A PM_{10} intercomparison exercise in Norway

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Summary

The purpose of the intercomparison was to estabilsh correction factors for samplers and automatic monitors commonly used in the Nordic countries.

To establish the correction factors, NILU performed a field test at three different locations in Norway during summer and winter conditions in the period 2001 to 2002. Each field test period lasted approx. 6 weeks. The intercomparison was performed according to the CEN/EN12341 standard.

A Kleinfiltergerät (KFG) and a high volume sampler (Andersen type) was used as reference instruments. Four other samplers and five continuous monitors participated as candidate instruments. All but two models participated in pairs as required by EN12341. TSP and meteorology was measured at all sites.

All instruments gave results close to the reference instruments. The slopes relative to the reference instruments varied from 0.80 to 1.07 and concequently the correction factors varied from 0.9 to 1.3. All instruments passed the comparison test, but not all instruments participated in the complete intercomparison exercise. Table 1 summarizes the correction factors for all intruments.

*Table 1. Correction factors..Column Use lists factors to be applied to results. Final result = Measured value * Factor.*

Candidate	Factor		Comments
	Calculated	Use	
Partisol	0.98	1.0	
NILU impactor	0.96	1.0	
Gent impactor	0.94	0.9	
IVL impactor	0.96	1.0	
EBERLINE	0.99	1.0	
TEOM w/SES	1.25	1.3	
TEOM wo/SES	1.14	1.1	
GRIMM	1.03	1.0	Only at Wood burning site
ADAM monitor	0.96	1.0	Only at Oslo Background winter
ADAM sampler	1.01	1.0	Only at Oslo Background winter

The concentrations of gaseous nitric acid were generally extremely low at all sampling sites, and was not correlated with other measured compounds.

1 Introduction

According to the European Council Directive 96/62/EC on ambient air quality assessment and management, and the first daughter directive, PM_{10} monitoring in the European countries should be harmonised. In order to harmonise PM_{10} monitoring the European Committee for Standardisation (CEN) has prepared a test procedure to ensure that the performance of candidate sampling instruments according to pertinent sampling conventions can be established reliably. The test procedure is described in the EN12341 standard, "Determination of the PM_{10} fraction of suspended particulate matter. Reference method and field test procedure to demonstrate equivalance of measurement methods".

In order to determine which PM_{10} monitors are most suitable for the Nordic conditions, NILU performed a field test at three different locations in Norway during summer and winter conditions. Each field test period lasted approx. 6 weeks. To cover the different ambient conditions as described in EN12341 the following types of test sites was been selected, indicating also the major contributing source:

- Roadside in the city (traffic, exhaust and road dust)
- Urban background in the city (all sources)
- Residential area in forrested area (wood burning for residential heating)

Wood burning, common in Nordic countries, give rise to high concentrations of VOCs, semi-volatile organic substances, and particulate matter. Ammonium nitrate is sometimes an important part of the particle mass in long range transported particles. In order to remove water from the particles before measuring the mass the air intake of monitors is usually heated. During this heating some of the volatiles may be lost. It is well-known that sampling on filters also has artifacts that can lead to loss of semi-volatile substances, but also can increase the apparent filter mass by adsorption of VOC into quarts filters. Samplers with denuders have been developed in order to reduce these effects (e.g. Ding et al., 2002a, 2002b).

Samplers that collect particles on a filter, as those used in this exercise, make use of filters that are conditioned before and after exposure for 48 hours at 20 ^{o}C and 50 % relative humidity in order to have one fixed and common basis. This process may, however, also lead to a loss of semi-volatile substances, which should be kept in mind when comparing results from monitors and filter samplers.

In order to study the performance of different reference methods under Nordic conditions two reference methods were included in the intercomparison.

2 Participants

The following institutions and companies participated in the intercomparison:

- NILU, provided several instruments, financial support and operations.
- University of Stockholm, Hans-Christen Hansson, evaluation of results.
- IVL, Martin Ferm, provided two candidate instruments.
- DMU provided one candidate instrument.
- ESM Eberline/Industriell Måleteknikk, provided two candidate instruments.
- GRIMM / Industriell Måleteknikk, provided two candidate instruments.
- Rupprecht & Patashnick (R&P)/ Oleico, provided two candidate instruments.
- NMR Hav- och Luftgrupp (HL), financial support.
- NMR Nordiska Arbetsgruppen för miljöövervakning och -Data (NMD), financial support.
- Naturvårdsverket in Sweden, financial support.

