, made of glass fibre material, were used. The gas volume flux, sucked into the impactor, was measured e.g. with a thermal mass flow meter.

3.3 Sampling and analysis

The sampling and analysis of the particle measurements have been carried out in accordance with the German guideline VDI 2066 Bl 5 [9]. In order to be able to determine the necessary sampling periods for the impactor measurements, at first the emission concentration of the total dust was measured. Thus on the one hand, overloading of the impactor stages should be prevented, on the other hand, a sufficient dust mass per impactor stage to permit weighing with sufficient accuracy should be collected.
To avoid very different loadings of the individual sampling plates, preliminary tests were carried out in order to take corresponding measures for the actual sampling (e.g. modification of the sampling period, application of a pre-separator). Because at the industrial plants, which were investigated, small emission concentrations of total dust were found, sometimes very long sampling periods (as many as 18 hours, in exceptional case 52 hours) were necessary to guarantee a sufficient filter covering.
On the other hand, the high dust loadings in the waste gas of the investigated iron stove (coal fired residential heating unit) required the determination of a sampling period, which is substantially shorter than the time for the complete burn-up of the fuel. Our own investigations have shown that in the first third of the combustion cycle (in this time the impactor measurements were carried out) the emission of the particles was almost complete. The impactor measurements were carried out as grid measurement according to the principles of guideline VDI 2066 Bl. 1 [10]. Only measuring points with approximately the same gas velocity were sampled. In an exceptional case this led to corrections of the number of measuring points. In the periods of the impactor sampling the operating parameters were recorded.
In some cases constituents of the individual dust fractions (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn, Sn and Tl) were analysed (according to guidelines VDI 3868 Bl. 1 [11] and VDI 2268 Bl. 1 [12]).

3.4 Quality assurance measures

All tests were accompanied by a set of quality assurance measurements according to DIN EN 45001 [13]. All pollutant emissions were detected with standardised measuring procedures (VDI guidelines, DIN EN). If applicable, certified continuously operating measuring instruments were used. The parameter of the sampling, analysis as well as the operational parameters, necessary for the evaluation of the detected emission concentrations, were recorded [14], [15], [16], [17], {18] according to the specifications of the standard form of the test report.
It must be considered that for the measurement of dust constituents (heavy metals) an impactor made of titanium and an impactor made of high-grade steel were used.
If you compare the size distributions of the dust constituents, detected with the two different materials of the impactors, you see that there is no significant difference between the results at the same plant. The metal blind values of the sample plates were determined for each impactor stage.

4. EXAMINED PLANTS, SAMPLING CONDITIONS AND RESULTS

The examined plants with their appropriate performance characteristics during the sampling periods, the used fuel and the parameters of the available gas cleaning units are specified in table 1.
Within the framework of the measuring programmes for the following types of industrial installations fine dust emission determination has been carried out and has been analysed:
- heat production, energy
- the industry of building materials
- ceramic and glass industry
- production and processing of steel, iron and other metals
- chemical industry

The types of industrial installations and domestic stoves and the measured average PM 10, PM 2.5 or PM 1.0-proportions in the emitted dust are specified in table 1. The analysis of these measurement programmes shall contribute to and deepen technology-referred knowledge of particle size distributions of emitted types of particles in exhaust gases.

Table 1: Plants with their appropriate performance characteristics during the sampling periods, average parts of PM 10, PM 2.5, PM 1.0

No.	plant, performance rating	reference	input substances fuel	number of sampling	performance during measurement period	dust separation	PM 10 in %	PM 2.5 in %	PM 1.0 in %
1a	rotary kiln, cement industry	[14], [18]	brown coal dust, waste oil, natural gas...	3	87 t/h clinker (do)[1)]	ESP[5)] horizontal	96.7	82.3	52.6
1b	rotary kiln, cement industry	[14], [18]	brown coal dust, waste oil, natural gas...	2	85 t/h clinker (co)[2)]	ESP[5)] horizontal	96.2	69.5	39.0
2a	grate cooler, cement industry	[14], [18]	clinker	3	72 t/h clinker	fabric filter	43.3	3.8	1.2
2b	grate cooler, cement industry	[14], [18]	clinker	3	72 t/h clinker	fabric filter	23.6	2.6	0.6
3	converter, copper industry	[14], [18]	brass scrap, copper dross, coke...	3	59 t/charge	fabric filter	96.3	76.3	51.3
4	industrial power station 180 MW, pulverized brown coal firing	[14], [18]	extracted dry brown coal	3	180 MW	ESP[5)] horizontal, scrubber	90.7	75.5	52.0
5	industrial power station 146 MW, grate firing	[14], [18]	brown coal briquettes, ground limestone	3	114 MW	ESP[5)] horizontal, drying-desulphurization	92.0	74.0	47.7
6	industrial power station 119 MW, fluidized bed combustion	[14], [18]	lignite	3	114 MW	ESP[5)] horizontal, desulphuri-zation, denoxing process	97.0	65.7	25.5
7aa	small scale firing unit, iron stove 6 kW	[14], [18]	LAUBAG-bcb[3)] (after cooling)	3	6 kW	without	93.4	85.4	76.6
7ab	small scale firing unit, iron stove 6 kW	[14], [18]	LAUBAG-bcb[3)] (before cooling)	2	6 kW	without	91.6	84.1	75.7
7ba	small scale firing unit, iron stove 6 kW	[14], [18]	MIBRAG-bcb[3)] (after cooling)	3	6 kW	without	95.9	83.5	63.8
7bb	small scale firing unit, iron stove 6 kW	[14], [18]	MIBRAG-bcb[3)] (before cooling)	3	6 kW	without	90.5	80.5	63.1
7ca	small scale firing unit, iron stove 6 kW	[14], [18]	Polish bcb[3)] (after cooling)	2	6 kW	without	95.8	80.8	65.0
7cb	small scale firing unit, iron stove 6 kW	[14], [18]	Polish bcb[3)] (before cooling)	2	6 kW	without	93.5	77.0	63.3
7d	small scale firing unit, iron stove 6 kW	[14], [18]	Bashkirian bcb[3)]	3	6 kW	without	91.3	80.8	70.6
7e	small scale firing unit, iron stove 6 kW	[14], [18]	MIBRAG-bcb[3)]	3	6 kW	without	94.0	85.6	75.4
8a	kiln, Lepol-processing	[15]	raw meal, coal, waste oil, used-tyre	6	41 t/h clinker (do)[1)]	ESP[5)]	92.4	50.1	39.2
8b	kiln, Lepol-processing, clinker cooler ex-haust gas	[15]	raw meal, coal, waste oil, used-tyre	6	41 t/h clinker (co)[2)]	ESP[5)], multi-cyclone, cyclone	98.0	64.5	23.2
9a	heat exchanger, cement industry	[15]	raw meal, heavy oil, used-tyre	6	118 t/h clinker (do)[1)]	ESP[5)]	99.4	75.2	42.9
9b	heat exchanger, cement industry	[15]	raw meal, heavy oil, used-tyre	6	118 t/h clinker (co)[2)]	ESP[5)]	100	62.1	25.0
10	isostatic compression press	[15]	porcelain substance	4	400 piece/h	fabric filter	94.9	57.4	38.3

