EMISSION MEASUREMENT OF PM 10 AND PM 2.5 AT INDUSTRIAL SOURCES

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1. Introduction

Recently new standards were set for ambient air concentrations of PM 10 and PM 2.5 in the European Union (Council Directive, 1999) [1]. First estimates and measurements show that both, annual and daily ambient air concentrations are expected to exceed the limit values for PM 10 at various locations in Europe. Thus, action plans will have to be set up to reduce PMx mass concentrations which requires knowledge of the sources of these particles.
For the determination of PM 10- and PM 2.5 concentrations from industrial sources there was no suitable measurement method. Measurements with multi-stage cascade impactors are used to determine the size distribution of particles in gas streams. Because they work with low gas flows, the sampling time is unacceptable long.

Therefore, the University of Duisburg and the North Rhine Westphalia State Environment Agency developed a three stage impactor ("GMU Johnas II") for the measurement of PM10 and PM2.5 emissions from industrial sources. In the following the design of the impactor will be described [2] and measurements performed at different types of plants will be presented.

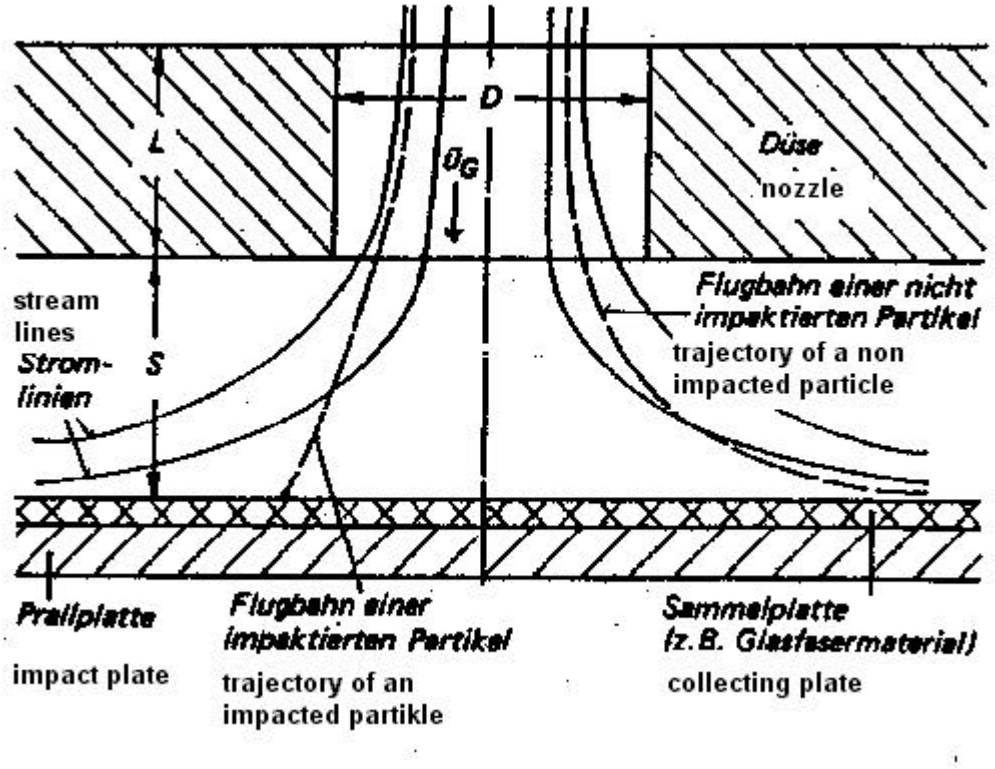
2. Definitions

PM 10; PM 2.5
Particles suspended in gas with an aerodynamic diameter < 10 µm and < 2,5 µm, respectively. The measurement method must be according to the DIN ISO 7708, where the form of the retention function of the different stages is defined.

Aerodynamic diameter
The aerodynamic diameter of a particle is the equivalent diameter of a spherical particle with a density of 1 g/cm³ showing the same aerodynamic behaviour.

Impactor
Impactors use the different inertias of particles to fractionate them into two fractions. An impactor stage is characterised by the particle diameter d50 where 50 % of the particles with this aerodynamic diameter are retained.

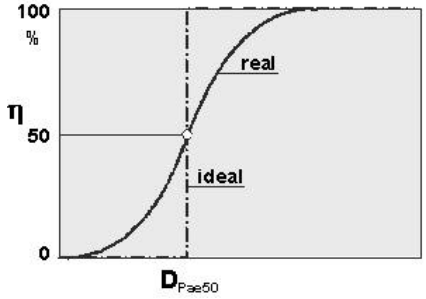
3. Principle of impaction



The retention of a particle on the impact plate depends on

- The aerodynamic diameter of the particle
- The distance between the nozzle an the impact plate
- The diameter of the nozzle
- The velocity of the gas (particle) in the nozzle
- The dynamic viscosity of the gas

The ideal function of retention can be expressed as a discontinuous function with the jump discontinuity being assigned to the cut off diameter d 50, but the real function of retention is a (sigmoidal) S curve. To characterise a stage the cut off diameter d 50 where 50 % of the particles with this aerodynamic diameter are retained is to be used.