At NILU the following people participated in the project:

- Leif Marsteen, project manager, data analysis, reporting.
- Jan Schaug, data analysis, reporting.
- Steinar Larssen, quality assurance of data and report
- Nils Ladegaard, running instruments, operations.
- Jan Wasseng, running instruments, operations.
- Jan Erik Hanssen, filter preparations, weighing, chemical analysis.

3 Methods

3.1 Instrumentation

This intercomparison exercise includes the most commonly used PM_{10} samplers and monitors in the Nordic countries.

PM_{10} sampling instruments can be split into two types based on operational technics; samplers and monitors. Samplers collect particulates on a filter, typically for 24 hours. The filter is conditioned and weighed before and after sampling. The sampler can be operated either manually, requiring manual filter change every day, or automatic, requiring change of exposed filters e.g. every two weeks. The latter sampler type utilises a stack of 14 filters, each filter being changed automatically every day.

A monitor measures the collected mass continuously and the values are recorded in a data logger, typically as 1 hour averages. The most commonly used measurement principles are the β-gauge and TEOM. In the β-gauge instrument the exposed filter is bombarded with β-particles and the number of β-particles penetrating the filter counted. As mass accumulates on the filter the fewer β-particles penetrate the filter. The number of β-particles penetrating the filter is proportional to the particulate mass on the filter. In the TEOM instrument (Tapered Element Oscillating Microbalance) a small filter is located on top of a hollow pin. The pin is forced to oscillate at its resonance frequency. As mass accumulates on the filter the resonance frequency decreases. The resonance frequency is inversly proportional to the accumulated mass on the filter.

In the present exercise monitors were kept running continuously while a filter change was performed manually every day on the samplers.

Three categories of instruments were included in the exercise:

- Reference instruments
- Candidate instruments
- Instruments characterising the measurement site

Table 2 lists the reference instruments. The reference instruments comply with the requirements of EN12341.

Table 2. Reference instruments.

#	Instrument	Make	Owner
1	Kleinfiltergerät, LVS3	Leckel	NILU
1	PM_{10} High volume sampler	Andersen	NILU

Table 3 lists the candidate instruments. EN12341 requires two instruments of each kind. This was accomplished for all candidate instruments except for the TEOM monitor without nafion drier, the ADAM monitor/sampler and the Partisol where only one instrument of each was available.

Table 3. Candidate instruments.

#	Instrument	Make	Owner
2	TEOM monitor with nafion drier	R&P	R&P
1	TEOM monitor without nafion drier	R&P	NILU
2	FH62 I-R monitor	Eberline	ESM
2	GRIMM monitor	GRIMM	GRIMM
1	SM200 ADAM monitor/sampler	OPSIS	OPSIS/DMU
1	Partisol sampler	R&P	Oleico
2	PModel S10 Sampler	IVL	IVL
2	EK sampler with University of Gent impactor	NILU	NILU
2	EK sampler with NILU impactor	NILU	NILU

EN12341 requires that the meteorology as well as the PM_{10} fraction of TSP at the measurement site is documented. To accomplish this a meteorology station and a TSP high volume sampler were installed at each site. In order to verify the chemical composition of the particles as far as possible , especially ammonium nitrates that is semi-volatile, a 3-filter sampler was included consisting of a teflon filter, an alcaline impregnated filter for acid gases and an acid impregnated filter for ammonia. Table 4 lists the instruments characterising the measurement site.

Table 4. Instruments characterising the measurement site.

#	Instrument	Make	Owner
1	TSP High volume sampler	Anderson	NILU
1	EK TAC 3-filter sampler	NILU	NILU
1	Meteorology tower with wind speed and direction, relative humidity, temperature and barometric pressure, indoor temperature	Aanderaa	NILU

The same instruments were used at all sites and in all measurement periods.

3.2 Instrument setup at the measurement sites

The instruments were located in two rows on a platform on top of a shelter. The instruments were always located in the same position. Figure 1 shows the instrument layout on the platform. The platform was approx. 2.5 m above ground. The sampling inlets were located at approx. 1.5 m above the platform. The meteorological tower was fixed to one corner of the shelter and the sensors were located in the tower at approx. 7.5 m above ground.