No.	plant, performance rating	reference	input substances fuel	number of sampling	performance during measurement period	dust separation	PM 10 in %	PM 2.5 in %	PM 1.0 in %
11	glass industry (batch glass)	[15]	cullet, batch, natural gas	6	223 t/d	lime-sorption, ESP[5)]	95.3	53.5	21.2
12	glass industry (flat glass)	[15]	cullet, batch, natural gas	6	508 t/d	desulphurization (lime), ESP[5)]	93.2	44.8	23.7
13	aluminum-remelt heat	[15]	natural gas, aluminium-scrap	1	without information	reactor (limehydrat), fabric filter	98.5	72.0	35.8
14a	aluminium-chippings drying plant	[15]	aluminium-chippings	2	1.8 t/h	thermic afterburning, fabric filter	95.4	46.1	20.3
14b	aluminium-chippings drying plant	[15]	aluminium-chippings	2	1.5 t/h	thermic afterburning, fabric filter	98.4	52.9	32.8
15	cupola	[15]	iron-scrap, coke, limestone	4	6.4 t/h	fabric filter	87.9	43.8	19.8
16	induction furnace, iron casting plant	[15]	iron-scrap	4	2.4 t/h	fabric filter	77.4	48.6	18.1
17	sand conditioning	[15]	sand, betonit	2	20 t/h	fabric filter	86.8	36.1	21.2
18	cupola	[15]	iron-scrap, coke	6	15 t/h	cyclone, venturi scrubber, recuperator	95.4	88.1	72.4
19	Cast iron processing	[15]	cast iron components	6	150 t/month	fabric filter	81.8	28.4	9.5
20	sand conditioning	[15]	arena, betonit, coal dust	6	27...30 t/h	ESP[5)]	74.0	15.5	1.4
21	mix plant	[15]	asphalt granulate, oil	6	150 t/h	fabric filter	93.1	29.2	8.3
22	electric shaft furnace, manufacture of silicium	[15]	coal, charcoal, coke, chips	6	3.2 t/h	fabric filter	92.8	45.4	24.9
23a	heavy oil firing (without SNCR[4)]) 10 MW	[15]	heavy oil	3	5 MW	additive	98.0	81.9	64.3
23b	heavy oil firing (without SNCR[4)]) 10 MW	[15]	heavy oil	1	8.5 MW	additive	91.3	64.7	49.9
23c	heavy oil firing (with SNCR[4)]) 10 MW	[15]	heavy oil, urea	1	5 MW	additive	97.1	77.5	55.8
23d	heavy oil firing (with SNCR[4)]) 10 MW	[15]	heavy oil, urea	1	8.5 MW	additive	93.5	68.0	50.7
24a	firing plant 175 kW	[16]	chips	1	177 kW	cyclone	93.5	84.4	80.0
24b	firing plant 175 kW	[16]	chip board	1	139 kW	cyclone	98.5	86.2	79.8
25a	firing plant 150 kW	[16]	chips	1	148 kW	without	95.1	72.1	66.9
25b	firing plant 150 kW	[16]	chips	1	43.4 kW	without	99.6	93.8	86.9
25c	firing plant 150 kW	[16]	joinery residues	1	133 kW	without	74.2	57.8	52.7
25d	firing plant 150 kW	[16]	coloured pencil residues	1	112.5 kW	without	71.3	43.7	39.0
26a	firing plant 450 kW	[16]	hogged wood	1	416.5 kW	multi-cyclone	100	96.5	89.0
26b	firing plant 450 kW	[16]	hogged wood	1	273 kW	multi-cyclone	98.0	79.7	63.1

No.	plant, performance rating	reference	input substances fuel	number of sampling	performance during measurement period	dust separation	PM 10 in %	PM 2.5 in %	PM 1.0 in %
27a	small scale firing unit 9kW	[16]	pieces of beech	5	9.4 kW	without	98.9	95.8	92.8
27b	small scale firing unit 9kW	[16]	pieces of beech	3	7.5 kW	without	98.2	90.2	70.9
27c	small scale firing unit 9kW	[16]	pieces of pine	1	8.2 kW	without	98.9	95.2	91.8
27d	small scale firing unit 9kW	[16]	pieces of pine	2	6.8 kW	without	99.2	97.6	94.1
28a	chimney stove 6 kW	[16]	pieces of beech	4	5.7 kW	without	99.7	98.4	87.3
28b	chimney stove 6 kW	[16]	pieces of beech	5	4.1 kW	without	97.8	95.5	86.6
29	pellet stove 8.5 kW	[16]	pellets of wood	1	8.0 kW	without	99.0	95.3	92.9
30a	grate firing 1.4 MW	[16]	saw chippings, saw dust	1	0.9 MW	cyclone	98.6	70.4	49.0
30b	grate firing 1.4 MW	[16]	saw chippings, saw dust	2	1.3 MW	cyclone	98.3	67.7	44.8
31	grate firing 0.8 MW	[16]	saw chippings, saw dust	3	0.8 MW	cyclone	98.3	62.6	36.0
32	grate firing 3 MW	[16]	hogged wood	4	1.3 MW	cyclone	98.0	91.7	85.2
33	grate firing 1.1 MW	[16]	piece of wood, saw chippings	5	1.0 MW	cyclone	89.8	55.0	43.2
34	grate firing 2 MW	[16]	hogged wood, wood waste	4	1.5 MW	ESP[5)]	89.2	67.1	62.4
35a	grate firing 4.8 MW	[16]	wood chippings, bark	2	2.9 MW	ESP[5)]	23.6	6.3	5.5
35b	grate firing 4.8 MW	[16]	wood chippings, bark	3	4.8 MW	ESP[5)]	52.8	29.6	27.5
36a	grate firing 7.9-9.5 MW	[16]	wood, wood chippings	3	5.8 MW	ESP[5)]	73.5	53.6	46.3
36b	grate firing 7.9-9.5 MW	[16]	natural gas, wood, wood chippings	3	8.6 MW	ESP[5)]	80.9	56.9	46.1
37	grate firing 15 MW	[16]	hogged wood, wood waste, wood chippings	4	15 MW	ESP[5)]	87.1	52.7	33.8
38a	grate firing 1.5 MW	[16]	hogged wood	2	0.5 MW	chimney gas condensation, multi-cyclone	99.8	99.5	95.6
38b	grate firing 1.5 MW	[16]	hogged wood	2	0.9 MW	chimney gas condensation, multi-cyclone	100	97.2	92.7

[1)] do = direct operation

[2)] co = compound operation

[3)] bcb = brown coal briquette

[4)] SNCR = selective non catalytic reduction

[5)] ESP = electrostatic precipitator

5 RESULTS AND DISCUSSION

5.1 Typical particle size distribution of the emission samplings

In the following figures the particle size distribution of the particular emission samplings in the distribution grid of Rosin-Rammler-Sperling-Bennett (RRSB) is shown. An average out of the particular size distribution is very difficult because of different aerodynamic diameters which are determined at each individual measurement. So only some representative distribution is shown (No. see table 1).
In figure 1 the particle size distribution of industrial power stations shows that the fluidised bed combustion results in a lower fine dust proportion in the diameter range < 4 µm but in a higher proportion for d_{ae50} > 4 µm.

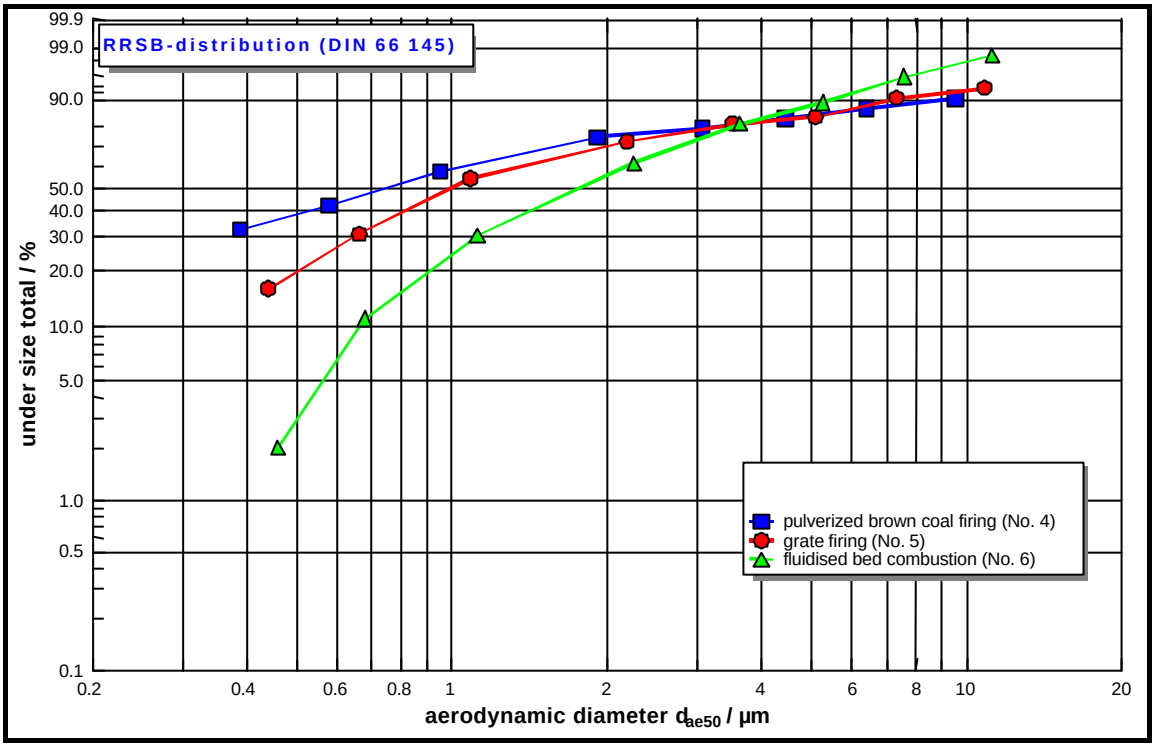

Figure 1: Industrial power stations

In the following figure 2 the determined particle size distribution of the dust in the purified gas of plants in the cement industry is shown.
The high to very high proportion of fine particles of the two types of rotary kilns with compound or direct operation is clearly visible. It is shown that the particle size distribution was somewhat coarser in the compound operation than in the direct operation.
The results of the tests at the examined grate cooler show a substantially coarser particle size distribution in comparison to the rotary kiln despite smaller total dust loadings (the at first unexpected results could be confirmed by repetition measurements after half a year). According to the information of the Research Institute of the Cement Industry Duesseldorf [19] this could be caused by the fact that the fabric filter, used in the grate cooler for dedusting, is subject to a high wear by clinker dust. Porosity in the filtering medium could lead to the passing of rougher particles, too. Since the raw gas dust concentration at the clinker cooler is usually relatively small, defects in a filter tube do not necessarily lead to a high emission concentration. From literature it is known that agglomerations are possible particularly behind fabric filters. This depends on the dust characteristics and the interactions of the particles at the filter cake in the purified gas. That could lead to a coarser particle size distribution. The repeated tests at the same plant also showed a further coarsening of the particle size distribution.