η = efficiency of the retention

4. Description of the GMU impactor Johnas II

The design of the impactor was made according to the theory of Marple [3, 4]. Therefore the cut off diameter d(ae)50 can be calculated by

$$d(ae)_{50} = \sqrt{\frac{9p\ Stk_{50}h\ d_j^3 N_i}{4Cr_P\dot{V}}}$$

with:
Stokes number Stk_{50} = 0.24, h : dynamic viscosity of the gas, d_j : nozzle width, N_i : number of nozzles, C : Slip correction factor, particle mass density r_P = 1000 kg/m³, $\dot{V}$: gas flow.

From a practical point of view the following conditions have to be fulfilled:

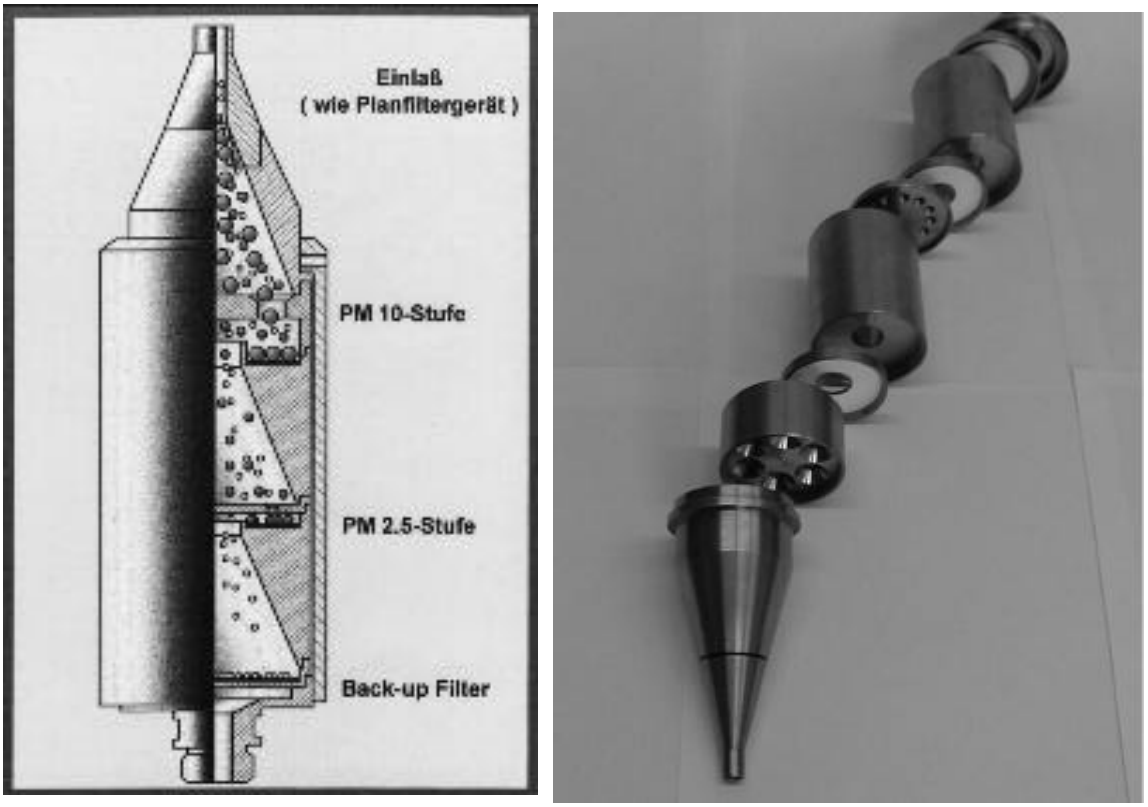
- A sample should only contain three fractions: > 2,5 µm, < 10 and > 2,5 µm and > 10 µm
- The gas flow through the impactor should be about 3 m³/h
- The measurement equipment has to be mounted in stack in line with the gas flow
- The dimensions of the impactor should fit into a 3" measurement hole
- The impactor material should be titanium due to less corrosion
- As collecting plates normal quartz filters should be used

The impactor had to be constructed in that way, that measurements are possible under the following conditions.

	mean	min	max
concentration [mg/m³] NTP	10	1	100
temperature [°C]	135	20	250
pressure [mbar]	1000	850	1100
humidity [g/m³] NTP	30	0	100
Composition of the gas	air		30% CO_2