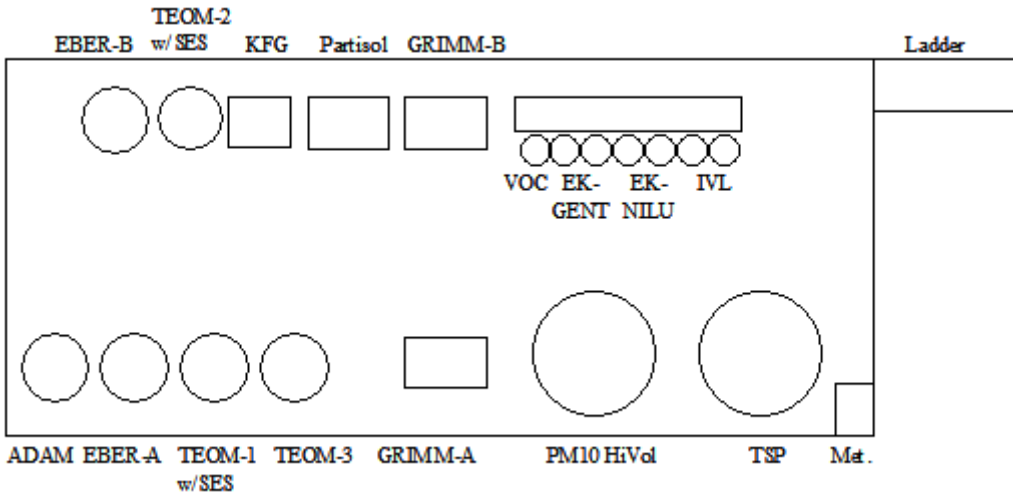

Figure 1. Instrument layout on platform.

The TSP HiVol, PM_{10} HiVol, KFG and Partisol samplers and GRIMM monitors were located on the platform. The TEOM, Eberline and ADAM monitors were located on benches along the walls inside the shelter. The IVL, EK NILU, EK Gent and VOC samplers were located in a separate room just inside the entrance. Temperature was measured continuously inside the shelter both in the monitor and sampler areas.

Figure 2 shows the shelter and platform at the road side site in Oslo.

Figure 2. Instrumentation on top of the platform.