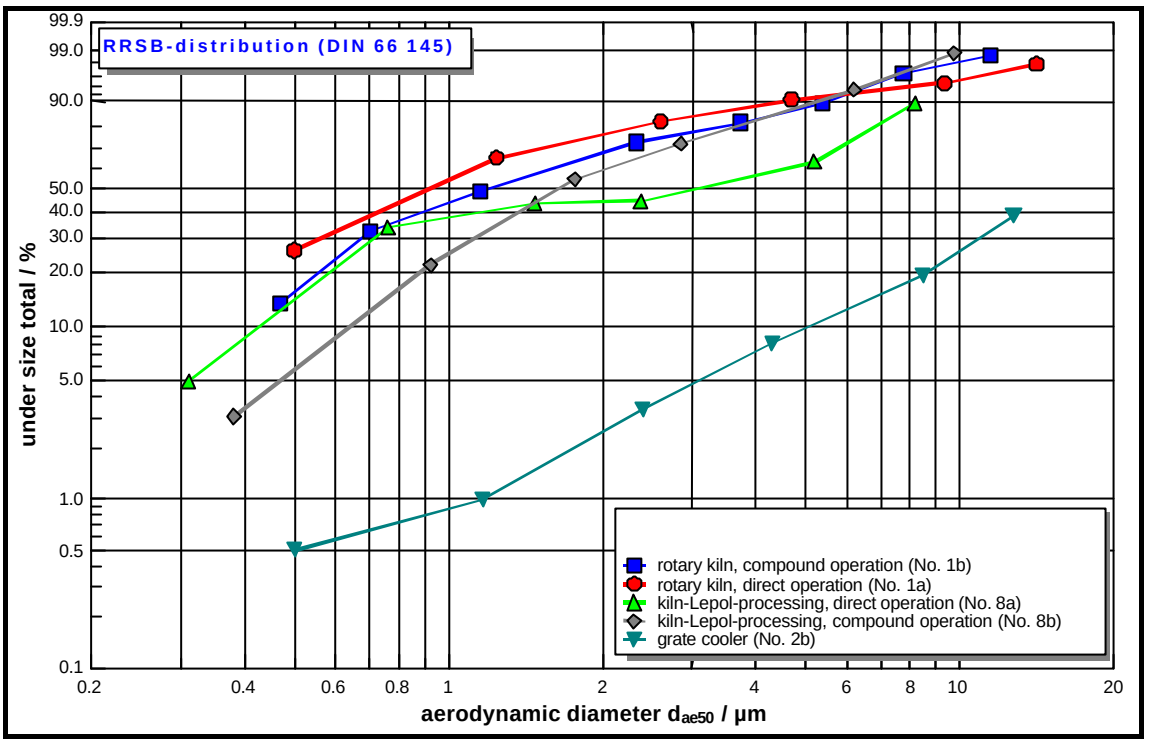

Figure 2: Cement industry

In figure 3 the particle size distribution of small scale firing units is shown. The influence of the fuel is significant. The wood fuel results in finer particles in proportion to the coal fuel.

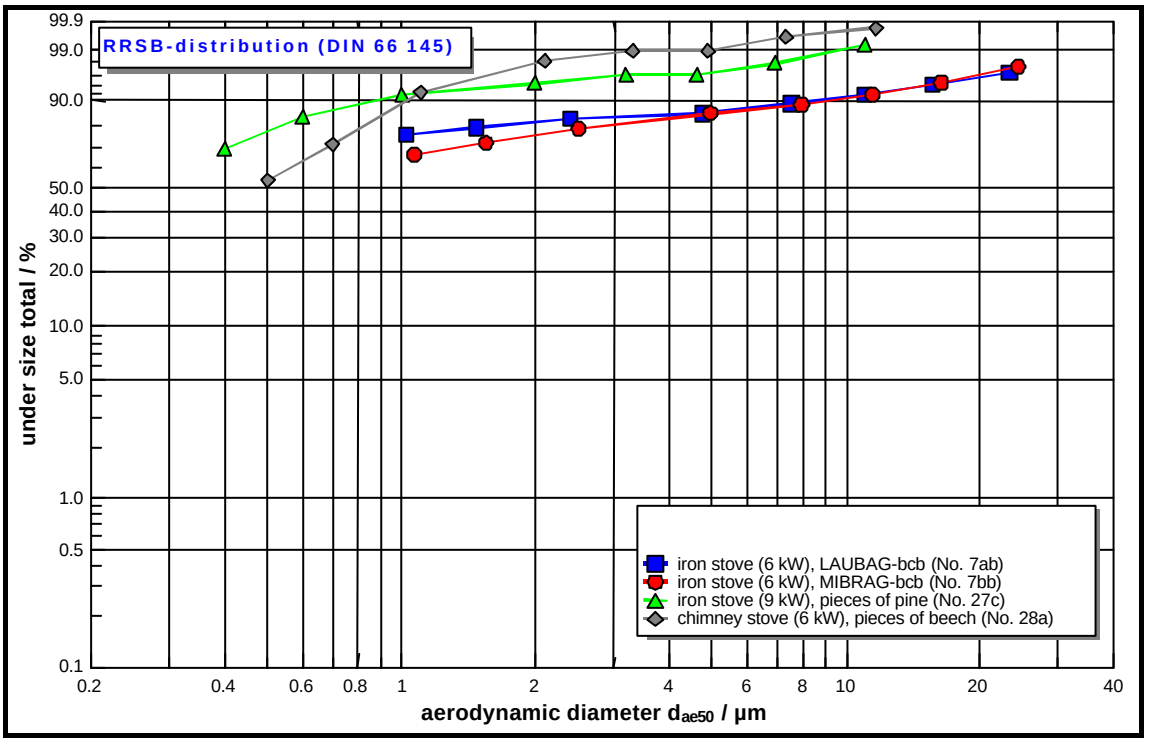

Figure 3: Small Scale Firing Units

It seems that at heavy oil firing plants only the power influences the particle distribution (see figure 4). The input of urea is not significant.

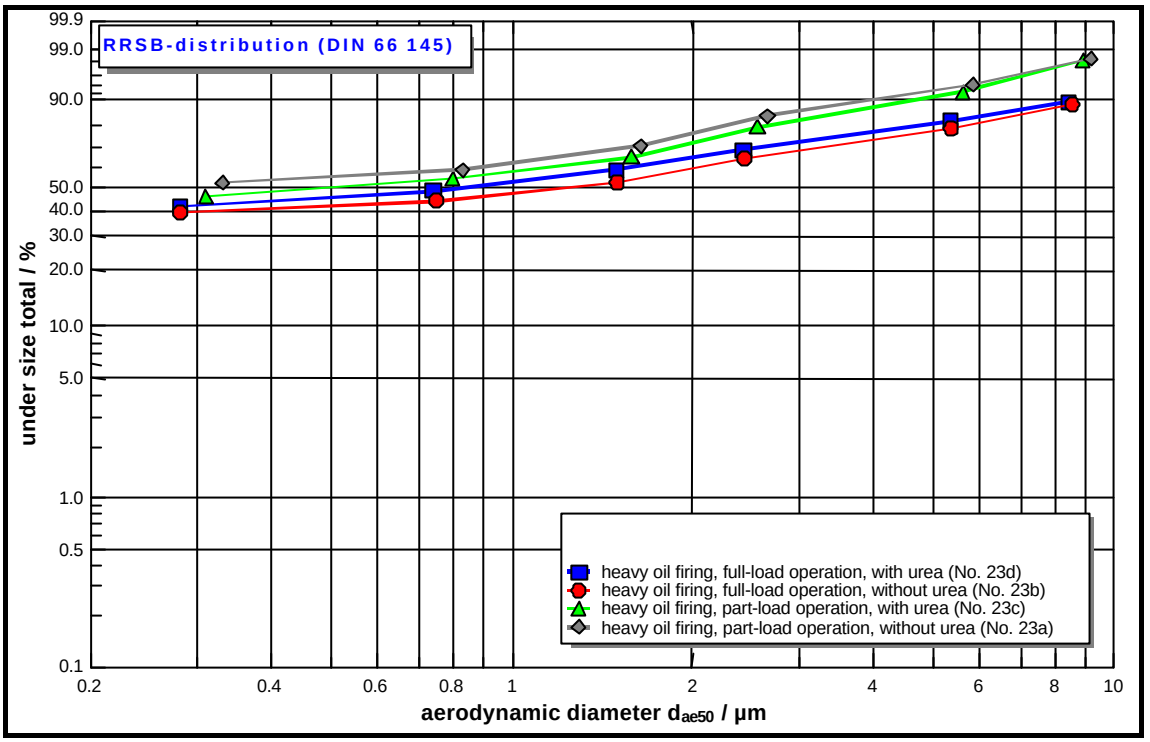

Figure 4: Heavy oil firing

Coarse particle size distribution has been found at sand conditioning plants (see figure 5).

