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PM_{10}, $PM_{2.5}$ and $PM_{1.0}$—Emissions from industrial plants—Results from measurement programmes in Germany

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Abstract

Emission measurement programmes were carried out at industrial plants in several regions of Germany to determine the fine dust in the waste gases; the PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ fractions were sampled using a cascade impactor technique. The installations tested included plants used for: combustion (brown coal, heavy fuel oil, wood), cement production, glass production, asphalt mixing, and processing plants for natural stones and sand, ceramics, metallurgy, chemical production, spray painting, wood processing/chip drying, poultry farming and waste treatment. In addition waste gas samples were taken from small-scale combustion units, like domestic stoves, firing lignite briquettes or wood.

In total 303 individual measurement results were obtained during 106 different measurement campaigns. In the study it was found that in more than 70% of the individual emission measurement results from industrial plants and domestic stoves the PM_{10} portion amounted to more than 90% and the $PM_{2.5}$ portion between 50% and 90% of the total PM (particulate matter) emission. For thermal industrial processes the $PM_{1.0}$ portion constituted between 20% and 60% of the total PM emission.

Typical particle size distributions for different processes were presented as cumulative frequency distributions and as frequency distributions. The particle size distributions determined for the different plant types show interesting similarities and differences depending on whether the processes are thermal, mechanical, chemical or mixed. Consequently, for the groups of plant investigated, a major finding of this study has been that the particle size distribution is a characteristic of the industrial process. Attempts to correlate particle size distributions of different plants to different gas cleaning technologies did not lead to usable results.

Keywords: PM_{10}; $PM_{2.5}$; $PM_{1.0}$; Stationary source emission; Emission measurement; Fine particles; Cascade impactor; Particle size distribution

1. Introduction

The health impact of fine dust has been evaluated at international and EU level (Council Directive, 1999; The APHEA Project, 1996; WHO, 2006; Second Position Paper on Particulate Matter, 2004).

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According to recent epidemiological investigations fine and ultrafine dust particles in the ambient air adversely affect human health so the European commission adopted a directive (Council Directive, 1999) limiting the mass concentration of PM_{10} in the ambient air; a proposed new European Directive seeks to limit the $PM_{2.5}$ mass concentration in ambient air. It is expected, however, that at present, strict limit values cannot be met in all areas of Germany and Europe (Second Position Paper on Particulate Matter, 2004) and therefore further emission reduction measures will be required. For the implementation of such air pollution abatement measures better data on fine dust emissions are necessary than currently available. The past legislation had required only the control of total PM emissions and so our knowledge of fine dust emissions is poor.

The results from a number of specific measurement programmes being conducted by the German Federal Environmental Agency and the German states were intended to improve the knowledge of fine dust emissions from different industrial and other sources. The presented study is based on results of measurement programmes in Saxony-Anhalt (Landesamt für Umweltschutz Sachsen-Anhalt, 2001), Bavaria (Bayerisches Landesamt für Umweltschutz, 2000), Saxonia (Sächsisches Landesamt für Umwelt und Geologie, 1999) and the Federal Environmental Agency (Baumbach et al., 1999; Dreiseidler et al., 2001). These programmes included measurements at industrial plants where total dust emissions are relevant or contain toxic materials such as heavy metals. Thus, for example, investigations were carried out at combustion plants (brown coal, heavy fuel oil, wood), cement production plants, glass production plants, asphalt mixing plants, processing plants for natural stones and sand, ceramic industry plants, metallurgy, chemical plants, spray painting plants, wood processing/chip drying, poultry farming and waste treatment plants. In total, 106 measurement campaigns were undertaken and 303 individual measurements were made. The first results have already been presented (Baumbach et al., 1999; Ehrlich et al., 1999, 2000; Bayerisches Landesamt für Umweltschutz, 2000; Dreiseidler et al., 2001). PM measurements were also carried out at a small stove burning different kinds of fuel briquettes; since burning brown coal briquettes on household fireplaces remains common practice in the new German federal states and in the new EU Member States like Czech Republic, Poland and others. Small wood-burning furnaces were also investigated.

2. Experimental investigations

2.1. General

Aerodynamic diameter d_{ae}, used to represent particle dispersion in gases, is defined as the diameter of a particle with the same sedimentation velocity in gases as spheres from material of the density $1\,g\,cm^{-3}$.

The reference method for sampling and measurement of PM_{10} concentration (as well as a provisional procedure for sampling and measurement of the $PM_{2.5}$ concentration) in ambient air is laid down (Council Directive, 1999/30/EC). The method is based on the inertial separation of the PM_{10} particle fraction and its subsequent gravimetric determination. In this study, PM_{10} (and $PM_{2.5}$) dust emission fractionation was carried out using cascade impactors, i.e. the same separation principle as used in the ambient air method. Cascade impactors are relatively simple to use for in situ sampling and avoids agglomeration effects that might distort the particle size distribution. Impactors fractionate suspended dust particles into different size categories according to their inertia. The basic components of an impactor are a nozzle and an impaction plate. When accelerated in the nozzle those particles having sufficient inertia strike the impaction plate and are collected. A cascade impactor contains several impactor stages and particles are segregated as a function of decreasing inertia, so that samples of decreasing aerodynamic particle size are obtained stage by stage. The particles not retained by any of the impactor stages are collected in a backup filter downstream of the impactor stages. The particle mass collected is determined by weighing each stage before and after sampling; the material collected can be used, when required, for further analyses e.g. on dust composition. A pre-separator is necessary if there is a large proportion of coarse particles $> PM_{10}$ (Lützke and Muhr, 1981).

2.2. Sampling techniques

The measurement campaigns used eight- or six-stage Anderson Impactors, type Mark III (material: stainless steel) and/or a six-stage Stroehlein type STF 1 impactor (material: Titanium). Preliminary investigations showed that under similar sampling

conditions both impactor types showed comparable particle size distributions in a range from 0.6 to 16 μm for the various processes tested. Both impactors are heat resistant up to 850 °C. Perforated collecting plates and glass–fibre backup filters were used for the particle separation. The sample gas flow suction rate was determined using thermal mass flow meters.

2.3. Sampling and measurement

The sampling and measurement was carried out in accordance with VDI 2066 Bl. 5 (1994). In contrast to the total dust emission measurements, however, the gas flow through an impactor must be kept constant at all points of the sampling grid to provide constant separation conditions in the impactor. The volumetric flow rate only depends on the waste gas conditions and can be calculated with special computer programs. Hence one measurement can only cover sampling points with similar gas velocity (maximum acceptable deviation is $\pm 30\%$), if the flow is not that homogeneous several measuring operations are required. The sampling period should be chosen such that sufficient PM mass is collected on each impactor stage to permit weighing with the required accuracy without overloading the stages.

First, the existing emission concentrations of total dust (according to EN 13284-1 (2001)) had been determined in order to be able to specify the required sampling times and suction rates for the impactor samplings. Thus, on the one hand overloading of the impactor stages was avoided and on the other hand sufficient PM masses could be ensured to allow weighing with adequate accuracy. Since, for the industrial plants examined, the total PM concentrations were usually low at the tested industrial plants, very long sampling times (up to 18 h and in extreme cases 52 h) had to be provided to obtain an adequate filter loading. By contrast, the high PM concentrations arising from domestic fire places required sampling times that were substantially shorter than the time taken for the complete burn cycle of the batch of fuel used. Investigations proved, however, that most of the particle load was emitted in the first third of the combustion cycle and so the impactor measurements were carried out at this time of higher emission.

The operating conditions of each installation were determined and documented during the sampling periods. All measurements took place during the process was operated normally. For each measurement campaign the flue gas conditions (waste gas velocity, static pressure, temperature and water vapour content) were determined.

2.4. Quality assurance

All measurements were performed according to the quality assurance measures specified in EN ISO/IEC 17025 (2005). Thus, all pollutant emissions were measured with standardized measurement methods (VDI guidelines, DIN, EN) and officially approved measurement instruments were used for the determination of gaseous components. All plant operating and measured parameters were carefully documented using standardized report guidelines.

3. Results

Tables 1–4 list the plants investigated, the performance characteristics pertaining during the sampling periods, the fuels used, and descriptions of the gas cleaning equipment and final results of PM percentage and total dust content (Landesamt für Umweltschutz Sachsen-Anhalt, unpublished; Bayerisches Landesamt für Umweltschutz, 2000; Baumbach et al., 1999; Ehrlich et al., 2000; Landesamt für Umweltschutz, 2001; Sächsisches Landesamt für Umwelt und Geologie, 1999; Dreiseidler et al., 2001).

4. Discussion

4.1. General

In the framework of this article it is not possible, to present all results from 106 emission measurement campaigns (see Tables 1–4) as particle size distributions in diagrams, therefore in the following section typical particle size distributions of emission samples were selected, presented and discussed. Nevertheless all obtained results were presented and assessed in Section 4.7 (Figs. 8–11).