Figure 3 shows some of the instruments on top of the platform

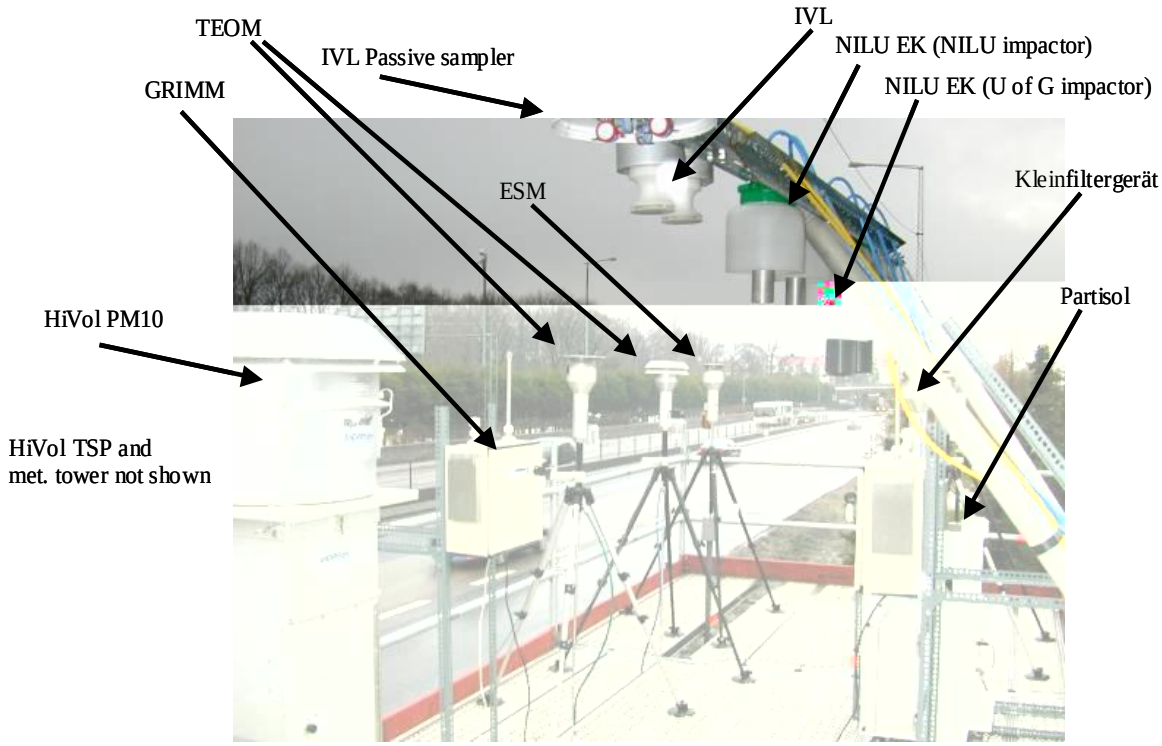

Figure 3. Instrument platform.

3.3 Measurement locations and measurement periods

The measurement sites were selected to represent both common and extreme situations in accordance with the European standard EN12341. Two sites were located in Oslo; roadside at E6, the main north-eastern highway connecting the city with residential areas outside Oslo, and in urban background surroundings. The third site was a background site located in Elverum about 120 km north of Oslo.

The Oslo roadside site is exposed to heavy traffic from the highway that carries about 45000 vehicles per day. The surrounding area consists of a mixture of fairly new office buildings and residential buildings from the fifties and sixties. A large churchyard is located at the opposite side of the highway at the site location.

The Oslo background site at Sofienberg was placed in a former churchyard that today is a green park with trees. The surrounding area is mainly residential and consists even today to a high degree of four to five floor buildings built between 1850 and 1900 for workers. Heating in these buildings is by electricity, but many probably also use stoves during cold weather. Many buildings in the area have been renovated the past 10 – 20 years and some have been taken down and replaced by modern residential houses. Four roads carry the traffic through the area at about 200 - 300 m distance to the west and east of the site.

Elverum is a community located northeast of Oslo with about 18000 inhabitants surrounded to a large extent by forests, and with activities connected to forestry, agriculture, besides some industry. Most people live in private houses, one to two floors high, and heated with a combination of electricity and wood stoves. The city was rebuilt at the end of the forties and in the fifties.

Table 5 lists the measurement sites and measurement periods.

Table 5. Measurement sites and periods.

Site	**Season**	**Start**	**End**
Oslo Roadside	Winter	16 January 2001	8 March 2001
	Summer	Aborted, no data	
Oslo Urban background	Summer	12 September 2001	25 October 2001
	Winter	28 October 2001	13 Desember 2001
Elverum Wood burning	Winter	30 January 2002	14 March 2002
	Summer	22 May 2002	2 July 2002

Due to construction work close to the Oslo Roadside station the summer season testing at the site had to be stopped there shortly after it had

started. The summer season at the Oslo Urban background site is thus actually in autumn, and the winter season at the same site started just after the autumn measurement period.

The instruments ran for approx. six weeks at each site collecting 4 samples from each analyser each week. With a total of 5 sites (not including the Oslo Roadside summer season) this totals 120 samples from each sampler which is far more then the 40 samples required by EN12341.

3.4 Sampling characteristics

The TEOM monitors with nafion driers were run at 30 °C inlet tube temperature. The TEOM without nafion drier (traditional type) and the Eberline monitors were run at 50 °C inlet tube temperature. The SM200 kept the inlet tube at ambient temperature by covering it with ambient air. The inlet tube was mounted inside an outer tube and ambient air was flushed through the outer tube continuously.

The measurement principles and sampling characteristics are shown in the tables below.

Table 6. Sampling characteristics, reference instruments.

Instrument	**Meas. principle**	**Impactor**	**Design flow**	**Reference temp/press**	**Meas. period**
Kleinfiltergerät, LVS3	Gravimetric	LVS3-PM_{10}/ EN12341	2.3 m^3/h	273 K/ 1 atm	23 h
PM10 High volume sampler	Gravimetric	HVS-PM_{10}/ EN12341	68 m^3/h	298 K/ 1 atm	23 h

Table 7. Sampling characteristics, candidate instruments.