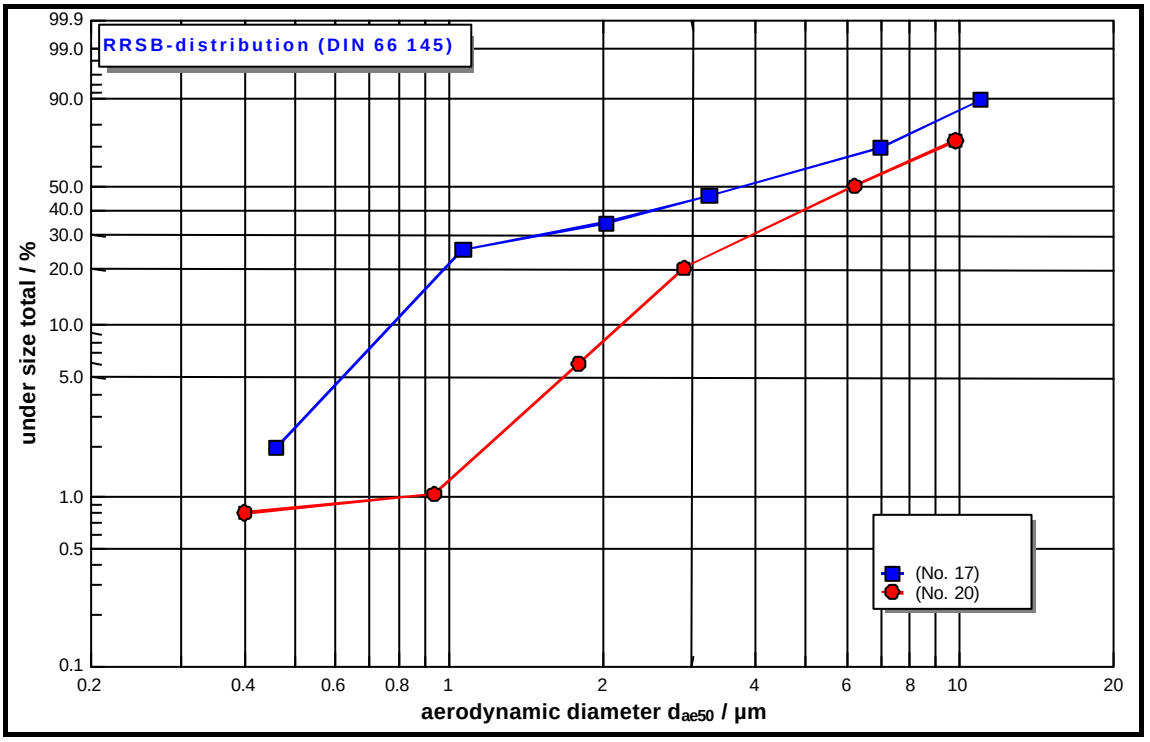

Figure 5: Sand conditioning

The input material is the major influencing factor at a wood firing plant (see figure 6).

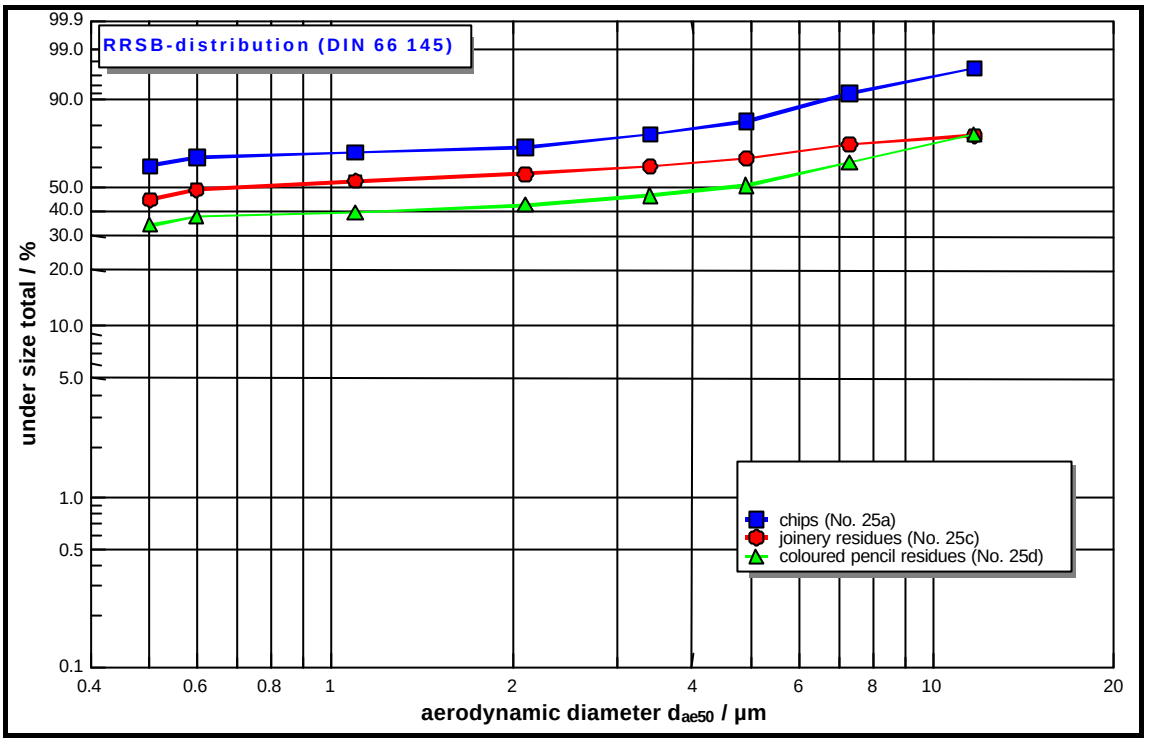

Figure 6: Wood firing plant

5.2 Analysis of technology-specific relationships between the proportions of PM 10, PM 2.5 or PM 1.0 in the emitted dust

To find out validated technology-specific determination of the proportions of PM 10. PM 2.5 or PM 1.0 in the emitted types of particles is very difficult in relation to the multiplicity of possible factors of influence and to the limited extent of available data.
Nevertheless, it was given a trial to find out trends at least. The most individual measured values have been determined for combustion plants and domestic stoves (altogether 63 measured values of the PM 10-proportion in the emitted dust).
But also for this field statistically validated analyses are difficult due to the multiplicity of possible measured variables on the amount of the proportion of PM 10, PM 2.5 or PM 1.0. The particle size distribution of particles in the exhaust gas can e.g. be influenced by the firing and/or dedusting technology, by the type of fuel, by the amount of the thermal output in the sampling period and by existing exhaust gas way and boundary conditions.
So it was expected that other proportions of fine particles in the exhaust gas of an industrial power station with fluidised-bed combustion of raw brown coal (lignite) and downstream electrostatic precipitator (ESP) as well as desulphurization would be found than in the exhaust gas of a wood-fired domestic stove.
In order to find out general trends, all average results of the examined plants and stoves were arranged according to the size of PM 2.5-proportions independently of the possible factors of influence such as firing technology, output, kinds of fuel as well as exhaust gas cleaning (figure 7).