The University of Duisburg calculated and designed a three stage impactor. An illustration and a picture are shown in figure 1. On the first impactor stage particles with an aerodynamic diameter > 10 µm are retained, on the second those < 10 µm and > 2,5 µm and on the back up filter those < 2,5 µm. Particles retained in the first diffusor are mostly particles > 10 µm. This was proved by REM.

figure 1, impactor GMU johnas



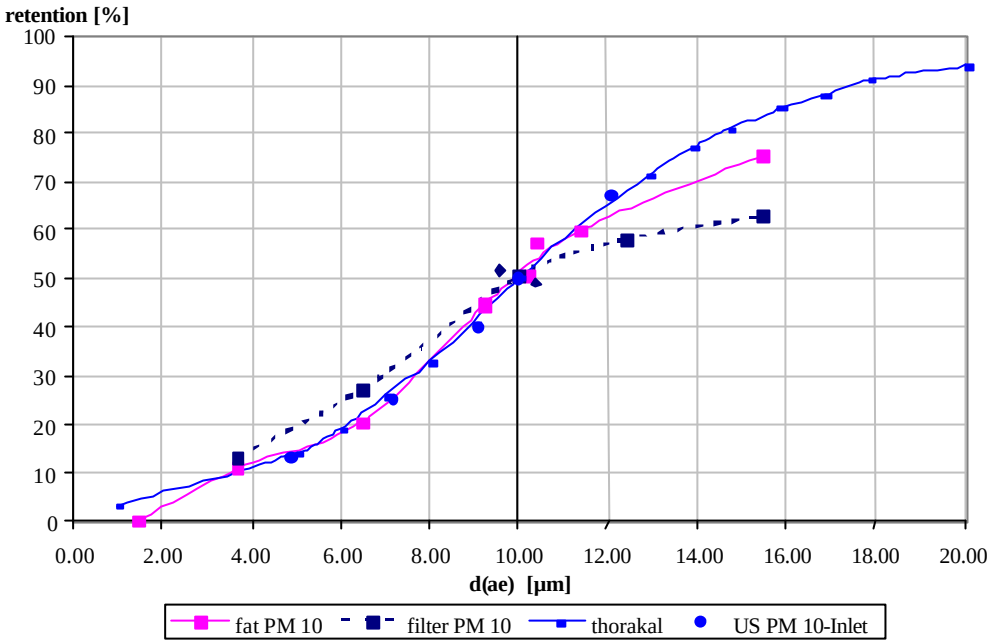
The back up filter consists of a normal 50 mm quartz fibre filter, the two impact plates consist of 50 mm quartz fibre filters with a centre hole. The impact plates are inserted into small titanium pots to ensure a quick change between two measurements.

To confirm the theoretical calculations, the two different impactor stages were calibrated with monodispersive aerosols.

The separation efficiencies of the PM 10 and PM 2.5 stage were according to DIN ISO 7708 and to the definition of PM 10 and PM 2.5.

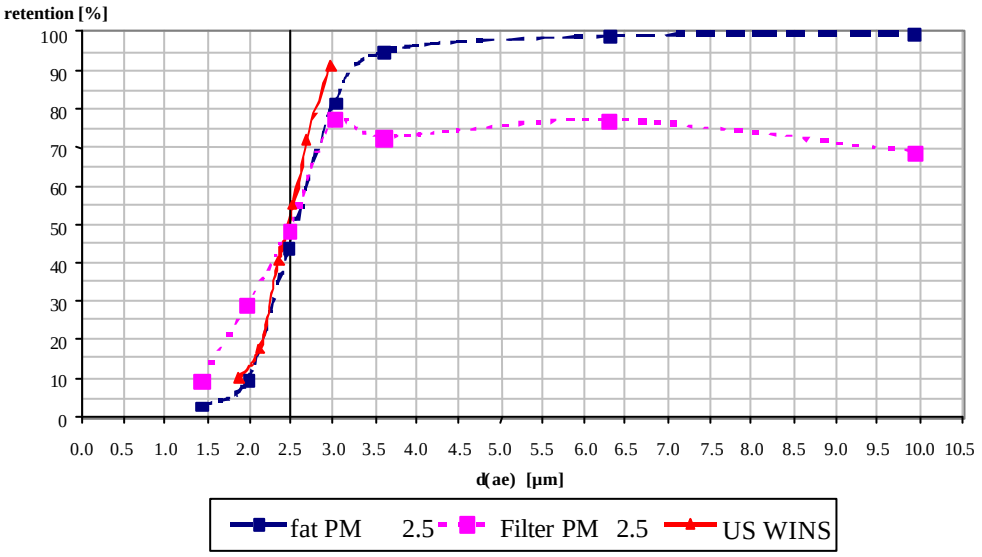
Figure 2 shows the collecting efficiency of the PM 10 stage compared with the thoracal fraction and the US PM 10 inlet.

Figure 2



The collecting efficiency of the PM 2.5 stage compared with the US WINS impactor is shown in figure 3.