Instrument	Meas. principle	Impactor	Design flow	Reference temp/pres	Meas. period
TEOM monitor w/nafion drier	Micro-balance	SA 246b	1 m^3/h	Operational	1 h
TEOM monitor wo/nafion drier	Micro-balance	SA 246b	1 m^3/h	Operational	1 h
FH62 I-R monitor	β-gauge	SA 246b	1 m^3/h	273 K/ 1 atm	1 h
GRIMM monitor	Light scatter	No impactor	1.2 l/min	Unknown	1 m / 5 m
SM200 monitor/ sampler	β-gauge/ gravimetric	LVS3-PM_{10}/ EN12341, size-adjusted to flow	1 m^3/h	273 K/ 1 atm	24 h
Partisol sampler Model 2025 *	Gravimetric	SA 246b	1 m^3/h	298 K/ 1 atm	23 h
PModel S10 sampler	Gravimetric	IVL inhouse	1 m3/h	Operational	23 h
EK sampler	Gravimetric	Gent inhouse	0.9 m^3/h	Operational	23 h
EK sampler	Gravimetric	NILU inhouse	0.6 m^3/h	Operational	23 h

SA 246b is the Sierra Andersen PM_{10} inlet and impactor.

* Partisol sampler Model 2025 has previously proven to be an equivalent sampler (Charron et al., 2004).

Table 8. Sampling characteristics, instruments characterising the measurement site.

Instrument	Meas. principle	Impactor	Design flow	Reference temp/pres	Meas. period
TSP High volume sampler	Gravimetric	No impactor	68 m^3/h	298 K/1 atm	23 h
EK TAC sampler	Sampling on filters	No impactor	~0.9 m^3/h no control	Operational	23 h

3.5 Filters and filter conditioning

All aerosol filters were conditioned 48 hours at 20 °C and 50 % relative humidity before weighing in a clean-room both before and after exposure.

Table 9. Filter characteristics, reference instruments.

Instrument	Filter type
Kleinfiltergerät, LVS3	Whatman QM-A, Quartz 47 mm
PM_{10} High volume sampler	Whatman QM-A, Quartz 8x10 inch

Table 10. Filter characteristics, candidate instruments.

Instrument	Filter type
Partisol sampler	Pall Zefluor 2µm 47mm Teflon membrane with Teflon backing
PModel S10 Sampler	Pall Fiberfilm 47 mm Teflon coated glass fiber
EK sampler with University of Gent impactor	Pall Zefluor 2µm 47mm Teflon membrane with Teflon backing
EK sampler with NILU impactor	Pall Zefluor 2µm 47mm Teflon membrane with Teflon backing

Table 11. Filter characteristics, instruments characterising the site.

Instrument	Filter type
TSP High volume sampler	Whatman QM-A, Quartz, 8x10 inch
EK TAC 3-filter sampler	1. Pall Zefluor 2µm Teflon membrane with Teflon backing 2. KOH-impregnated Whatman 40 cllulose filter 3. Oxalic acid imoregnated Whatman 40 cellulose filter All 47 mm

At the start of the measurement campaign quartz aerosol filters were attempted used both in the NILU and the Gent samplers. The construction of the filter holder used in these samplers with parts being screwed together caused quartz filters to be cut and parts of the filters lost during filter handling. This caused wrong filter weights after exposure and unrealistic PM_{10} masses. The first week's results for the two samplers in the comparison have therefore not been used. Membrane filters that are normally applied in these samplers then replaced the quartz filters. Additionally one result from each of these four samplers from the city background site on one specific day was invalidated. These measurements gave about fifty per cent too high PM_{10} masses, both with respect to the three equivalent methods, the KFG, the High Volume PM_{10}, and the Partisol sampler, and other samplers and monitors. The specific reason for this deviation still remains unidentified.

Similar to the four samples above, the eight last KFG reference results in the comparison, at the summer period at the wood burning site, were twice the concentrations of the all other samplers and monitors results the corresponding days. These data were also invalidated, but the specific cause of the error was not found.