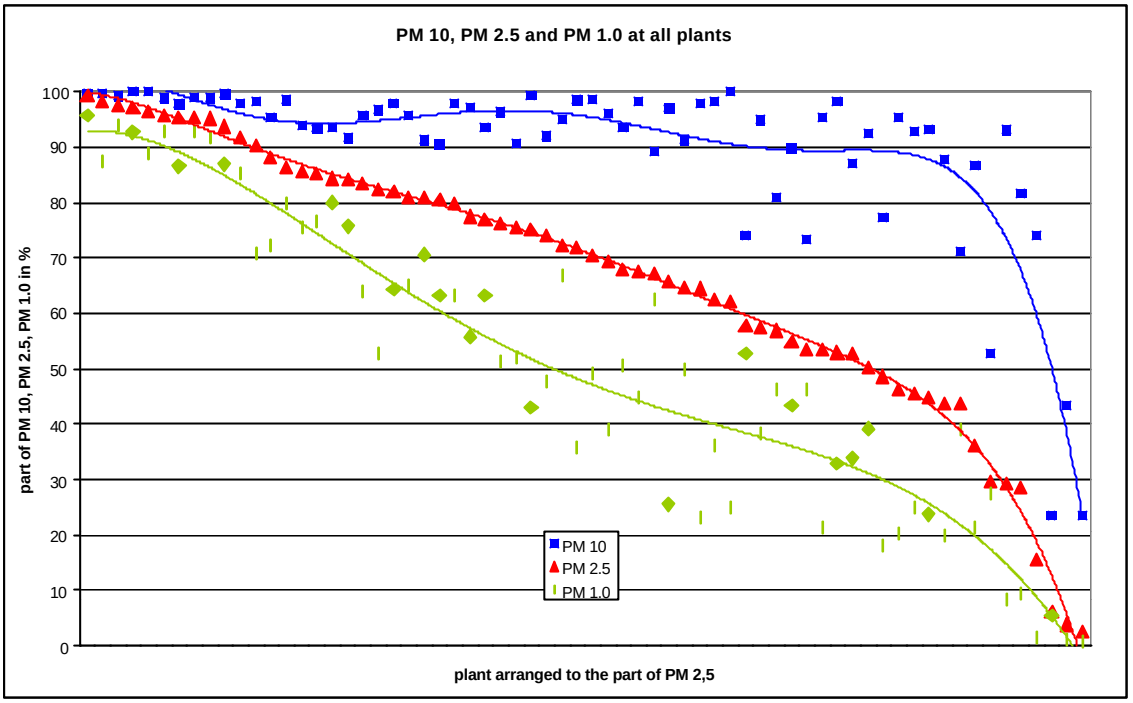

Figure 7: Average proportion of PM 10, PM 2.5 and PM 1.0 at all examined plants

It is obvious that the largest part of the examined installations emits dust with a high proportion of fine particles (more than 90 % particle size < 10 µm). Approximately 75 % of the examined installations have at least an average PM 10-proportion > 90 %. Considerably smaller PM 10 – and as a rule smaller PM 2.5 and PM 1.0-proportions were found at a grate cooler in the cement industry (two measuring periods) with fabric filter (plant number 2), at a combustion unit (150 kW) without dedusting, burning waste wood or coloured pencils (plant number 25), at a combustion unit with electrical gas cleaning during the firing of wood chippings and bark (plant number 35) in the part and full load operation, at a combustion unit with electrical gas cleaning during the firing of wood and wood chippings (plant number 36) as well as at a sand reprocessing system with electrical gas cleaning (plant number 20).
Far higher PM 10-proportions in the exhaust gas were found at this installation without dedusting when other kinds of fuel (hogged wood) were used. A dependency of the PM 10-emission concentration on the kind of the used exhaust-gas cleaning system could not be determined. Thus also smaller PM 10-proportions were determined- partly considerably below 90 % at systems with fabric filter (e.g. grate cooler). On the other hand, domestic stoves for solid fuel without separators contained very high amounts of fine particles in the exhaust gas.
It must be assumed that other technology-related influences on the amount of the fine particle proportions in the exhaust gas are more dominating, than the influence by the kind of the exhaust gas cleaning technique. The technical status of the dedusting or the separation function of the dedusting unit play probably an important role in dependence of the raw gas particle size distribution. But this was not further dealt with.
If the proportions of PM 1.0 and PM 10 are analysed similar to those of PM 2.5 (figure 7) you can see that they essentially follow the trend of the PM 2.5-proportion. But in many cases there are considerable deviations from this trend line. It was generally noticed that deviations from the trend to smaller PM 2.5 and PM 1.0-proportions were found; that means to a coarser particle size distribution in the emitted dust when firing fuel in pulverized or in liquid form. That concerns such fuel like wood chippings, saw chippings, pencil residues, raw brown coal, dried brown coal (approx. 1mm grain size) or added limestone for the desulphurization of the exhaust gases. This is also right for the examined heavy fuel oil firings.
The figures 8 and 9 show a comparison of fine particle proportion in the exhaust gas of firing installations using solid fuel, structured in coarse pieces and firing installations using fine structured (pulverized) solid or liquid fuel.

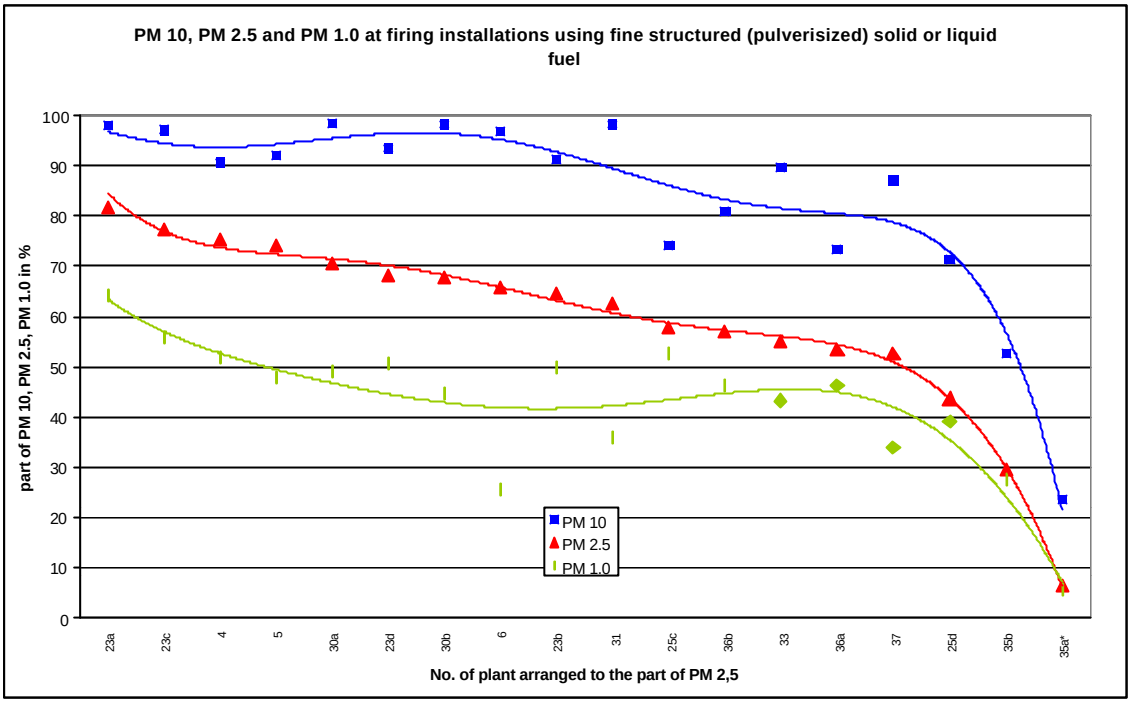

Figure 8: Average proportion of PM 10, PM 2.5 and PM 1.0 at firing installations using fine structured fuel

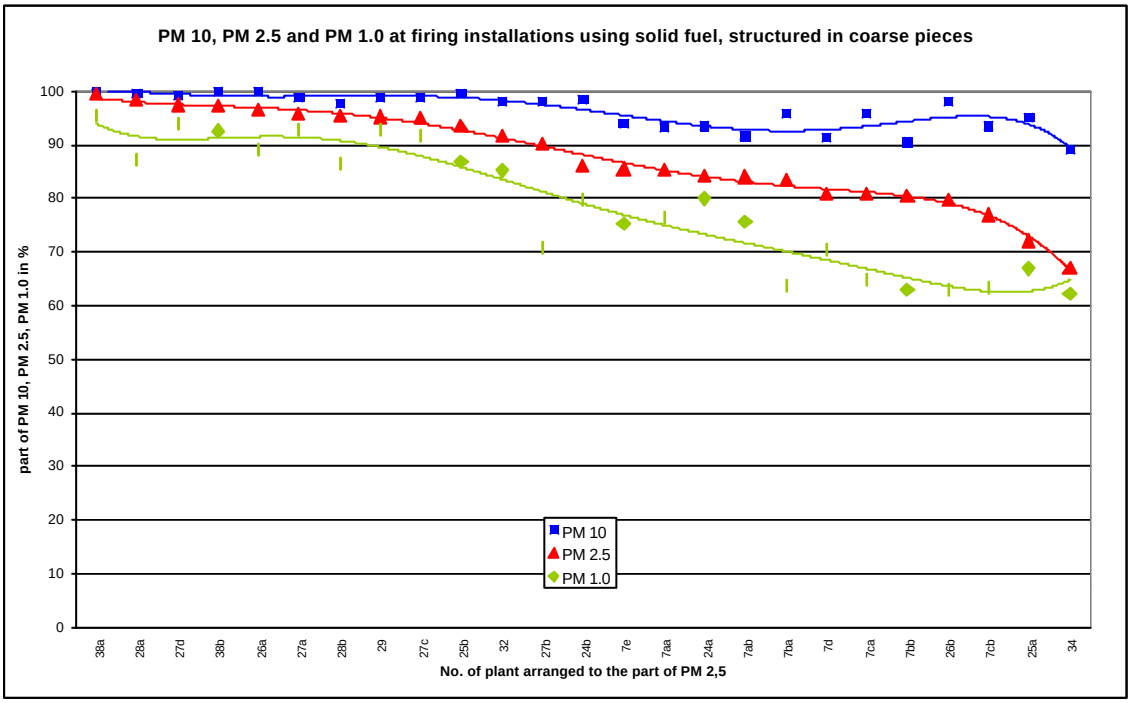

Figure 9: Average proportion of PM 10, PM 2.5 and PM 1.0 at firing installations using fuel, structured in coarse pieces