It is common practice to plot size distribution data in such a way that a straight line results. This assists in more detail data analysis, e.g. for reading of essential parameters like PM_{10}, $PM_{2.5}$ or $PM_{1.0}$. This can be done if the distribution fits a standard law. In this study the best fit with the experimental data was gained using the cumulative frequency particle size distribution (D) according to Rosin,

Table 1
Measurement results for combustion plants

No.	Plant, maximum performance, fuel or basic input material	Samplings	Performance during measurement period (as thermal input)	Dust separation	Total dust in $mg\,m^{-3}$	Mean value		
						PM_{10} in %	$PM_{2.5}$ in %	$PM_{1.0}$ in %
Combustion plant/coal								
1	Power plant, 180 MW, dry brown coal	3	180 MW	ESP, scrubber	4.3	90.7	75.5	52.0
2	Power plant, 146 MW, bcb limestone	3	114 MW	ESP, FGD	4.6	92.0	74.0	47.7
3	Fluidized bed combustion, 119 MW, brown coal	3	114 MW	ESP, FGD, NO_x	14.4	97.0	65.7	25.5
4	Power plant, 1000 MW, pulverized hard coal	2	1000 MW	ESP, FGD, NO_x	0.9	88.4	69.2	51.3
5	Power plant, 1000 MW, pulverized hard coal	2	500 MW	ESP, FGD, NO_x	0.7	91.3	71.6	50.0
Combustion plant/heavy oil								
6	Combustion, 10 MW, heavy oil	3	5 MW	Additive	44.4	98.0	81.9	64.3
7	Combustion, 10 MW, heavy oil	3	8.5 MW	Additive	52.3	91.3	64.7	49.9
8	Combustion, 10 MW, heavy oil, urea	3	5 MW	Additive, SNCR	56.7	97.1	77.5	55.8
9	Combustion, 10 MW, heavy oil, urea	3	8.5 MW	Additive, SNCR	70.3	93.5	68.0	50.7
10	Combustion, 20 $t\,h^{-1}$ steam, heavy oil	3	9.7 $t\,h^{-1}$ steam (output)	SNCR	9.2	92.5	68.8	49.1
11	Combustion, 270 MW, heavy oil	3	270 MW	NO_x	45.8	87.1	71.1	63.2
12	Combustion, 270 MW, heavy oil	2	270 MW	Additive, NO_x	33.4	93.4	86.3	76.5
13	Combustion, 270 MW, heavy oil	2	200 MW	NO_x	26.8	96.5	88.9	76.3
14	Combustion, 270 MW, heavy oil	2	200 MW	Additive, NO_x	26.2	95.9	87.1	69.2
Combustion plant/wood								
15	Grate firing, 1.4 MW, saw chips, saw dust	1	0.9 MW	CC	194.0	98.6	70.4	49.0
16	Grate firing, 1.4 MW, saw chips, saw dust	1	1.3 MW	CC	172.0	98.3	67.7	44.8
17	Grate firing, 0.8 MW, saw chips, saw dust	1	0.8 MW	CC	251.0	98.3	62.6	36.0
18	Grate firing, 3 MW, hogged wood	1	1.3 MW	CC	184.0	98.0	91.7	85.2
19	Underfeed stoker, 2.3 MW, rest of chipboards	3	1.3 MW	Multi-CC	94.3	95.1	73.4	58.7
20	Grate firing, 1.1 MW, piece of wood, saw chips	1	1.0 MW	CC	123.0	89.8	55.0	43.2
21	Grate firing, 2 MW, hogged wood, wood waste	1	1.5 MW	ESP	27.0	89.2	67.1	62.4
22	Grate firing, 7.9-9.5 MW, wood, wood chips	1	5.8 MW	ESP	23.0	73.5	53.6	46.3
23	Grate firing, 7.9-9.5 MW, natural gas, wood, chips	1	8.6 MW	ESP	27.0	80.9	56.9	46.1
24	Grate firing, 15 MW, hogged and rest wood, chips	1	15 MW	ESP	7.0	87.1	52.7	33.8
25	Grate firing, 1.5 MW, hogged wood	1	0.5 MW	CGC, multi-CC	24.0	99.8	99.5	95.6
26	Grate firing, 1.5 MW, hogged wood	1	0.9 MW	CGC, multi-CC	31.0	100.0	97.2	92.7
27	Power station, 31 $t\,h^{-1}$ steam, matured wood	3	27 $t\,h^{-1}$ steam (output)	CC, FF, NO_x	19.9	80.2	31.4	13.8

Table 2
Measurement results for cement, ceramic, glass industry

No.	Plant, maximum performance, fuel or basic input material	Samplings	Performance during measurement period	Dust separation	Total dust in $mg\,m^{-3}$	Mean value		
						PM_{10} in %	$PM_{2.5}$ in %	$PM_{1.0}$ in %
Cement/furnaces								
28	Rotary kiln, brown coal dust, waste oil	3	$87\,t\,h^{-1}$ clinker (do)	ESP horizontal	8.2	96.7	82.3	52.6
29	Rotary kiln, brown coal dust, waste oil	2	$85\,t\,h^{-1}$ clinker (co)	ESP horizontal	n.m.	96.2	69.5	39.0
30	Lepol-kiln, raw meal, coal, waste oil, used tyre	6	$41\,t\,h^{-1}$ clinker (do)	ESP	15.1	92.4	50.1	39.2
31	Heat exchanger, raw meal, heavy oil, used tyre	6	$118\,t\,h^{-1}$ clinker (do)	ESP	2.3	99.4	75.2	42.9
32	Heat exchanger, raw meal, heavy oil, used-tyre	6	$118\,t\,h^{-1}$ clinker (co)	ESP	4.8	100.0	62.1	25.0
33	Rotary kiln, brown coal dust, raw meal	2	$334\,t\,h^{-1}$ clinker (do)	ESP	3.5	95.5	78.2	41.6
34	Rotary kiln, brown coal dust, raw meal (1 mill)	4	$325\,t\,h^{-1}$ clinker (co)	ESP	7.1	89.9	56.4	25.4
35	Rotary kiln, brown coal dust, raw meal (2 mills)	2	$287\,t\,h^{-1}$ clinker (co)	ESP	12.9	90.9	49.2	24.5
Cement/cooler fabric filter								
36	Grate cooler, clinker	3	$72\,t\,h^{-1}$ clinker	FF	3.4	43.3	3.8	1.2
37	Gate cooler, clinker	3	$72\,t\,h^{-1}$ clinker	FF	21.1	23.6	2.6	0.6
Cement/cooler ESP, CC								
38	Clinker cooler, raw meal, coal, waste oil, used tyre	6	$41\,t\,h^{-1}$ clinker (co)	ESP, multi-CC, CC	15.3	98.0	64.5	23.2
Glass industry/bath								
39	Manufacture batch glass, cullet, batch, natural gas	6	$223\,t\,d^{-1}$	Lime-sorption, ESP	3.0	95.3	53.5	21.2
40	Manufacture of flat glass, cullet, batch, natural gas	6	$508\,t\,d^{-1}$	FGD (lime), ESP	5.1	93.2	44.8	23.7
41	Manufacture of goblets and beakers, bath, cullet, batch, natural gas	3	$46\,t\,d^{-1}$	FF	0.8	93.4	53.3	37.7
Glass industry /treatment								
42	Manufacture of goblets and beakers, treatment, cullet, batch	2	$46\,t\,d^{-1}$	FF	0.2	83.9	49.5	41.7
Asphalt mix plant								
43	Asphalt mix plant, asphalt granulate, oil	6	$150\,t\,h^{-1}$	FF	19.0	93.1	29.2	8.3
Manufacture of porcelain/press								
44	Isostatic compression press, porcelain substance	4	$400\,piece\,h^{-1}$	FF	0.1	94.9	57.4	38.3
Treatment natural stone, sand								
45	Crusher plant, lime stone, dolomite	3	$600\,t\,h^{-1}$	FF	1.2	69.2	14.2	5.0
46	Screening plant, lime stone, dolomite	3	$225\,t\,h^{-1}$	FF	10.9	59.2	5.9	1.2
47	Sand conditioning, natural sand	3	$30\,t\,h^{-1}$	FF	6.2	68.7	11.9	2.9
48	Sand drying, cinder sand, heavy oil	1	$30\,t\,h^{-1}$	FF	28.3	87.3	18.9	6.0
49	Preparation of ceramic raw materials, loam, clay, porosity material	3	Full load	FF	0.8	80.4	34.4	16.5
Tunnel oven ceramic industry								
50	Oven (without additive), loam, clay, gas	1	Full load		5.3	93.9	85.0	79.7
51	Oven (with additive), loam, clay, gas, lime	3	Full load		3.4	95.4	88.6	84.9

Table 3
Measurement results for metallurgy, chemical plants, spray painting and others