3.6 Aerosol sampling and analysis

The aerosol filter in the EK TAC three-filter sampler was analysed for sulphate, nitrate, chloride, ammonium, sodium, potassium, magnesium, and calcium by IC. Nitric acid and sulphur dioxide collected at the KOH impregnated filter were determined by IC. Ammonia from the acid impregnated filter was analysed by automatic spectrophotometric method.

The sampling and analytical procedures follow the EMEP manual (EMEP/CCC-Report 1/95). Weekly field blanks and laboratory blanks were used to control contaminations and detection limits.

The quality of the inorganic chemical analyses can be checked by investigation of the ion balances in the aerosol filter results. The results from the highway site and at the "wood-burning" sites were generally very good. The ion balance in the samples from the Oslo city background site were also generally good, but had in some cases an imbalance more than 20 per cent with an apparent anion deficit.

3.7 Data treatment

The samplers ran from Monday to Friday starting each day at 13:00 and ending the next day at 12:00, hence the normal sampling time was 23 hours. The GRIMM monitor reported 1 minute or 5 minute averaged mass concentrations. The TEOM and Eberline monitors reported 1 hour averaged mass concentrations. For instruments reporting 1 minute, 5 minute or 1 hour averages the 23 hour average was computed. The hour between 12:00 and 13:00 was removed from the monitor data. The SM200 monitor reported only 24 hour averaged mass concentrations.

Concentrations and volumes were converted to standard conditons (273 K / 1 atm) before results were compared. The hourly averaged concentration reported at operational conditions by the TEOM instrument were converted to standard conditions by applying the ambient hourly averaged temperature and pressure values collected by the met. sensors. The TEOM scales its measured values according to the formula ReportedValue = 1.03 * MeasuredValue + 3. The scaled values are used is this report.

The control unit of the IVL, NILU and Gent impactors were located inside the shelter where the temperature differed from the ambient temperature. The 23 hour air volume reported at indoor conditions by these samplers were converted to standard conditions by applying the 23 hour averaged indoor temperature and ambient pressure values collected by the met. sensors.

When more then 2 hours of monitor data, not counting the hour between 12:00 and 13:00, were missing, the 24 hour average of that day was invalidated. The monitors run through the weekends and these 24 hour averaged data were included in the precision testing of the monitors.

Values from periods of instrument failures were removed from the data sets.

The intercomparison included two candidate instruments of each kind. A precision test was performed on each pair of instruments based on the 23 hour averaged data. When comparing a candidate instrument with a reference sampler, all valid 23 hour averages from the two candidate instruments were included giving twice the number of samples in the precision testing compared to the comparison with the reference data. The reference values were based on averages of the two reference samplers, the KFG and the High-volume PM_{10} samplers. The 23 hour reference value was based on one single instrument only when the other instrument did not produce a valid 23 hour measurement a specific day.

A statistical outlier test, Grubbs test, has been applied on all data sets as described in the evaluation of the designed CEN field test procedure (CEN/TC 264/WG 6). When testing data from two equivalent instruments, including the KFG and the High volume PM_{10} samplers, the results revealed 8 outliers in the dataset from the two TEOM monitors with SES, 6 outliers in the Eberline monitors' data and in the KFG - High volume sampler' data set, 5 outliers in the data from the two Gent samplers, and 4 outliers in the data from the NILU samplers and the IVL samplers. Outliers were also detected when comparing candidate data with the reference data. The total number of outliers did not allow for removing them without being in conflict with the requirement that not more than 5 per cent of the original data can be rejected on the basis of statistical tests. It was therefore decided to keep all data, and no measurements have been rejected on statistical reasons in the results in the following pages.

4 Results

All results except for the GRIMM data were converted to 273 ^{o}K and 1013 hPa. 23 hour averages (from 13:00 – 12:00 next day) have been calculated from the monitor data and the averages referred to as "daily averages" below.

Data from periods with technical problems have been rejected and are not included in this report. This includes mainly the last part of the KFG results from the wood-burning site, which did not compare with any other data and the GRIMM monitor data from both the roadside site and the city background site in Oslo.

No data have been rejected as a result of the statistical outlier tests that were performed.

The data from the candidate samples and the monitors have all been compared with reference data in view of the EN12341 standard although the standard is not prepared for automated methods. The reference data set is the average of the High-volume and the KFG results when results from both samplers were available. When results from only one of the two reference samplers were valid, those measurement have been used in the reference data set. Data from both samplers in each pair of candidates have been put into one data set (not averaged) and compared with the reference data set.