Figure 3



In the first calibration experiment the impact plates were coated with fat to reduce bouncing of the particles. This variant cannot be used for emission measurements. In the second calibration experiment impact plates consisting from quartz fibre filters were used. The calibrations showed good agreements for greased impaction plates to the ambient PM 10 and PM 2.5 cut-offs and acceptable compliance if quartz fibre filters were used. The difference in

the calibration curves is in agreement with the theory on the influence of impaction surfaces on the performance of an impactor.

Because the cut off diameter of 10 µm and 2,5 µm of the different impactor stages is depending on the gas flow through the nozzles, a calculation of the gas flow - depending of the gas conditions- has to be done before any measurement can begin.

Therefore an Excel® calculation sheet was made, which is shown in figure 4. The grey fields have to be filled in with the flue gas data, the program then calculates the gas flow through the impactor and – depending on the velocity of the gas in the duct – the diameter of the extraction probe inlet. Further the Reynolds numbers for the two impactor stages are calculated. They shall be between 500 and 3000. At least the cut off diameters are calculated from the input data.

Figure 4, Excel® sheet for the calculation

Atmospheric pressure p_a :	**1021,0**		**[mbar]**
Static differential pressure p_d :	**10,0**		**[mbar]**
Temperature of the gas T :	**125,0**		**[°C]**
humidity f_0 NTP:	**30,0**		**[g/m^3]**
Percentage of CO_2 in the dry gas $r_0(CO_2)$:	**10,0**		**[%]**
Percentage of O_2 in the dry gas $r_0(O_2)$:	**11,0**		**[%]**
Percentage of N_2 in the dry gas $r_0(N_2)$:	**79,0**		**[%]**
Percentage of air in the dry gas r_0(Luft air?) :	**0,0**		**[%]**
Velocity of the gas in the duct $v_{Abgas\ waste\ gas?}$:	**13,0**		**[m/s]**

Diameter of the extraction probe inlet d_s:	8,8	[mm]
Gas flow $\dot{V}$ (eff TP):	2,85	[m^3/h]
Gas flow (NTP) $\dot{V}_0$:	1,92	[m^3/h]

Absolute pressure in the impactor p :	1031	[mbar]
Percentage of CO_2 in the humid gas $r(CO_2)$:	9,64	[%]
Percentage of O_2 in the humid gas $r(O_2)$:	10,60	[%]
Percentage of N_2 in the humid gas $r(N_2)$:	76,16	[%]
Percentage of air in the humid gas r(air) :	0,00	[%]
Percentage of water vapor in the humid gas r(aq-vap) :	3,60	[%]
Density of the humid gas (eff. TP) ρ :	0,924	[kg/m^3]
Temperature dependent dynamic viscosity of the gas η(T) :	2,1432E-05	[kg/ms]
Mean free path of the gas λ :	8,7077E-08	[m]
Velocity in the 2,5µm-nozzle $v_{2,5µm}$:	14,84	[m/s]
Velocity in the 10 µm-nozzle $v_{10µm}$:	3,88	[m/s]
Reynolds number for 2,5 µm $Re_{2,5µm}$:	1523	[1]
Reynolds number for 10 µm $Re_{10µm}$:	1557	[1]

with $\dot{V}_{mean}$ new calculated aerodyn. diameter $d_{2,5µm}(ae)_{mean}$:	2,50	[µm]
with $\dot{V}_{mean}$ new calculated aerodyn. diameter $d_{10µm}(ae)_{mean}$:	9,99	[µm]

Measurements can only be done under constant conditions. Sampling has to be done isokineticly. Because the gas flow through the impactor may not be changed during a measurement, sampling must be either performed at a representative measuring point in the duct or one has to accept the errors combined with non - isokinetic sampling if measuring is carried out at diverse points on the axis.
If measurements are made under more extrem conditions as described above, the Reynolds numbers of the two stages have to be in the range of 500 up to 3000.

Measurements in flue gases containing condensed water droplets cannot be carried out. Using the heated impactor out of the stack is not yet proved.

The aim of the measurement is to get the concentration of PM 10 and PM 2.5 in the gas flow.

The evaluation of a measurement is performed as follows.

- PM 2.5 is represented by the particle mass of the back up filter
- PM 10 is represented by the sum of the particle masses on the second impactor stage and the back up filter
- The particle mass on the first impactor stage and the conical diffusor behind the inlet are not considered in the evaluation

This procedure is similar to measurements in ambient air, where only the particle mass is collected, which passes the PM 10 inlet

The mass concentration of PM 10 and PM 2.5 is calculated by dividing the particle mass by the sampling gas volume (NTP).