No.	Plant, maximum performance, fuel or basic input material	Samplings	Performance during measurement period	Dust separation	Total dust in $mg\,m^{-3}$	Mean value		
						PM_{10} in %	$PM_{2.5}$ in %	$PM_{1.0}$ in %
Metallurgy/melt								
52	Converter, brass scrap, copper dross, coke…	3	$59\,t\,charge^{-1}$	FF	2.4	96.3	76.3	51.3
53	Aluminium-remelt heat, natural gas, al-scrap	2	Without information	Reactor (lime), FF	0.2	98.5	72.0	35.8
54	Cupola, iron-scrap, coke, limestone	4	$6.4\,t\,h^{-1}$	FF	7.1	87.9	43.8	19.8
55	Induction furnace, iron casting plant, iron-scrap	4	$2.4\,t\,h^{-1}$	FF	0.4	77.4	48.6	18.1
56	Cupola, iron-scrap, coke	6	$15\,t\,h^{-1}$	CC, scrubber,	58.3	95.4	88.1	72.4
57	Electric arc furnace, iron/steels scrap, aggregates	3	$100\,t\,h^{-1}$	FF	4.6	96.1	58.1	24.6
58	Cupola, iron/steels scrap, coke…	1	$16\,t\,d^{-1}$	FF	0.2	96.2	75.8	64.4
Metallurgy /drying, cooling								
59	Drying plant, aluminium chips	2	$1.8\,t\,h^{-1}$	Afterburning, FF	1.0	95.4	46.1	20.3
60	Drying plant, aluminium-chips	2	$1.5\,t\,h^{-1}$	Afterburning, FF	0.4	98.4	52.9	32.8
61	Tunnel to cooling castings, castings, water	2	No information	No information	n.m.	94.6	42.7	23.2
62	Rotary screen, casting transport, castings	6	150 t/month	FF	25.4	81.8	28.4	9.5
Metallurgy, treatment								
63	Sand conditioning, pit-iron sand, betonite, coal dust	6	$27–30\,t\,h^{-1}$	ESP	21.4	74.0	15.5	1.4
64	Sand conditioning wet casting, sand, betonite	2	$20\,t\,h^{-1}$	FF	0.6	86.8	36.2	21.2
65	Treatment, moulding sand, consumable material	2		FF	4.1	91.9	39.7	20.6
Chemical plants/drying, furnace								
66	Flow dryer A fabrication of acrylonitrile fibre, acrylonitrile	4	$800\,kg\,h^{-1}$	CC, water injection	16.9	91.4	62.1	50.9
67	Flow dryer B fabrication of acrylonitrile fibre, acrylonitrile	6	$800\,kg\,h^{-1}$	CC, water injection	8.9	83.9	48.6	17.4
68	Fertilizer dryer, fertilizer, heavy oil, carbon-monoxide	6	$450\,t\,d^{-1}$	FF, packed scrubber	13.3	96.3	33.4	n.m.
69	Spray dryer for drying resin, liquid condensation resins	6	$0.65\,m^3\,h^{-1}$	FF	2.6	78.3	15.3	3.3

Table 3 (*continued*)

No.	Plant, maximum performance, fuel or basic input material	Samplings	Performance during measurement period	Dust separation	Total dust in $mg\,m^{-3}$	Mean value		
						PM_{10} in %	$PM_{2.5}$ in %	$PM_{1.0}$ in %
70	Rotary dryer (bentonite), natural gas, podsol	3	$2\,t\,h^{-1}$	FF	4.7	76.7	32.1	11.1
71	Run around dryer (bentonite), natural gas, podsol	3	$3\,t\,h^{-1}$	CC, ESP	79.8	96.7	63.9	21.3
72	Milling dryer (bentonite), natural gas, bentonite, soda	6	$9\,t\,h^{-1}$	ESP	3.3	95.2	49.9	11.8
73	Low-shaft electric arc furnace (manufacture of silicium), coal, charcoal, coke, chips	6	$3.2\,t\,h^{-1}$	FF	1.2	92.8	45.4	24.9
Chemical installations								
74	Urea prill processing, urea	2	$45\,t\,h^{-1}$		80.0	97.0	68.5	15.5
75	Urea prill processing, urea	4	$25\,t\,h^{-1}$		45.0	91.7	61.4	38.3
76	Fluid catalytic cracker carbon-monoxide, vacuum gas oil	6	$160\,m^3\,h^{-1}$	Multi-CC, ESP	21.0	97.1	71.4	26.4
77	Fluid catalytic cracker, wax distillate, refinery residuals	6	$100\,t\,h^{-1}$	ESP	2.7	90.2	57.7	19.3
Spray painting								
78	Spray painting of automobiles, organic solvents	3	About $400\,kg\,d^{-1}$	Venturi-washer	0.6	97.6	82.8	30.6
Wood working/shaving dryer								
79	Rotating drum dryer, shavings	4	58 MW	CC, FF	28.7	90.9	66.7	52.3
80	Chipping dryer, shavings, gas or oil	2	Full load	ESP	2.1	99.2	92.8	87.5
Poultry farming								
81	Floor keeping, fryer	6	14500 animals		6.2	60.6	8.2	1.0
82	Battery keeping, laying hen	4	15700 animals		3.1	36.5	2.7	0.4
Utilization of waste material								
83	Old growth shredder, lumber, wood, timber waste	4	$30\,t\,h^{-1}$	FF	4.5	92.6	16.7	11.5

Table 4
Measurement results for small-scale firing units

No.	Plant, maximum performance, fuel or basic input material	Samplings	Performance during measurement period (as thermal input)	Dust separation	Total dust in $mg\,m^{-3}$	Mean value		
						PM_{10} in %	$PM_{2.5}$ in %	$PM_{1.0}$ in %
Small-scale firing units/coal								
84	Household stove, 6 kW, LAUBAG-bcb (after cooling)	3	6 kW	Without	20.5	93.4	85.4	76.6
85	Household stove, 6 kW, LAUBAG-bcb (before cooling)	2	6 kW	Without	13.2	91.6	84.1	75.7
86	Household stove, 6 kW, MIBRAG-bcb (after cooling)	3	6 kW	Without	56.4	95.9	83.5	63.8
87	Household stove, 6 kW, MIBRAG-bcb (before cooling)	3	6 kW	Without	80.6	90.5	80.5	63.1
88	Household stove, 6 kW, Polish bcb (after cooling)	2	6 kW	Without	n.m.	95.8	80.8	65.0
89	Household stove, 6 kW, Polish bcb (before cooling)	2	6 kW	Without	66.2	93.5	77.0	63.3
90	Household stove, 6 kW, Bashkirian bcb	3	6 kW	Without	147.8	91.3	80.8	70.6
91	Household stove, 6 kW, MIBRAG-bcb	3	6 kW	Without	n.m.	94.0	85.6	75.4
Small-scale firing units/wood								
92	Firing plant, 175 kW, chips	1	177 kW	CC	24.3	93.5	84.4	80.0
93	Firing plant, 175 kW, chip board	1	139 kW	CC	75.1	98.5	86.2	79.8
94	Firing plant, 150 kW, chips	1	148 kW	Without	23.2	95.1	72.1	66.9
95	Firing plant, 150 kW, chips	1	43.4 kW	Without	22.3	99.6	93.8	86.9
96	Firing plant, 150 kW, joinery residues	1	133 kW	Without	119.9	74.2	57.8	52.7
97	Firing plant, 150 kW, coloured pencil residues	1	112.5 kW	Without	33.7	71.3	43.7	39.0
98	Firing plant, 450 kW, log wood	1	416.5 kW	Multi-CC	56.7	100	96.5	89.0
99	Firing plant, 450 kW, log wood	1	273 kW	Multi-CC	55.1	98.0	79.7	63.1
100	Small-scale firing unit, 9 kW, log wood beech	1	9.4 kW	Without	78.0	98.9	95.8	92.8
101	Small-scale firing unit, 9 kW, log wood beech	1	7.5 kW	Without	54.0	98.2	90.2	70.9
102	Small-scale firing unit, 9 kW, log wood pine	1	8.2 kW	Without	47.0	98.9	95.2	91.8
103	Small-scale firing unit, 9 kW, log wood pine	1	6.8 kW	Without	85.0	99.2	97.6	94.1
104	Chimney stove, 6 kW, log wood beech	1	5.7 kW	Without	98.0	99.7	98.4	87.3
105	Chimney stove, 6 kW, log wood beech	1	4.1 kW	Without	104.0	97.8	95.5	86.6
106	Pellet stove, 8.5 kW, wood pellets	1	8.0 kW	Without	17.0	99.0	95.3	92.9

do = direct operation, co = compound operation, bcb = brown coal briquette, SNCR = selective non-catalytic reduction, ESP = electrostatic precipitator.
n.m. = no measurement, FGD = flue-gas desulphurization, CGC = chimney gas condensation, FF = fabric filter, CC = cyclone, NO_x = NO_x removal.

Rammler, Sperling and Bennet (RRSB) (Batel, 1964).