Clearly lower proportions of PM 1.0. PM 2.5 and partly of PM 10 are found at firing installations using fine structured fuel than at firing installations using fuel, structured in coarse pieces. At the first mentioned installations either an almost complete combustion of the fuel particles takes place or it comes to an agglomeration of fine particles in the cooling process of waste gas. The geometrical structure of the fuel and the associated quality of the complete combustion are obviously essential for the amount of the proportion from fine to ultra fine particles in the exhaust gas.

The following figure shows the proportions of PM 1.0. PM 2.5 and PM 10 at other non firing plants.

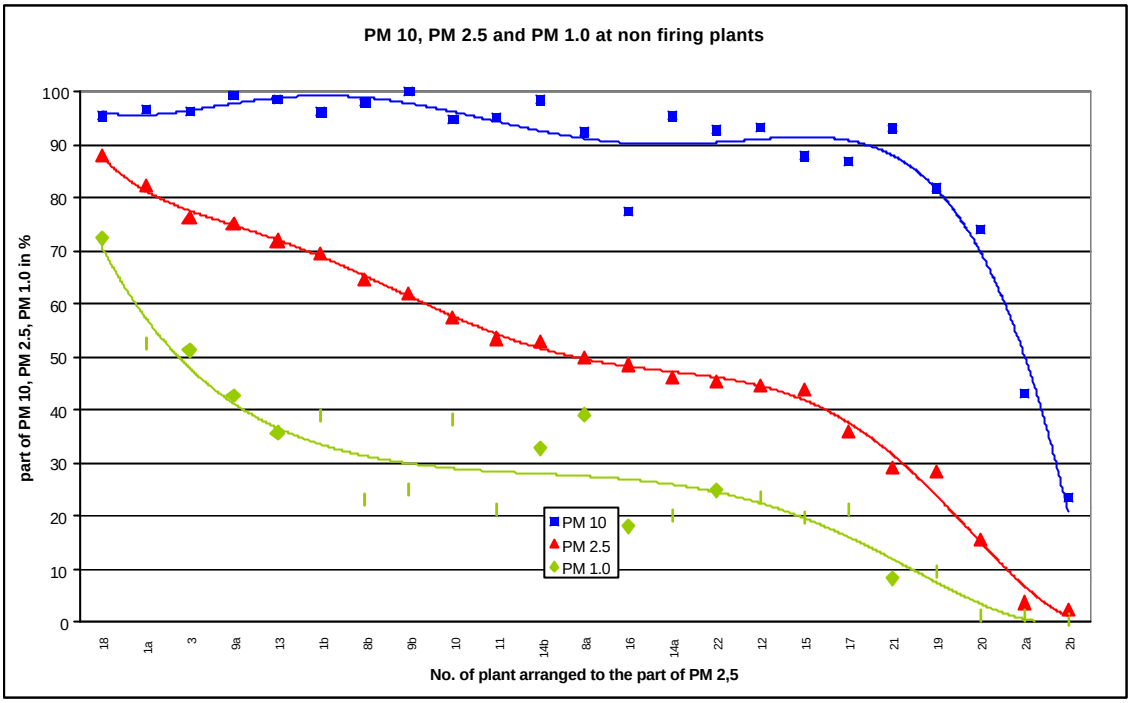

Figure 10: Average proportion of PM 10, PM 2.5 and PM 1.0 at non firing installations

5.3 Distribution of the dust constituents

Discussing the health damage mechanisms of fine and ultra fine types of dust, the dust constituents always play a special role, since particles are not homogeneous substances but much more complicated ones both in physical and chemical regard. Therefore dust and dust are not identical, but the health effect of the dust particles also depends on the origin of the dust.
A distribution analysis of the dust constituents of the examined fine dust fractions shows a differentiated picture. A set of particle size distribution of the examined systems shows a similar undersize total distribution for the particle masses and for the masses of the constituents in a certain range e.g. for the investigations into the converter system (figure 11).

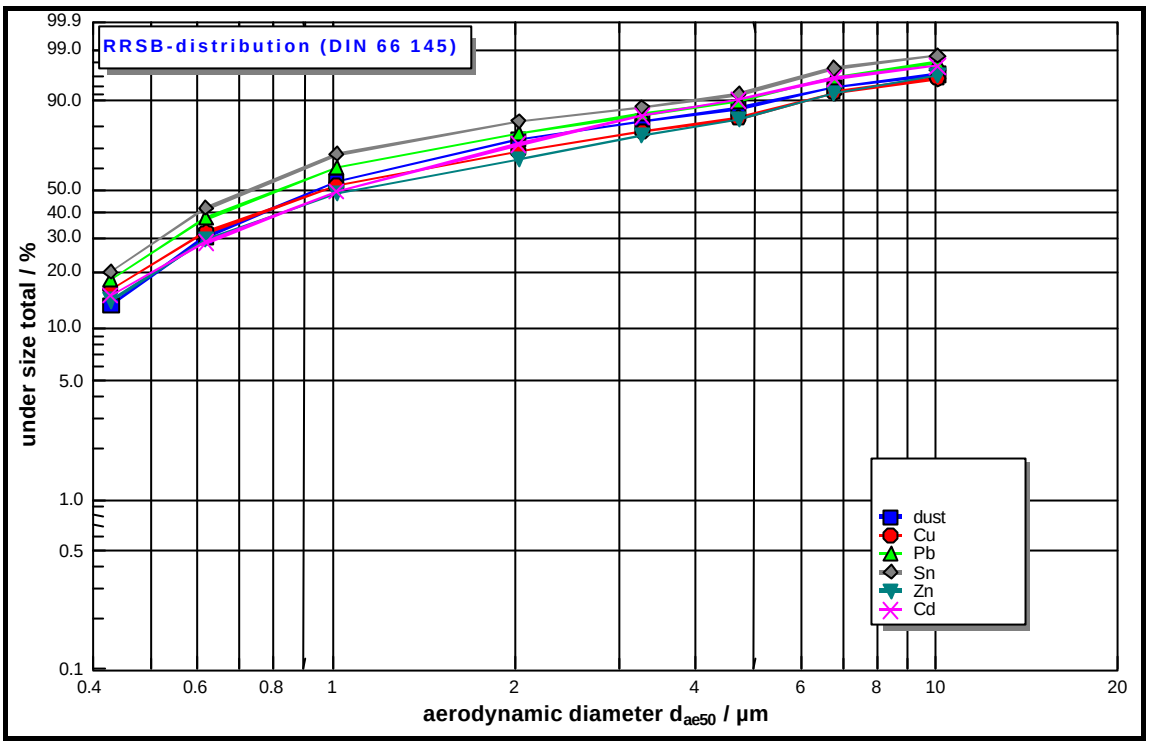

Figure 11: Converter, distribution of dust and dust constituents

The distribution of the dust constituents of the particle fractions at the examined grate cooler system (figure 12) is very different. These results could also be confirmed by repeated measurements after half a year with clearly increased total dust contents.