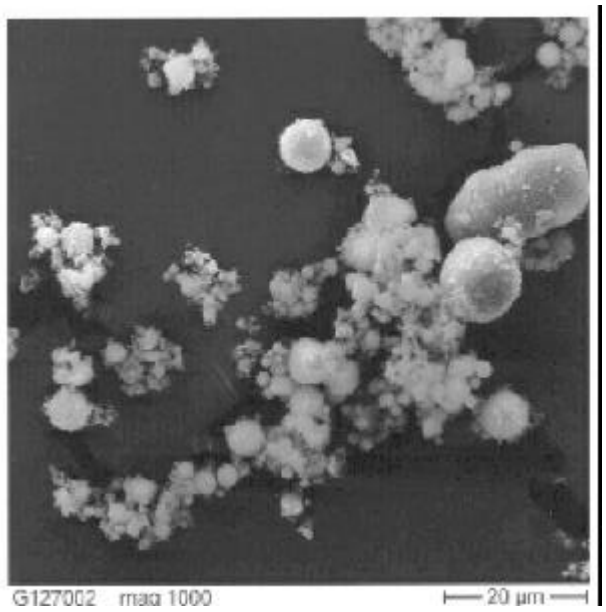
5. Measurements

Different measurements were carried out at the ESA (emission simulation installation) of the Hessian State Environmental Agency at Kassel. In this installation it is possible, to dose definite quantities of dust into a gas stream in order to realise definite concentrations of dust.

For testing a dust was used from the third stage of an electrical precipitator of a lignite fired power plant. (density: 3 g/cm³)

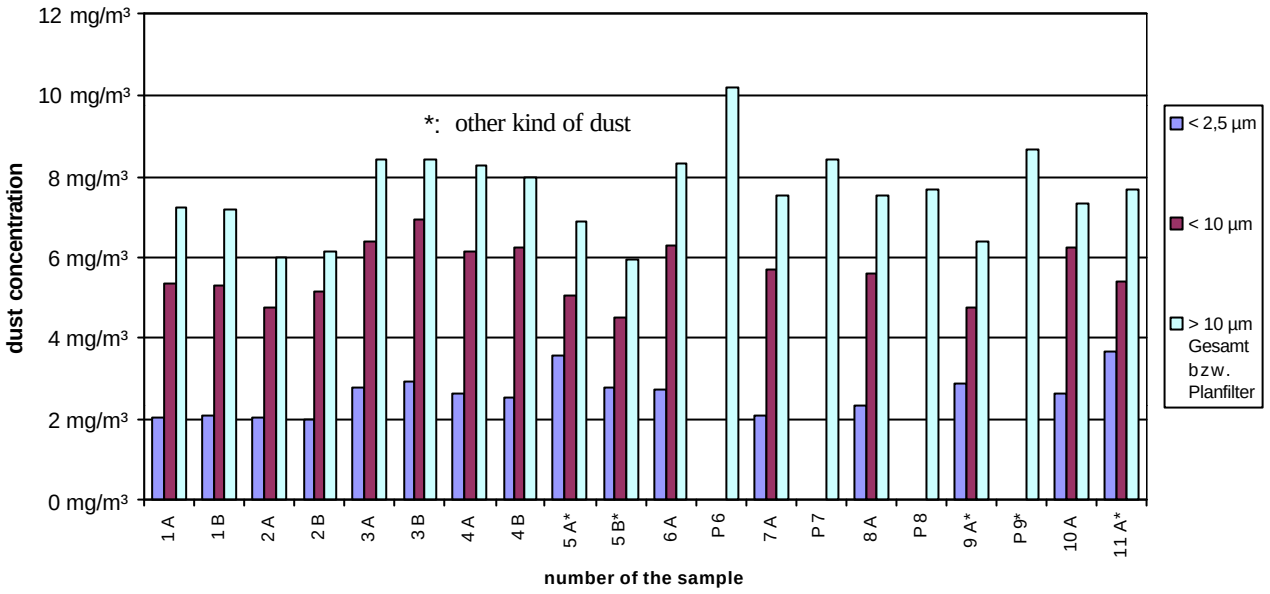
Figure 5 shows a REM picture of the dust. Most of the particles are spheric and they are not very agglomerated. Thus this dust appeared suitable for the experiments.

Figure 5, test dust



The results of the measurements are shown in figure 6

Figure 6



Samples 1A and 1B were parallely collected by the impactor. The samples PX and XA are parallel measurements with the total dust method according to VDI standard 2066 part 7.

The results are calculated in the following manner

- PM 2.5 ⇒ mass on the back up filter divided by the gas volume
- PM 10 ⇒ mass on the back up filter and the second stage divided by the gas volume
- The mass on the first stage and the dust in the conical diffusor are not considered

Figure 6 shows the concentration of PM 2.5, PM 10 and the calculated mass concentration from the addition of all masses in the impactor (1st stage plus diffusor plus PM 10 plus PM 2.5) divided by the gas volume.

From the parallel measurements with the test dust standard deviations were calculated for the PM 10 and PM 2.5 concentrations.