The RRSB function is well suited for particle distributions arising from comminution processes, a common basic processes in a variety of the plants investigated in this study. Another illustration method is the depiction of the frequency distribution (Y) over the PM diameter, which has been used too: $Y = -\mathrm{d}R/\mathrm{d}(\log d)$ (R is the cumulative residue distribution, $R = 1–D$) (Batel, 1964).

4.2. Coal-fired thermal power plants

Fig. 1(a) and (b) show the particle size distributions of the emissions from various types of pulverized coal-fired thermal power plants. In

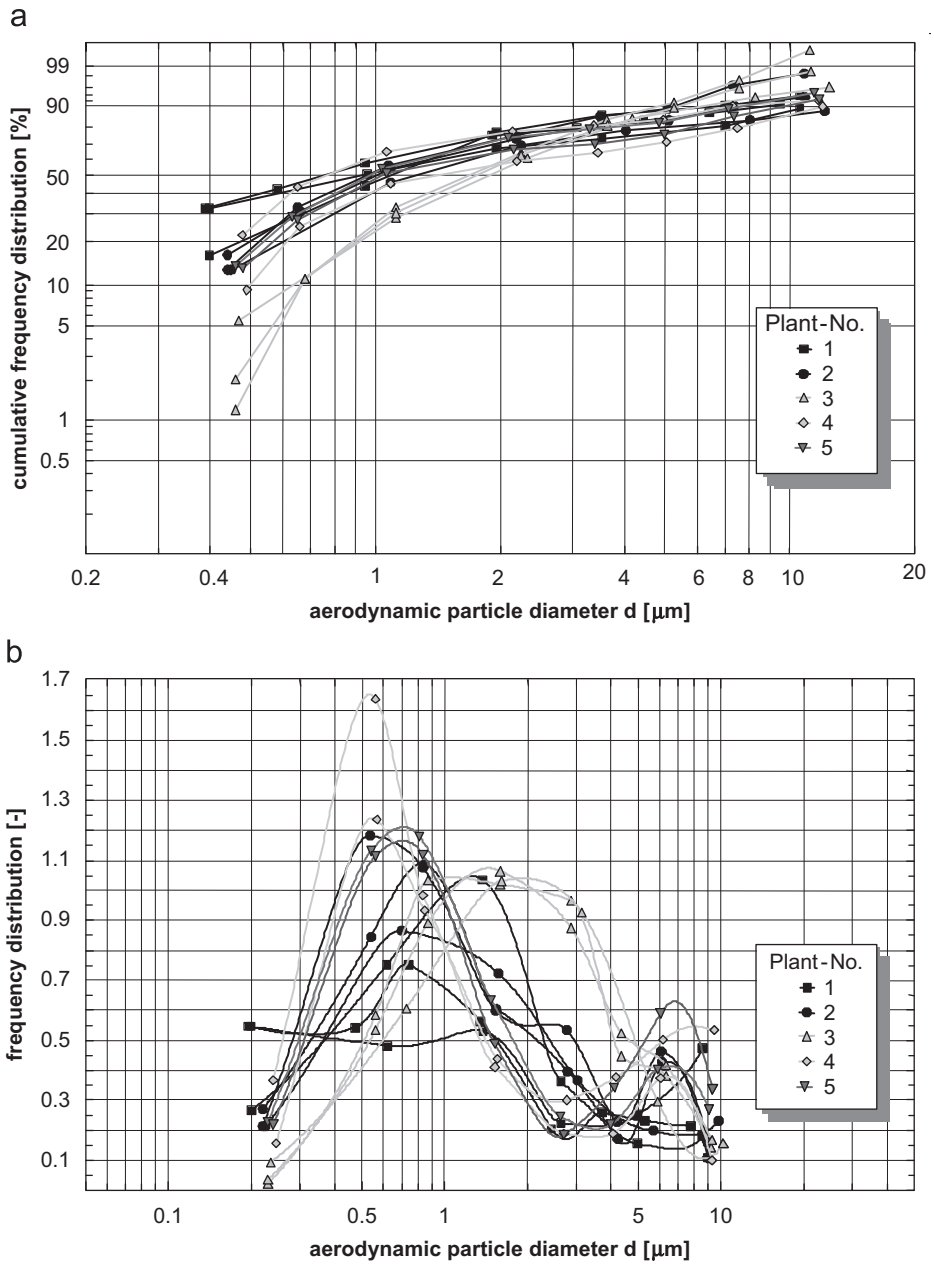

Fig. 1. (a) PM size distributions of thermal power plants (brown coal firing), RRSB cumulative frequency distribution; plant numbers according to Table 1. (b) PM size distributions of thermal power plants (brown coal firing), frequency distribution; plant numbers according to Table 1.

general a bi-modal particle size distribution appears in the combustion processes (Szpila et al., 2003). The density distribution in the Fig. 1(b) shows two modes, one in the submicrometer range (0.5–0.8 μm) and the other in the coarse range (5–10 μm).

In the flue gases of the fluidized bed combustion (plant No. 3) the maximum of the fine portion is shifted to around 2 μm (compared to 0.5–0.8 μm for the other coal combustion processes), the second maximum occurs around 6 μm for the fluidized bed combustion. The shift of the fine-grained mode to a more coarser grain is probably related to an interaction of the bed material with the ash components (Szpila et al., 2003).

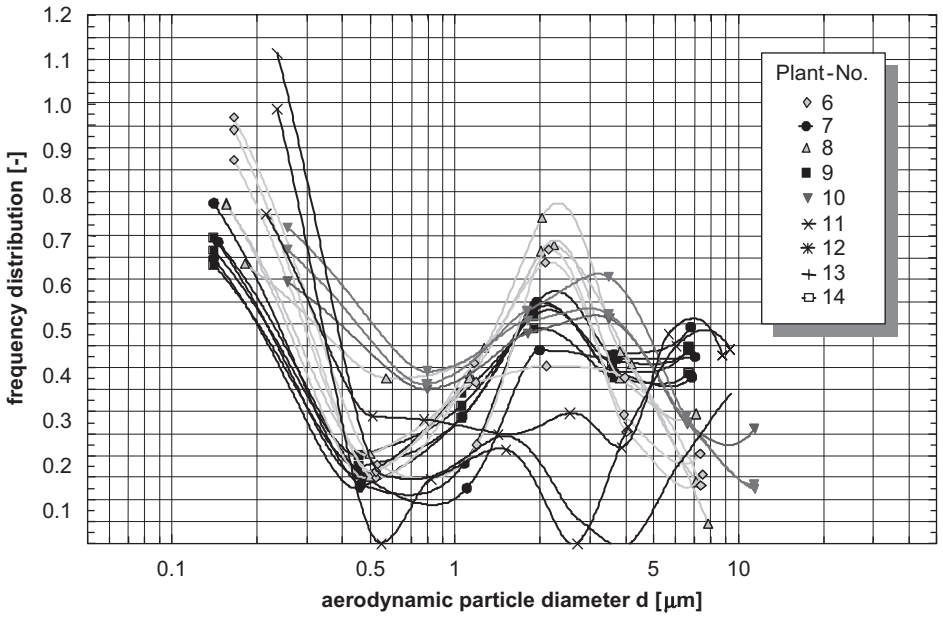

Fig. 2. PM size distribution in flue gases of heavy oil firings, frequency distribution; plant numbers according to Table 1.

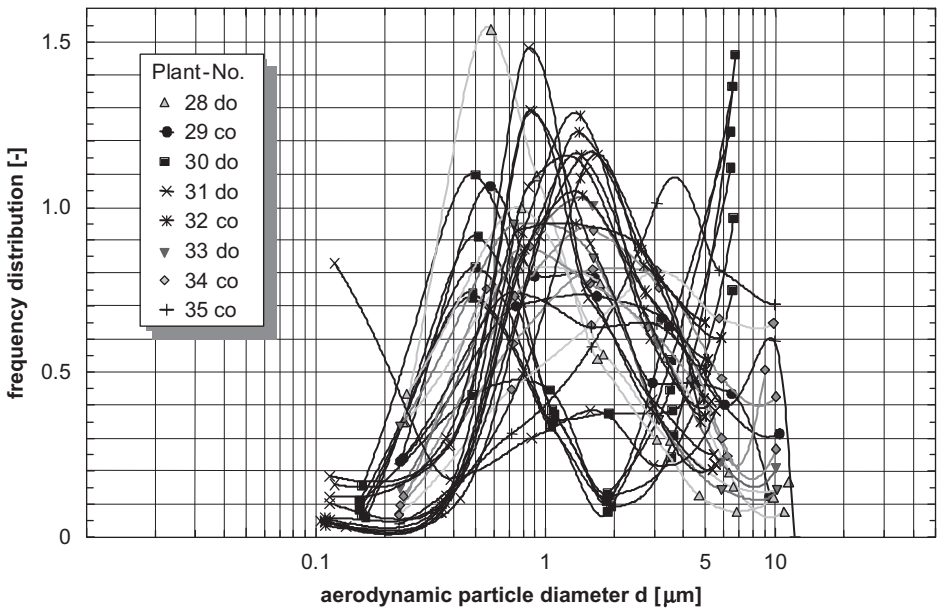

Fig. 3. PM size distribution in flue gases of cement factories, frequency distribution; plant numbers according to Table 2.