All PM_{10} measurements in this comparison were lower than 100 $\mu g_{10}/m^3$.

4.1 Summary of results

Correction factors

Table 12 lists the correction factors for each instrument type that participated in the intercomparison. When calculating the final result the measured or computed mass concentration value shall be multiplied by the correction factor.

The factor is applied as:

Final result = Measured value * Factor

Table 12. Correction factors..Column Use lists factors to be applied to results.

Candidate	Factor		Comments
	Calculated	Use	
Partisol	0.98	1.0	
NILU impactor	0.96	1.0	
Gent impactor	0.94	0.9	
IVL impactor	0.96	1.0	
EBERLINE	0.99	1.0	
TEOM w/SES	1.25	1.3	
TEOM wo/SES	1.14	1.1	
GRIMM	1.03	1.0	Only at Wood burning site
ADAM monitor	0.96	1.0	Only at Oslo Background winter
ADAM sampler	1.01	1.0	Only at Oslo Background winter

The factors apply for the TEOM, Eberline and ADAM monitors under the following conditions only:

TEOM with SES with an inlet tube heating set to 30 °C
TEOM without SES with an inlet tube heating set to 50 °C
Eberline with an inlet tube heating set to 50 °C
ADAM with ambient sheet air covering the inlet tube.

TEOM without SES is the classical TEOM.

Precision tests

Table 13 lists the results from the precision test. The test requires two identical instruments of each kind hence the Partisol is not included.

Table 13. Precision test. Candidate B or 2 vs candidate A or 1.

Instrument	**N_{pairs}**	**Slope**	**Intercept**	**r2**	**Cl95<5 ?**
PM_{10} HiVol vs KFG	120	1.09	-2.26	0.99	Pass
NILU B vs A	119	0.99	0.49	0.98	Pass
Gent B vs A	121	0.98	0.58	0.99	Pass
IVL B vs A	127	1.01	0.09	1.00	Pass
EBER 2H vs 1H	209	1.04	-0.75	0.99	Pass
TEOM 2H vs 1H	198	1.08	-0.84	0.99	Pass
TEOM 3H vs 1H [1)]	158	1.12	-1.90	0.99	Pass
GRIMM B vs A	35	0.99	-0.03	0.99	Pass
ADAMM vs S [1)]	47	1.14	-3.95	0.97	Fail

1) This is not true precision since the measurement method in each pair of instruments is not equal.

TEOM 1 and 2 are with SES. The GRIMM B vs GRIMM A results are for the Wood burning site only (winter and summer season). TEOM 3 is a

traditional at 50 °C without SES. ADAM M is values measured by the monitor part of the ADAM. ADAM S is gravimetric (sampler) analysis of the mass concentration. The ADAM participated only during the winter season at the Oslo Urban background site.

All instruments passed the precision test except for the ADAM when comparing the ADAM gravimteric analysis and the ADAM β-gauge measurement.

Comparisons

Table 14 lists the results from the comparison of candidates and reference.

Table 14. Comparison test. Candidate vs reference.

Candidate	Npairs	Slope	Intercept	r2	r2>=0.95 ?
Partisol	107	1.02	0.39	1.00	Pass
NILU impactor	239	1.04	-0.31	0.98	Pass
Gent impactor	243	1.07	-0.56	0.98	Pass
IVL impactor	254	1.05	-1.16	0.99	Pass
EBERLINE	236	1.01	0.06	0.98	Pass
TEOM w/SES	223	0.80	2.74	0.98	Pass
TEOM3H wo/SES	93	0.88	1.70	0.98	Pass
GRIMM	50	0.97	0.95	0.98	Pass
ADAMM [1)]	26	1.05	2.55	0.99	Pass
ADAMS [1)]	26	0.99	3.27	0.98	Pass

1) ADAMM is ADAM monitor and ADAMS is ADAM filter sampler

All instruments passed the comparison test. The ADAM participated only during the winter season at the Oslo Urban background site. The results of the GRIMM instrument are for the Wood burning site only (winter and summer season).