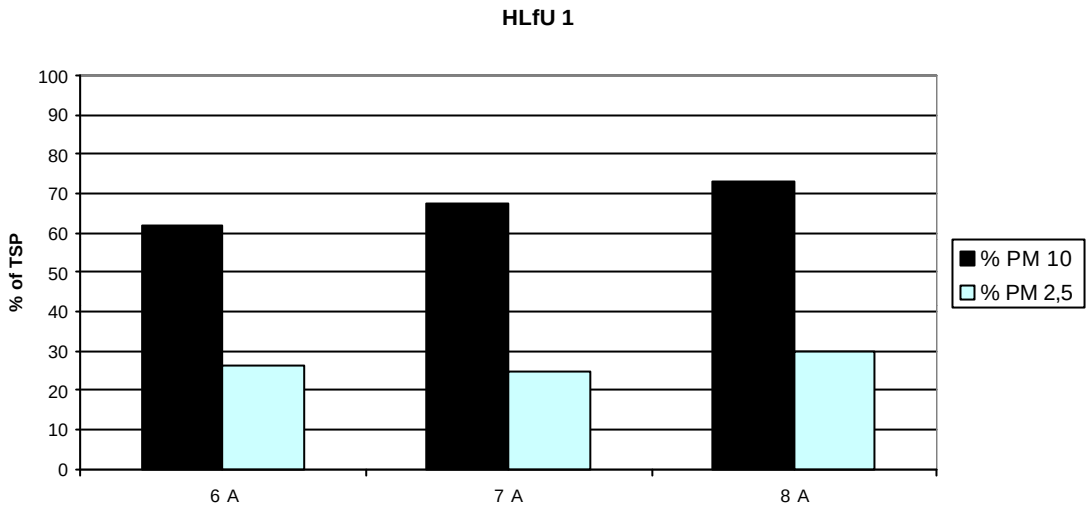
sample	PM 2.5 [mg/m³]	PM 10 [mg/m³]		sample	PM 2.5 [mg/m³]	PM 10 [mg/m³]
1 A	2,04	5,35		1 B	2,06	5,32
2 A	2,03	4,73		2 B	2,01	5,16
3 A	2,76	6,40		3 B	2,91	6,92
4 A	2,62	6,14		4 B	2,55	6,24

	mg/m³	
s (PM2.5)	0,1	2,5%
Mean value	2,4	

	mg/m³	
s (PM10)	0,2	4,2%
Mean value	5,8	

Figure 7 shows the percentage of PM 10 and PM 2.5 of the TSP resulting from parallel measurements with the impactor and the TSP method according to VDI 2066 part 7 [5].

Figure 7



6. Measurements at industrial plants

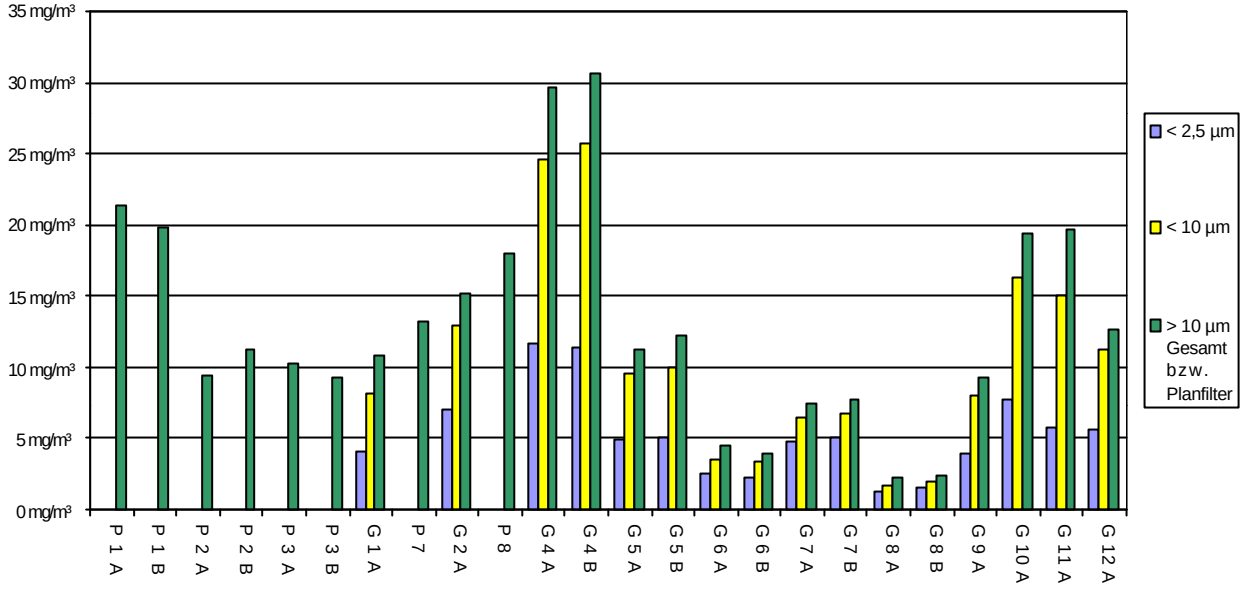
With the GMU impactor Johnas II the North Rhine Westphalia State Environment Agency made different measurements at industrial plants.