4.3. Heavy fuel oil firings

The heavy fuel oil-fired combustion plants investigated (Fig. 2) also show a bi-modal particle size distribution. Here, the fine mode is strongly shifted to the ultrafine range (smaller than 0.1 μm) and so it is not completely covered by the measurement method; the coarser mode lies here within the range of 2–3 μm.

4.4. Cement factories

Fig. 3 represents the particle size distributions in the clean gas of cement kilns. The large to very large

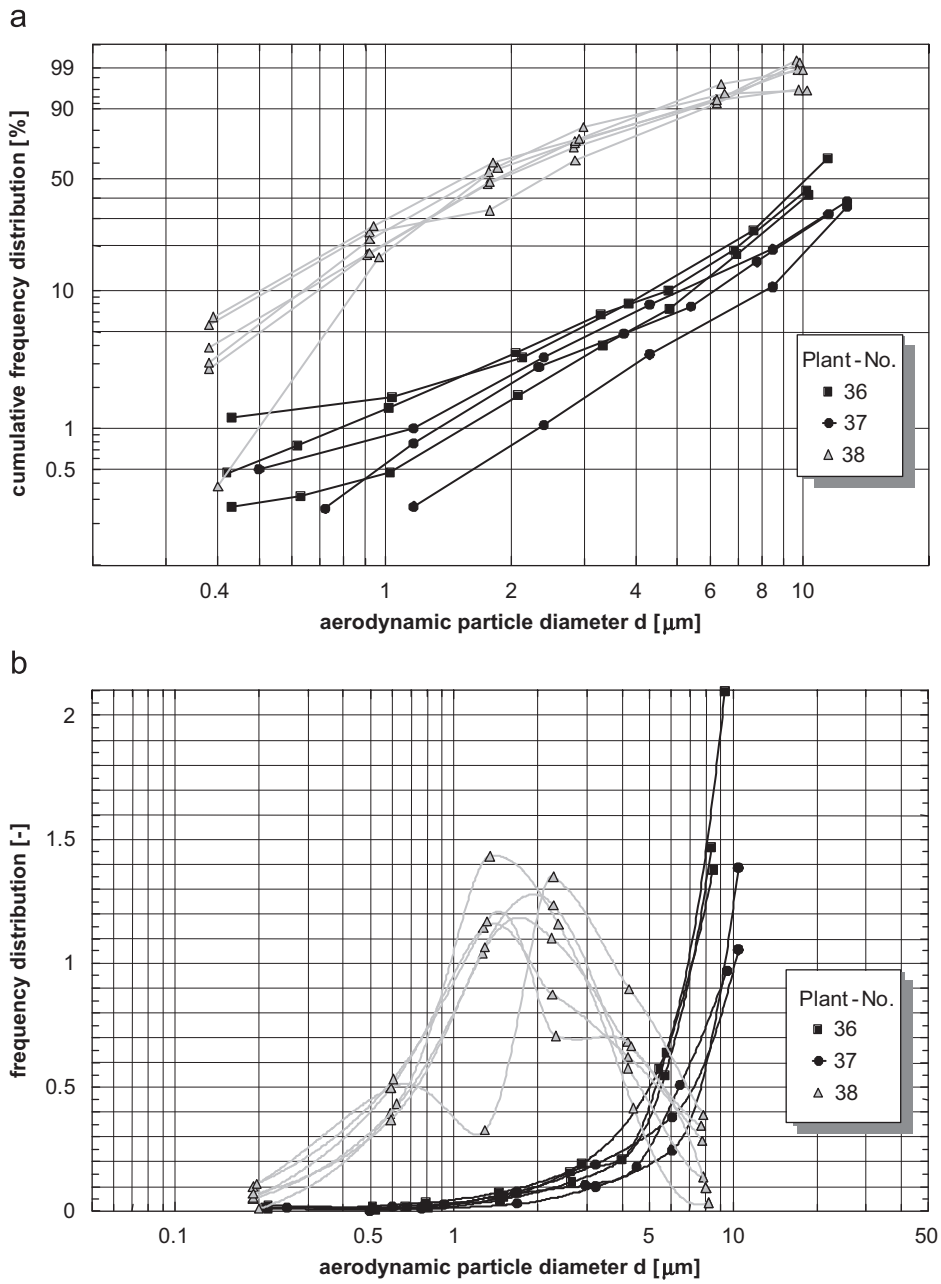

Fig. 4. (a) PM size distributions of cement clinker cooler plants, RRSB cumulative frequency distribution; plant numbers according to Table 2 (plant Nos. 36 and 37: unexpected but proved results). (b) PM size distribution in flue gases of cement clinker cooler plants, frequency distribution; plant numbers according to Table 2 (plant Nos. 36 and 37: unexpected but proved results).

portion (<3 μm) in the flue gas of the two rotary kilns (plant Nos. 28, 30, 31, 33) in direct (do) or compound (co) operation is clearly to be seen. Moreover a little coarser particle size distribution can be recognized in the compound operation (plant Nos. 29, 32, 34, 35). The bi-modal distribution typical for thermal processes, is again well identifiable. The compound operation means that portions of particles from mechanical processes (e.g. raw mills) are contained in the flue gas and thus the fine mode of 0.5 μm also shifts to 1.5–2 μm.

Measurements show (Fig. 4(a) and (b)) that while emitting lower concentrations of total PM (see Table 2), cement plants with grate cooler plants and baghouse filters (plant Nos. 36 and 37) have a more pronounced coarse particle size fraction than that of

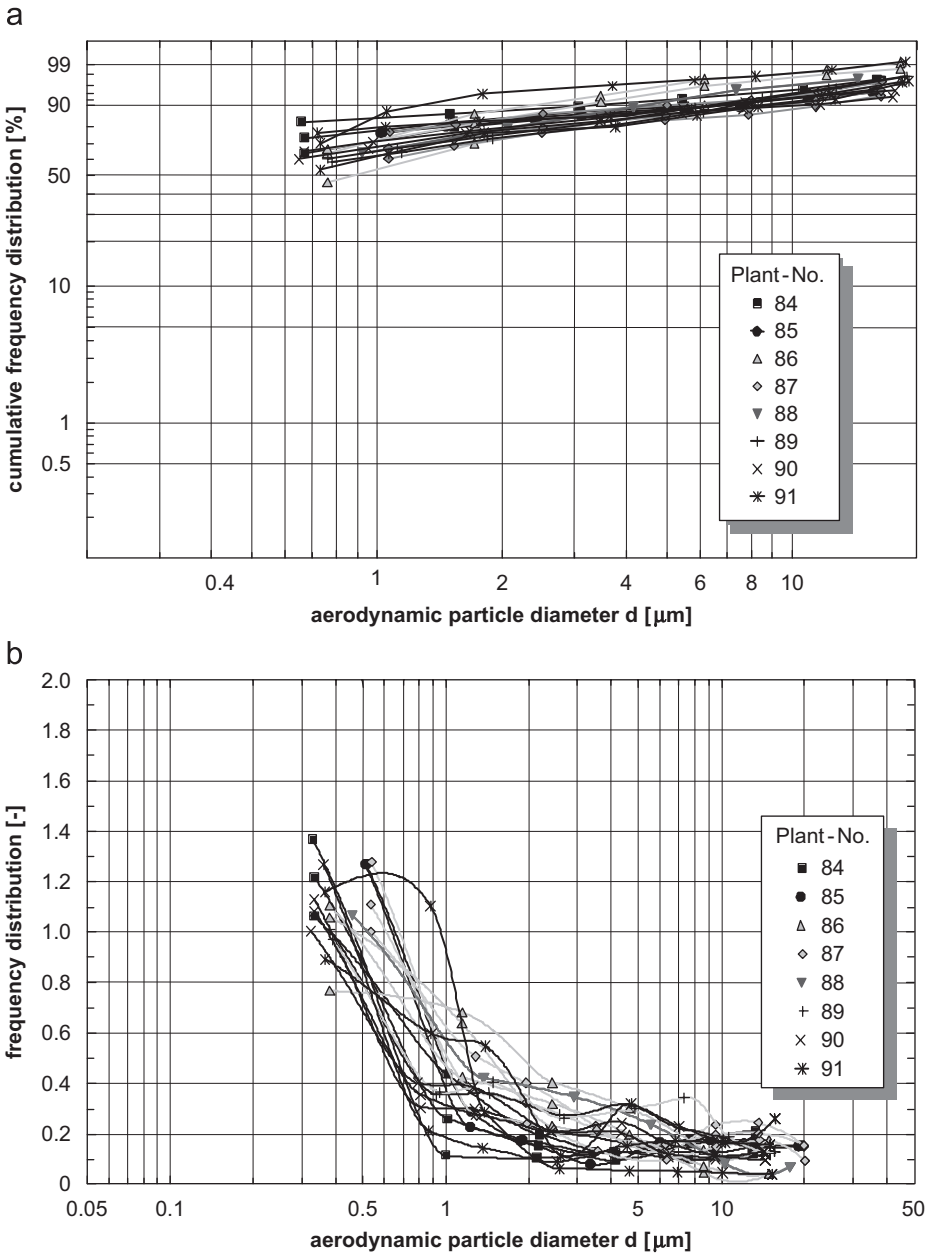

Fig. 5. (a) PM size distributions of small firing plants using brown coal briquettes, RRSB cumulative frequency distribution; plant numbers according to Table 4. (b) PM size distribution in flue gases of small firing plants using brown coal briquettes, frequency distribution; plant numbers according to Table 4.