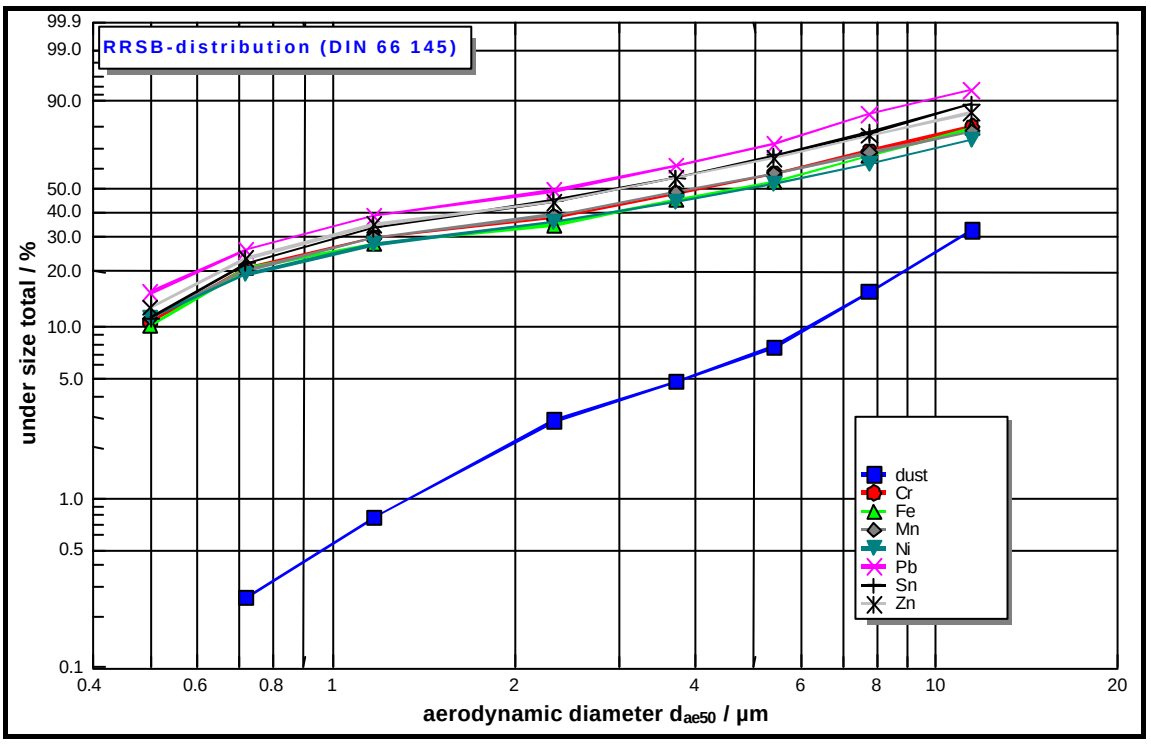

Figure 12: Grate cooler, distribution of dust and dust constituents

In a second analysis the dust constituents of the fine dust emissions (proportions of PM 10) of different plants were compared. As expected, very high heavy metal proportions (e.g. lead and cadmium) were found in the fine dust of the examined system in the secondary copper industry, which reach the per cent range (figure 13 and 14).

Clearly increased proportions of iron in the fine dust fraction (< 10 µm) were found at systems of the cement industry.

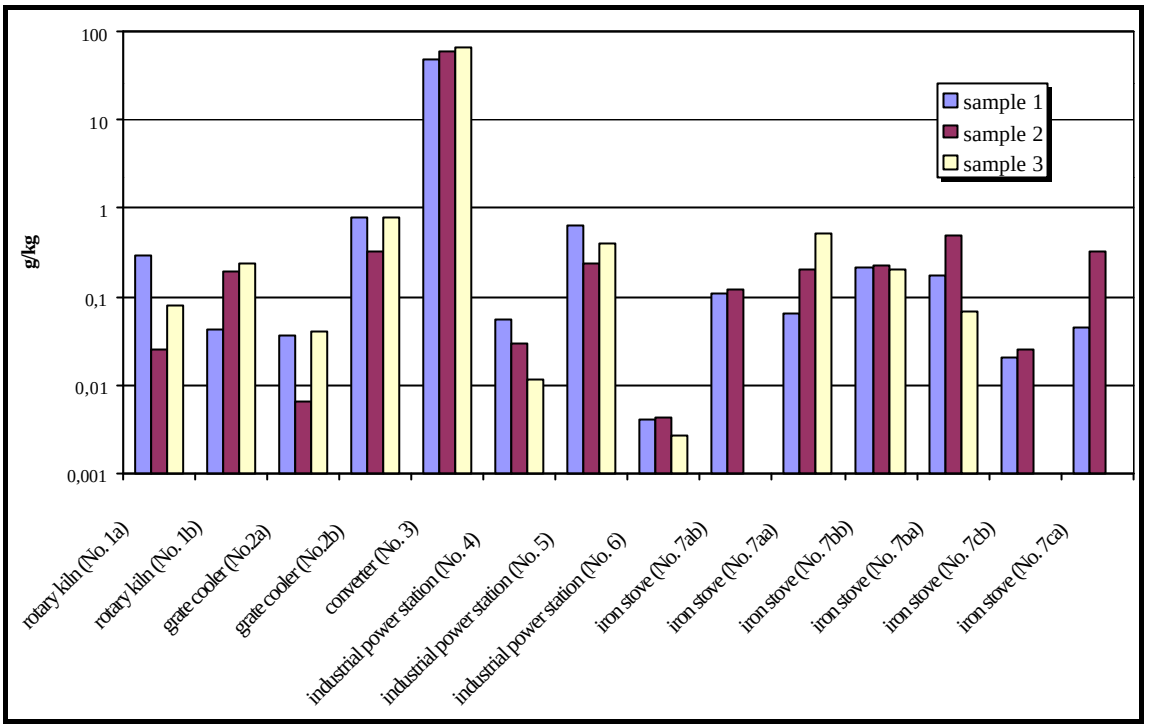

Figure 13: Lead proportion in the emitted PM 10-dust

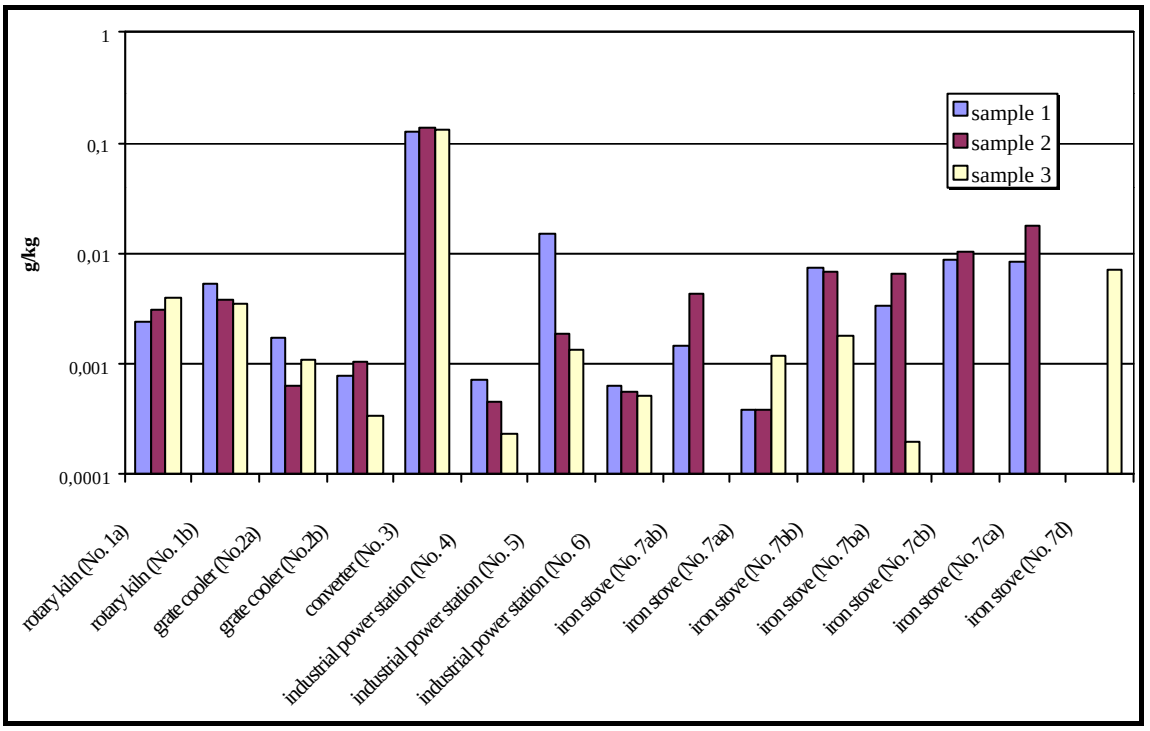

Figure 14: Cadmium proportions in the in the emitted PM 10-dust

In another diagram (figure 15) the proportions of the dust constituents related to the mass of the particles for each examined impactor stage show a considerable enrichment of heavy metals in the finer particle fractions for the grate cooler system (cf. [20]). For other systems examined so far this strong enrichment effect could not be

proven. On the contrary, reverse results were found e.g. at the examined firing installations or a similar proportion of dust constituents related to the mass of the particles for each impactor stage (cf. converter figure 15).