- Oxygen steel plant (electrostatic precipitator)
- Iron ore sintering plant (electrostatic precipitator)
- Cement kiln (electrostatic precipitator)
- Cement mill (fabric filter)
- Non ferrous metal industry (lead), 3 plants (fabric filter)

Most of the measurements were carried out as parallel measurements with 2 inpactors and additional comparative measurements with an impactor and the TSP method.

As an example the results at one of the plants are shown in figure 8. The flue gas was cleaned by an electrostatic precipitator.

Figure 8



Samples indicated with a P refer to measurements of TSP, samples marked with a G are measurements with the impactor.

The first 3 measurements were performed as 3 parallel measurements with two equipments of the TSP method at the two sampling points in the cross section of the stack. They show that the homogeneity of the dust concentration was acceptable. (Parallel are P1A/P1B, P2A/P2B, P3A/P3B)

In this plant the dust concentration varied over the time because of different discontinuous processes in the plant which were all dedusted applying the same cleaning system.

From the parallel impactor measurements the standard deviation for PM 10 and PM 2.,5 was calculated.

Standard deviation from parallel measurements

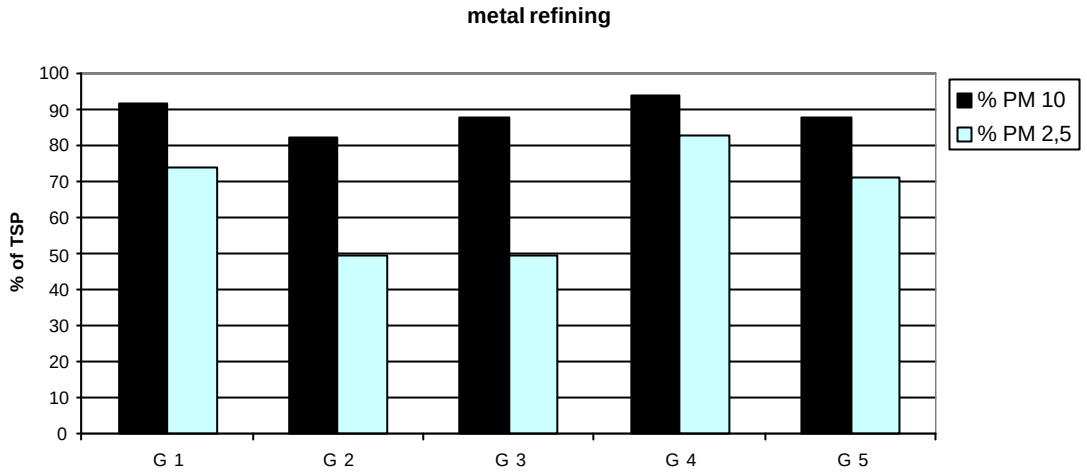
sample	PM 2.5 mg/m³	PM 10 mg/m³	sample	PM 2.5 mg/m³	PM 10 mg/m³
G 4 A	11,7	24,7	G 4 B	11,4	25,8
G 5 A	4,9	9,6	G 5 B	5,0	10,0
G 6 A	2,6	3,6	G 6 B	2,3	3,4
G 7 A	4,8	6,5	G 7 B	5,0	6,8
G 8 A	1,4	1,8	G 8 B	1,6	2,0

	mg/m³	
s(PM2.5)=	**0,2**	**3,0%**
Mean value	**5,1**	

	mg/m³	
s(PM10)=	**0,4**	**4,3%**
Mean value	**9,4**	

In the following figures the results of measurements in three industrial plants are depicted. They are results from parallel measurements of TSP and the impactor GMU johnas.

Figure 9



In this plant different discontinuous processes of metal refining were performed. The gas was cleaned by a fabric filter. The level of the dust concentration was about 1 mg/m³, the sampling duration about 6 h.

While sampling numbers G2 and G3 a special refining oven was not in use.

Figure 10 shows the results from a cement mill. The gas was cleaned by a fabric filter. The level of the dust concentration was about 1 mg/m³, the sampling duration about 4 h.