the plant with grate cooler and electrostatic precipitator (plant No. 38). According to the information supplied by the Research Institute of the Verein Deutscher Zementwerke e.V. (1998) the reason for these coarse particles in the waste gases of the plant Nos. 36 and 37 could be that porous sites in the filter medium or leaks may, in addition, allow coarser particles to pass the filter. Since the raw gas PM concentrations in the clinker cooler (plant No. 38) are usually relatively low, defects in a filter tube do not necessarily lead to high emission concentrations. Furthermore it is documented in literature (Lützke and Muhr, 1981) relating to baghouse filters that the dust quality may be dependent on the interactions of the particles with the filter cake and associated agglomeration

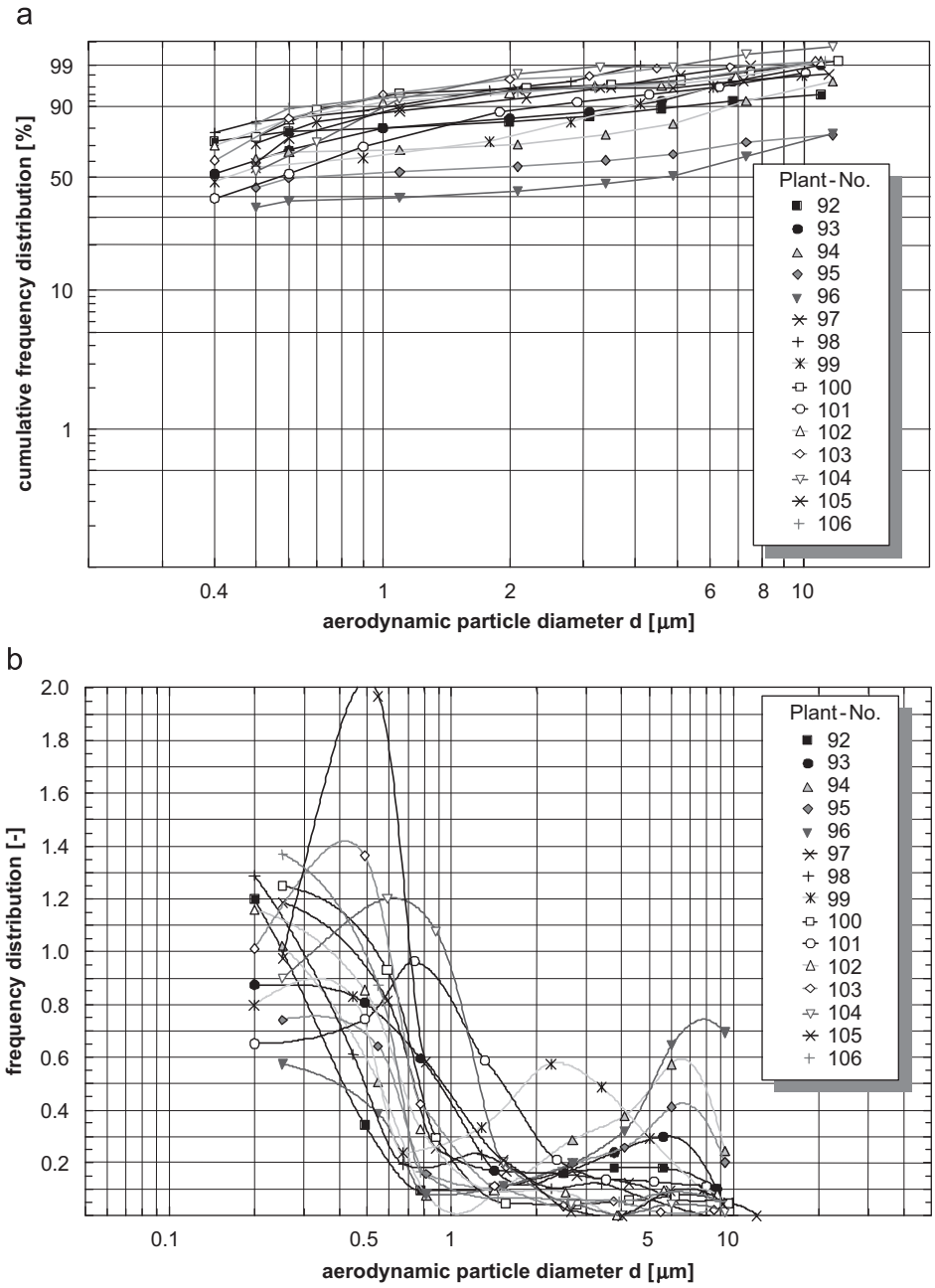

Fig. 6. (a) PM size distributions of wood combustion plants, RRSB cumulative frequency distribution; plant numbers according to Table 4. (b) PM size distribution in flue gases of wood combustion plants, frequency distribution; plant numbers according to Table 4.

phenomena, and thus coarser particles have been found in the clean gas.

These unexpected results of plant No. 36 were confirmed by repeated measurements taken half a year later. The new data, at the same plant, also showed a further coarsening in the particle size distribution (plant No. 37). Thus, the mean PM_{10} portion changed from 45% to 22%, in the mean time the total dust content in the flue gas had also increased from 3.4 to 21.1 mg m^{-3}.

4.5. Small-scale firing units

Figs. 5(a), (b), 6(a) and (b) typify the particle size distributions in the flue gases of small combustion plants. The particle size distributions of the 9 kW furnaces fired with log wood (beech, spruce) (see plants No. 100–103 in Figs. 6(a) and (b)) showed a greater proportion of fine particulates (see Table 4) than briquette-fired plants of a similar capacity (plants No. 84–91 in Figs. 5(a) and (b)). The fine

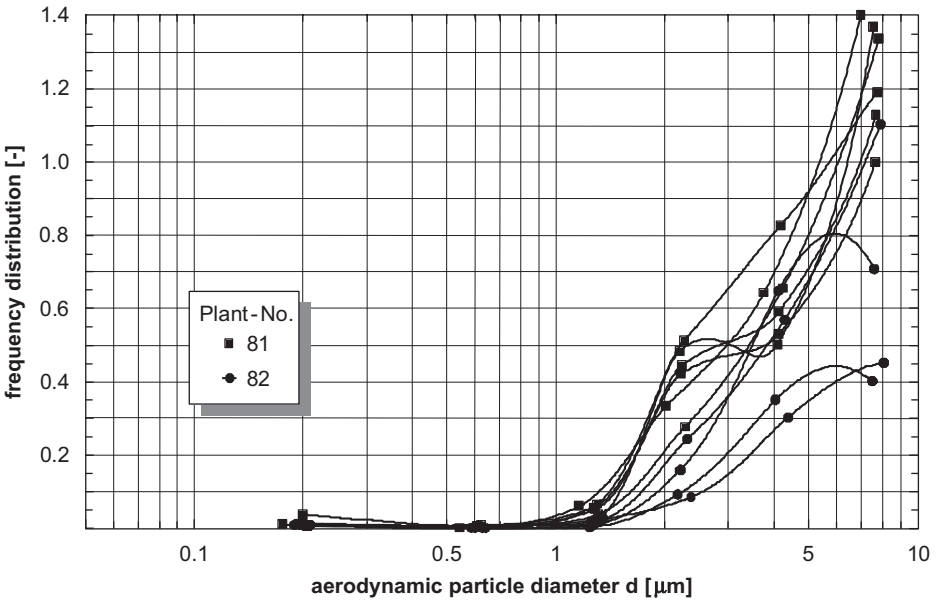

Fig. 7. PM size distribution in waste gases of poultry farming plant, frequency distribution; plant numbers according to Table 3.