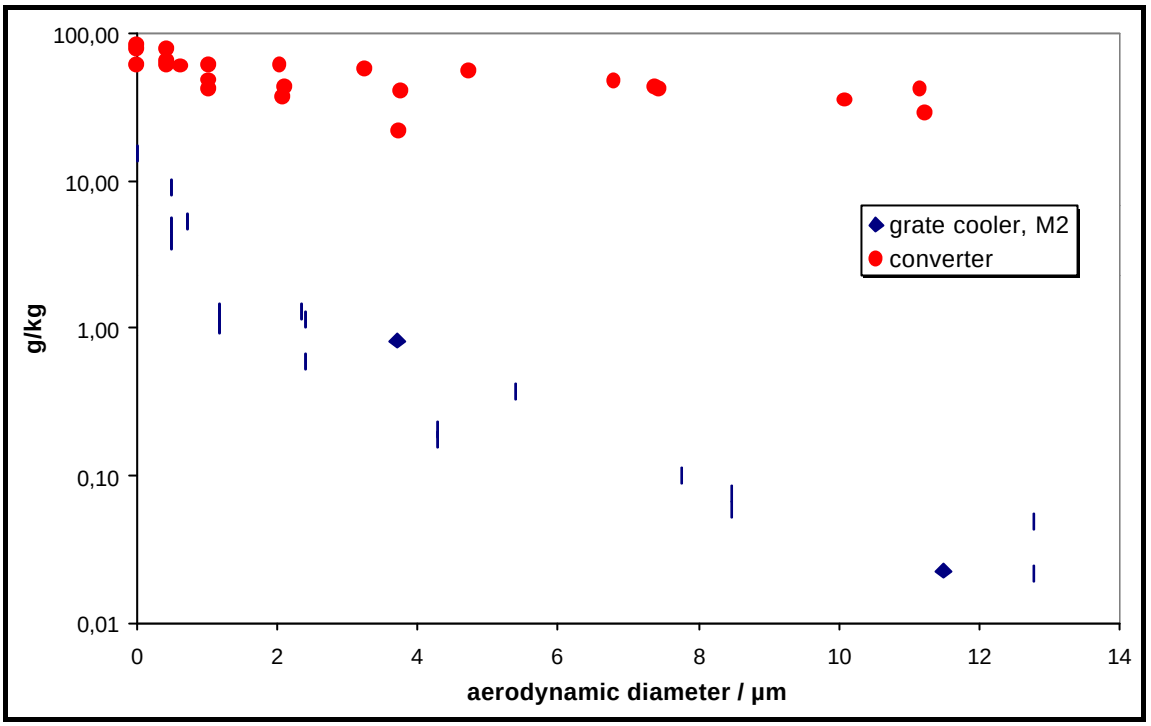

Figure 15: Proportion of lead in the emitted fine particle fractions (grate cooler and converter)

6 ACKNOWLEDGEMENT

The authors thank all involved co-workers of the Bayerisches Landesamt für Umweltschutz and the Universität Stuttgart. Also thanks to all co-workers of the Landesamt für Umweltschutz Sachsen-Anhalt who have taken part in the investigations, in particular Mr. Pohle, Mr. Maiwald, Mr. Lux and Mr. Reifert of the department "Emission and Immission Monitoring" for the sampling and for the analyses as well as Dr. Schumann, Dr. Jacob, Dr. Bärwald, Mrs. Lange and Mrs. Voigt of the department "Analytics Air".

7 REFERENCES

[1] Richtlinie 1999/30/EG des Rates vom 22. April über Grenzwerte für Schwefeldioxid, Stickstoffoxide, Partikel und Blei in der Luft. ABl. Nr. L163 v. 29.06.1999, p. 41

[2] EPA, Air Quality Criteria for Particulate Matter. National Center for Environmental Assessment. Office of Research and Development. US Environmental Protection Agency. Report No. EPA /600/P-95/001aF-cF, 1996

[3] The APHEA project. Short term effects of air pollution on health: an European approach using epidemiological time series data. J. Epidemiol. Commun. Health 50 (1996). Suppl. 1.

[4] Landesanstalt für Umweltschutz Baden-Württemberg und UMEG, Schwebstaubbelastung in Baden-Württemberg., 1998, Karlsruhe.

[5] WHO (World Health Organisation): Air Quality Guidelines for Europe. Second Edition (in printing)

[6] Bruckmann, P., Die neuen Richtlinien der Europäischen Union zur Luftqualität. Zeitschrift Immissionsschutz, Berlin, 3 (1998) pp. 100 –108

[7] Ehrlich, Chr., Noll, G., Kalkoff, W.-D.: Messtechnische Ermittlung von PM 10-und PM 2.5-Emissionen aus Industrieanlagen und Hausbrandfeuerstätten. Neuere Entwicklungen bei der Messung und Beurteilung der Luftqualität, Deutschland, Heidelberg, 27. Bis 29. April 1999. VDI-Verlag Düsseldorf, VDI-Berichte: Nr. 1443. pp. 117-129

[8] Protokolle zu den Fachgesprächen am 27.01.98 und 22.07.98 im Umweltbundesamt zum Thema ”Untersuchungen zu Emissionsquellen von Feinstaub”. Unpublished
[9] VDI 2066 Bl. 5 Messen von Partikeln. Staubmessung in strömenden Gasen. Fraktionierende Staubmessung nach dem Impaktionsverfahren - Kaskadenimpaktor
[10] VDI 2066 Bl. 1 Messen von Partikeln. Staubmessung in strömenden Gasen. Gravimetrische Bestimmung der Staubbeladung. Übersicht
[11] VDI 3868 Bl. 1 Messen der Gesamtemission von Metallen, Halbmetallen und ihren Verbindungen. Manuelle Messung in strömenden, emittierten Gasen. Probenahmesystem für partikelgebundene und filtergängige Stoffe
[12] VDI 2268 Bl. 1 Stoffbestimmung an Partikeln. Bestimmung der Elemente Ba, Ca, Cd, Co, Cr, Cu, Ni, Pb, Sr, Zn in emittierten Stäuben mittels atomspektrometrischer Methoden
[13] DIN EN 45001 Allgemeine Kriterien zum Betreiben von Prüflaboratorien
[14] Landesamt für Umweltschutz Sachsen-Anhalt Halle/Saale, Emissionsermittlungs-Berichte 01-97, 06-98, 07-98, 08-98, 10-98, 01.99, 02-99, 03-99, 01-00, 04-99, 05-00. Unpublished
[15] Bayerisches Landesamt für Umweltschutz, Messberichte des Programms „Ermittlung von Korngrößenverteilung im Abgas verschiedener Emittenten“. Unpublished
[16] Feinstaubuntersuchungen an Holzfeuerungen. Universität Stuttgart, Institut für Verfahrenstechnik und Dampfkesselwesen, Bericht Nr. 44-1999
[17] Ehrlich, Chr., Noll, G., Kalkoff, W.-D.. Emissionsuntersuchungen von PM 10, PM 2.5 und PM 1.0 an Industrieanlagen und Hausbrandfeuerstätten. Zeitschrift Immissionsschutz, Berlin, 4 (2000) pp. 141 – 148
[18] Feinstaubemissionsuntersuchungen in Sachsen-Anhalt: PM 10-, PM 2.5- und PM 1.0– Emissionen aus Industrie und Hausbrand, Berichte des Landesamtes für Umweltschutz Sachsen-Anhalt, Sonderheft 1/2001
[19] Schreiben des Forschungsinstituts der Zementindustrie Düsseldorf vom 22.09.98. Unpublished
[20] Lützke, K., Muhr, W., Erprobung von Emissionsmessverfahren zur Feststellung von Korngrößenfraktionen, UBA Forschungsbericht 81-10402121, Oktober 1981

Dr. Christian Ehrlich

Saxony-Anhalt Environmental Protection Agency
Landesamt für Umweltschutz Sachsen-Anhalt
Reideburger Str. 47
D - 06116 Halle (Saale), Germany

e-mail: ehrlich@lau.mu.lsa-net.de

After having studied physics at the Leipzig University I have been involved in environmental and air pollution issues since 1977. As Head of Department "Air Pollution Prevention and Chemical Safety" at the Saxony-Anhalt Environmental Protection Agency in Halle (Saale), Germany, I am responsible for emission inventories, air pollution management plans, air quality monitoring networks, emission measurements, noise abatement and chemical safety of the Federal German State Saxony-Anhalt.
I have been a member of the Advisory Committee of the German States on Air Quality Monitoring since 1992 and a member of the Expert Committee for Environmental Measurement Technology of the Commission on Air Pollution Prevention (KRdL) of VDI and DIN - Standards Committee since 1995.