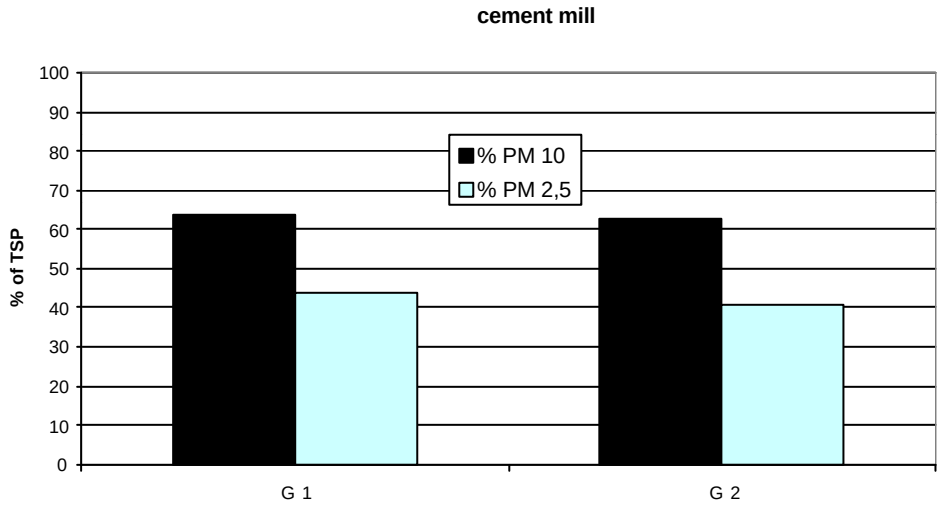


Figure 10

The third example was performed at a sintering plant during a more or less constant state of the plant. The gas cleaning system was an electrostatic precipitator. The dust concentration was between 15 and 20 mg/m³, the sampling duration 0,5 h.

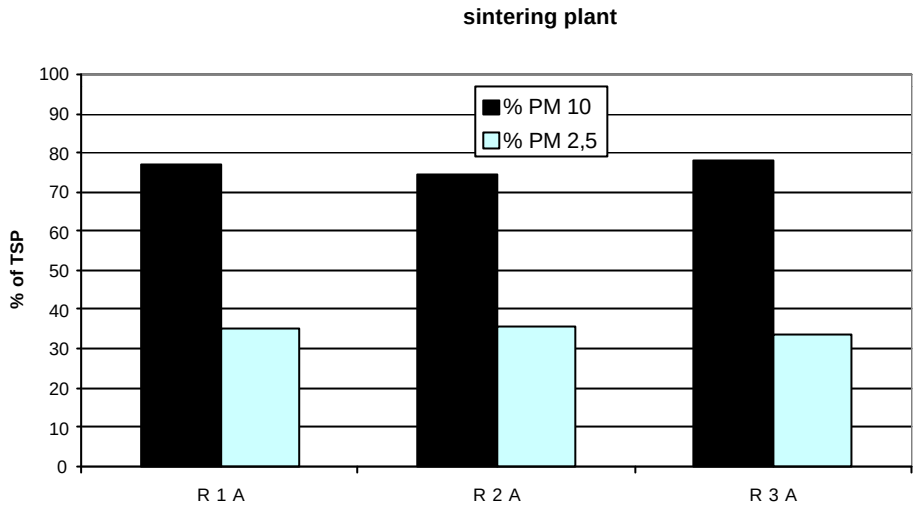


Figure 11

As a conclusion from these measurements one sees, that the percentage of PM 10 and PM 2.5 from TSP varies with different stages of the plant and the dependence of the gas cleaning system for this ratio is of second order.

7. Summary

The North Rhine Westphalia State Environment Agency and the University of Duisburg developed and validated a 3 stage impactor (GMU impactor Johnas II) for the measurement of industrial emissions of PM 10 and PM 2.5.

As the gas flow through the impactor is more than 2 m³/h (NTP), shorter measurement times are possible as compared with other impactors especially compared with the method described in VDI guideline 2066 Bl. 5 (Andersen stack sampler Mark III).

The concentration of PM 10 and PM 2.5 in the flue gas can be measured directly by weighing one impact plate and the back up filter. The change of the impact plates and the back up filter between two measurements can be carried out quickly and easily in order to avoid contamination of the samples. The loaded quartz filters can be used for further analysis of the metal content.

The GMU impactor Johnas II was tested at different industrial plants. Dust concentrations ranges varied between 80 and 0,8 mg/m³.

In Germany this measurement method will be a VDI guideline as a standard for the measurement of PM 10 and PM 2.5.

The described impactor is commercially available.

8. Literature

1. Council Directive 1999/30/EC relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air, Official J. L163, 41-60, 29/06/1999.
2. John, A.C., T.A.J. Kuhlbusch, H. Fissan, G. Bröker, K.-J. Geueke (1999). Development of a PM10/2.5-cascade impactor for in stack measurements of fine particulate matter, *Gefahrstoffe – Reinhaltung der Luft*, 59, 449-454.

3. Marple, V.A., Liu, B.Y.H., Characteristics of laminar jet impactors, *Env. Sci. and Technology* **8** (1974) S. 648.

4. Marple, V.A.; Willeke, K., Impactor design, *Atmos. Env.* **10** (1976) S. 891-896.

5. VDI Guideline 2066 Bl. 7, (1993)