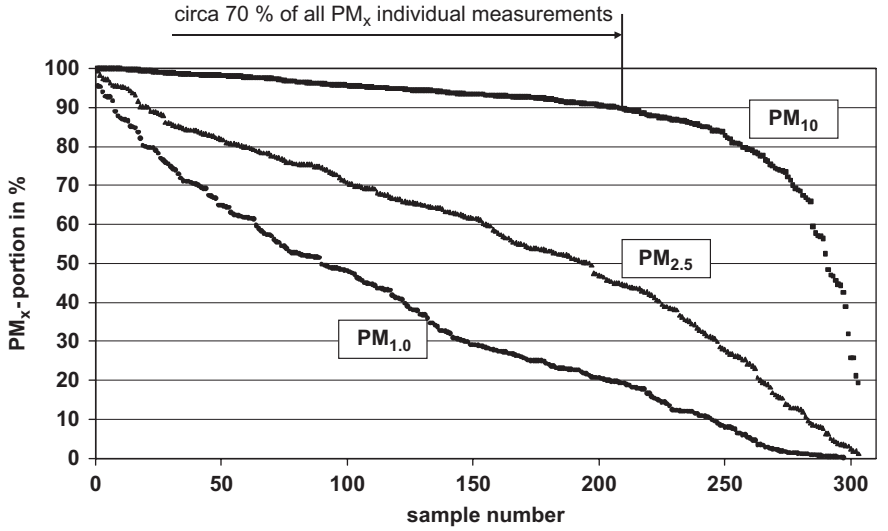

Fig. 8. Schematic depiction of the PM_x fraction of individual measurements—in descending order.

particulate fraction, which extends into the ultrafine range, cannot be completely collected using the measuring technique used here (VDI 2066 Bl. 5, 1994). These fine and ultrafine particles are formed from volatile exhaust components that condense during cooling in the exhaust duct and deposit on the surfaces of existing fine particles (such as soot) or form new particles. In the case of biomass combustion, wood in this case, volatile alkali compounds such as potassium salts are also present (Nussbaumer, 2003).

Wood combustion plants with larger capacity (175 kW, plant Nos. 92–99, Figs. 6(a) and (b)) show distinctly coarser PM distributions. Here, it would seem that coarser mineral ash components carried away by highflow caused of the suction ventilators thereby enhancing the "coarse" mode.

4.6. Poultry farming plant

Fig. 7 shows the PM size distributions of emissions from a poultry farm. The emissions from eight barnstables with 14.500 broilers per stable were determined. Here the predominant influence on PM formation is mechanical particle resuspension rather than any thermal process. As might therefore be expected the resultant distribution shows more coarse particles and is not bi-modal. The measurement results show that 50% of the particles are larger than 10 μm (see Table 3).

4.7. Comparison of plant groups

It is difficult to define PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ profiles that are abatement technology specific. There are too many factors that might influence

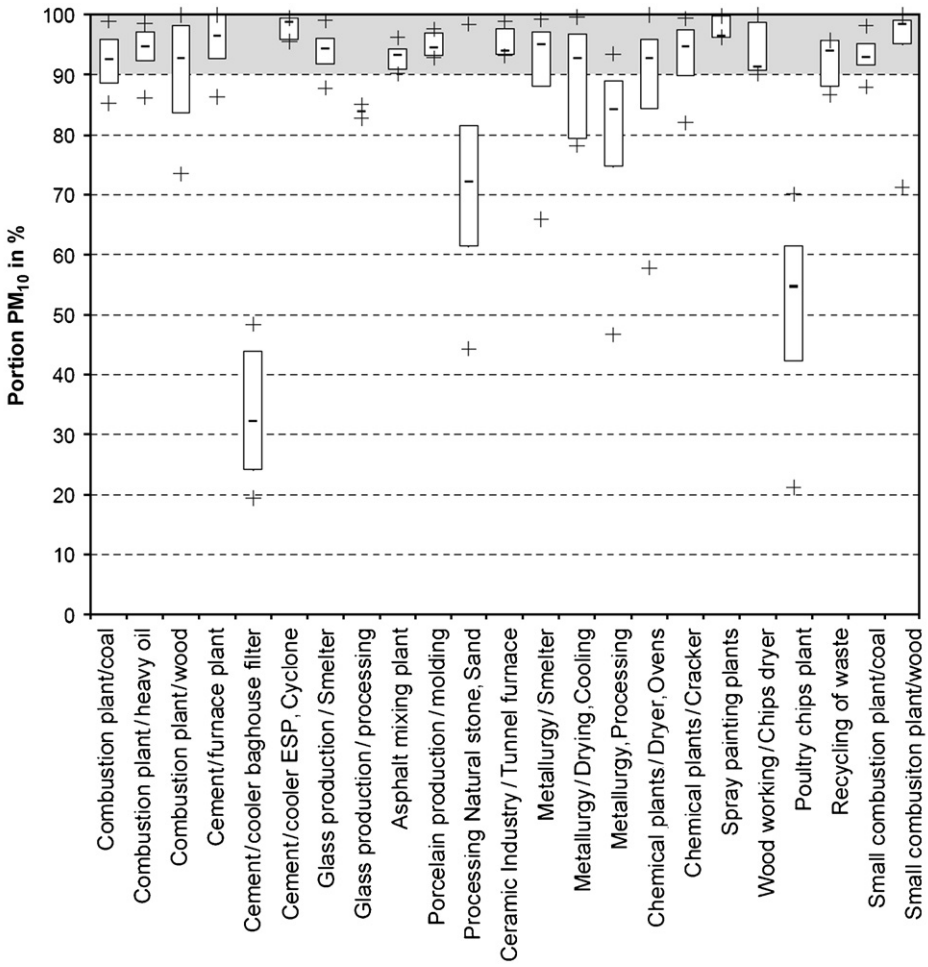

Fig. 9. Portion of PM_{10} for all investigated plant groups.

particle size distributions and a limited number of data is available to test possible relationships. The emitted PM spectra from the industrial processes tested showed a great variety. The most important factor appeared to be whether the main process is thermal, mechanical or a mixed process. It is well known from the scientific and technical literature that ultrafine particles (smaller than 0.1 μm) are formed in thermal processes, due to sublimation or condensation. On the other hand, particles formed by mechanical processes can be expected to be larger than 1 μm. In addition to these factors, the particle size distributions in the exhaust gas of combustion plants can also be influenced by: the firing and/or flue gas cleaning technologies, the fuel type and the operating conditions (load/capacity) during the PM sampling period. The results (individual measurements) from all the plants tested within the programme have been arranged according to the levels of $PM_{1.0}$, $PM_{2.5}$ and PM_{10} portion at the total dust. The result is shown in Fig. 8. It is evident that the majority of the plants investigated emit dust with a high portion of fine particles; for more than 200 of the 303 individual measurements (70%) the PM_{10} fraction accounted for greater than 90% of the total dust emitted.

Further evaluations were made of the individual plant groups investigated, the results were arranged according to the order given in the Annex of the IPPC-Directive (Council Directive, 1996) the respective PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ values are represented in box whisker diagrams—see Figs. 9–11. A box indicates the range of the 25 and 75 percentile of the measured values and the biggest and smallest value of the measurement are indicated with a cross, a (−) indicates the 50 percentile.

Fig. 9 shows a PM_{10} portion in the total dust of >90% for many plant groups (flagged area in

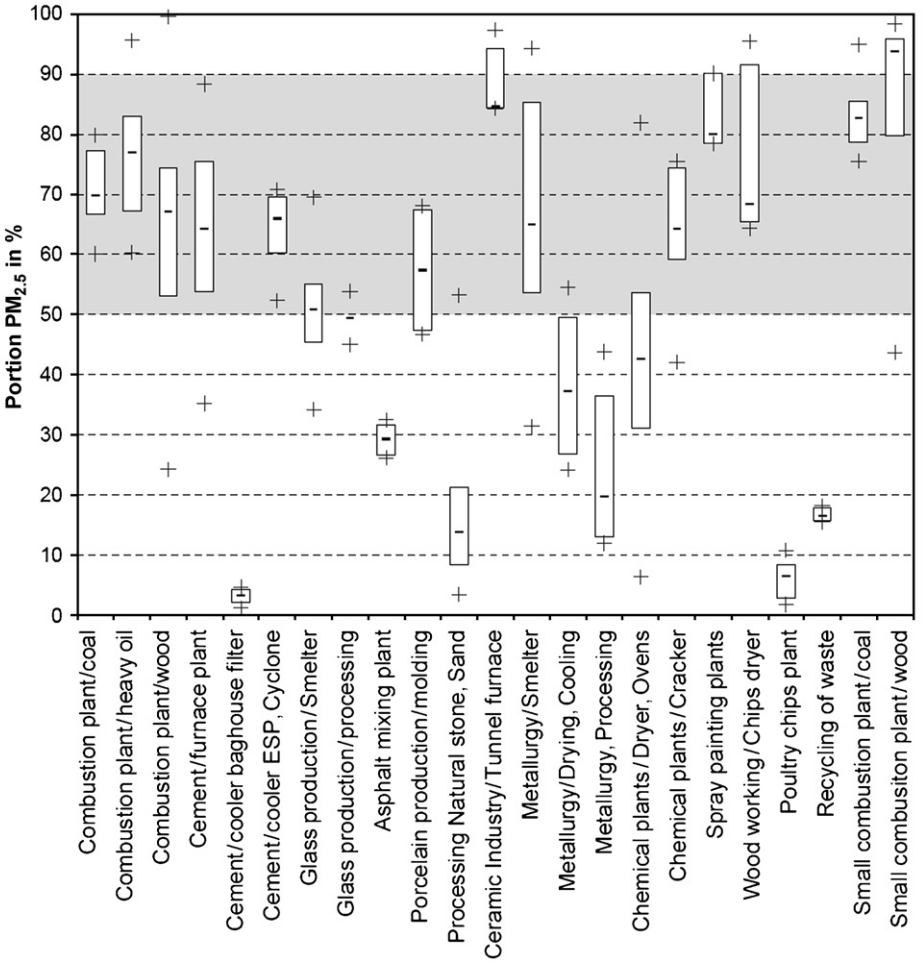

Fig. 10. Portion of $PM_{2.5}$ for all investigated plant groups.

Fig. 9). Exceptions are: Grate cooler plant with baghouse filters with the explained problems, natural stone and sand processing, poultry farming plant.

The picture is different for the $PM_{2.5}$ fraction of total dust emitted by the groups of plant under investigation: For most of the plant groups the $PM_{2.5}$ portion ranges between 50% and 90% (flagged area in Fig. 10). A $PM_{2.5}$ portion of 2–50% has been determined for the more mechanical processes such as natural stone and sand processing, poultry farming plant, drying, cooling, processing, recycling of waste and asphalt mixing plant.

A different picture also results for the $PM_{1.0}$ portion in the total dust too and three groupings can be distinguished: For the thermal processes the $PM_{1.0}$ portion lies between 20% and 60% of the total dust. The mechanical processes such as natural stone and sand processing, poultry farming plant, drying, cooling, processing, recycling of waste, asphalt mixing plant exhibit low $PM_{1.0}$ fraction of 0–30% (flagged area in Fig. 11). The third group, covering the small combustion plants (wood, brown coal briquette), chip dryers and tunnel furnaces, exhibit very high levels of $PM_{1.0}$ ranging from 60% to more than 90% of total dust.

5. Conclusion

For more than 70% of the individual emission measurements carried out at the industrial plants and at the domestic stoves, the PM_{10} portion accounted for more than 90% of the emitted dust. The $PM_{2.5}$ portion moves predominantly between 50% and 90% of the total dust. For the thermal industrial processes the $PM_{1.0}$ portion lays between 20% and 60% of the total dust.

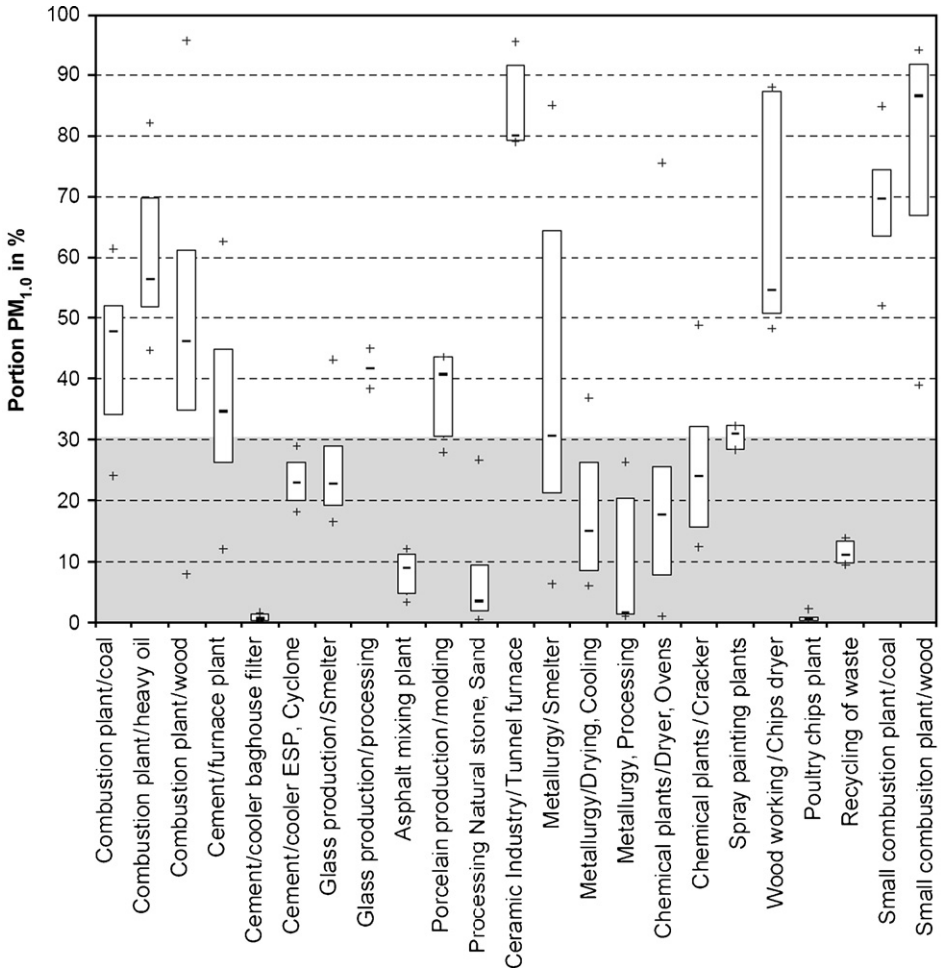

Fig. 11. Portion of $PM_{1.0}$ for all investigated plant groups.

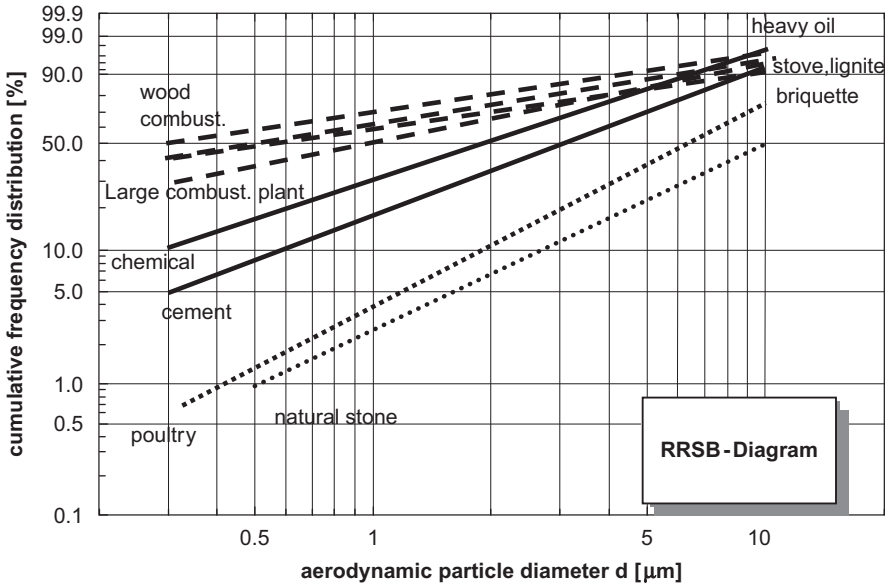

Fig. 12. Schematic comparison of typical particle size distributions in the waste gases of thermal (- - - -) (wood combustion, lignite briquette combustion, heavy oil combustion and large combustion plants), mechanical (· · · · ·) (poultry farming, natural stone processing) and mixed processes (——) (cement factories, chemical plants).

The particle size distributions determined for different plants can be clearly differentiated according to whether the emitting source is thermal, mechanical or mixed as well chemical processes, see Fig. 12. Thermal processes according to Fig. 12 are small-scale firing units using wood or lignite briquettes as fuel and large combustion plants (heavy oil, coal); mechanical processes are natural stone and poultry farming and mixed processes in this context are chemical plants and cement factories.

Consequently, for the groups of plant investigated, a major finding of this study has been that the particle size distribution is a characteristic of the technical process. Attempts to assign particle size distributions, of different plant, to different gas cleaning technologies did not give usable results.

The measurement results are of special interest for the Implementation of the European Commission's "Thematic Strategy on Air Pollution" (Communication, 2005), which was developed as a long-term, strategic and integrated policy advice to protect against significant negative effects of air pollution on human health and the environment (WHO report, 2006). In the framework of modelling of air pollution in Europe e.g. for the RAINS model of IIASA, which determines baseline scenarios towards 2020 (Amann et al., 2006), experimental determined emission factors for PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ are necessary. A contribution to that can be given by this article. Experimental determined emission factors for PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ are also very useful for establishing and revising of the "National Emission Ceilings Directive" of the European Commission (Directive, 2001/81/EC, 2001) regarding the Emission of primary particle fractions.

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