Aerosol analysis

The concentrations of gaseous nitric acid were generally extremely low at all sampling sites, and was not correlated with other measured compounds. Nitric acid will easily react chemically with particles in air. Sea salt particles will in this case give hydrochloric acid that like nitric acid is a gas and will be collected on the KOH-impregnated filter. Sulphuric acid is a hygroscopic particle that like nitric acid will give hydrochloric acid in air when reacting with sea salt particles. Hydrochloric acid was, however, not measured due to technical reasons. Interactions on the aerosol filter during sampling may have increased the measured concentrations of nitric acid and at the same time reduced the concentrations of particulate nitrate, and of particulate chloride.

The ammonium concentrations on the aerosol filter were lower than the sum of the sulphate and nitrate concentrations on equivalent basis in nearly all samples, and the median of sulphate and nitrate bound to metals ranged from about 40 per cent at the roadside site to 90 per cent at the Oslo background site during the winter measurements.

4.2 Intercomparison results

Reference samplers

Figure 4 is a comparison of the measurement results obtained with the two reference methods, the Kleinfiltergerät (KFG) and the Andersen high volume sampler with PM_{10} inlet. As seen from the Figure the difference between the two samplers was not negligible, and the High-volume PM_{10} sampler tended to give higher concentrations than the KFG at the highest concentrations. The KFG results from the end of the comparison were invalidated as commented above.

The statistics on the regression between the two reference sampler is given in Table 15 below.

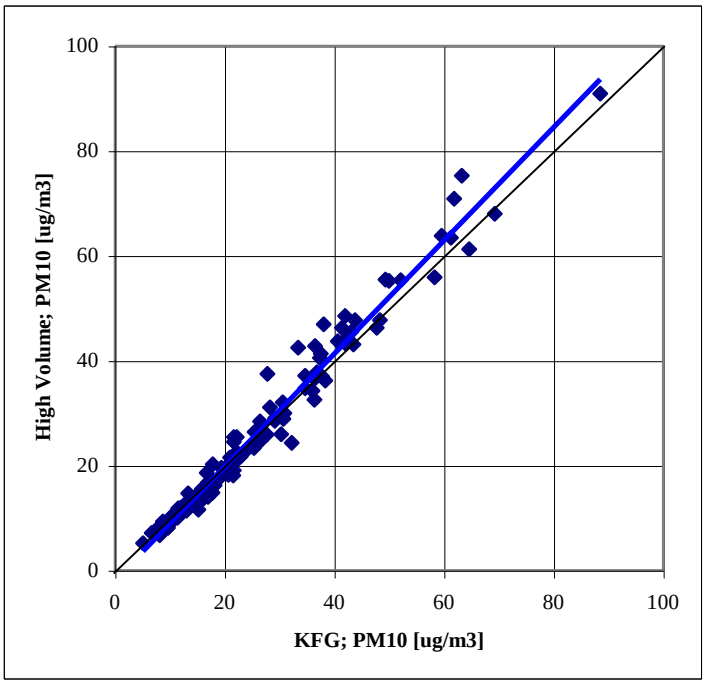

Figure 4. High Volume PM_{10} measurements compared with KFG results.

Table 15. Orthogonal regression of the High-volume PM_{10} sampler with the KFG sampler.

High-volume PM_{10} = a KFG + b*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
120	1.09	1.05	1.14	-2.26	-3.18	-1.35	0.99

The slope is 1.09, and the statistics confirm a significant difference at a 95 per cent confidence level. There is also an intercept lower than zero, which likewise is significant. The coefficient of determination (R^2) was, however, not much different from 1, and the majority of the measurements correspond very well. The overall correspondence between the two samplers was nevertheless not as good as expected.

Partisol sampler

One Partisol sampler was operated in this intercomparison. As seen from Figure 5 and Table 16 this sampler compares well with the reference data set. Table 16 gives the statistics from the regression of the Partisol sampler with the reference data. The regression line is not significantly different from a 1:1 correspondence with the reference, the intercept being near zero and the coefficient of determination (R^2) being 1.00.

Table 16. Orthogonal regression of the Partisol sampler with the reference data set.

Partisol = a Reference + b*

Num. samples	Slope			Intercept µg/m^3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
107	1.02	1.00	1.04	0.39	-0.03	0.81	1.00

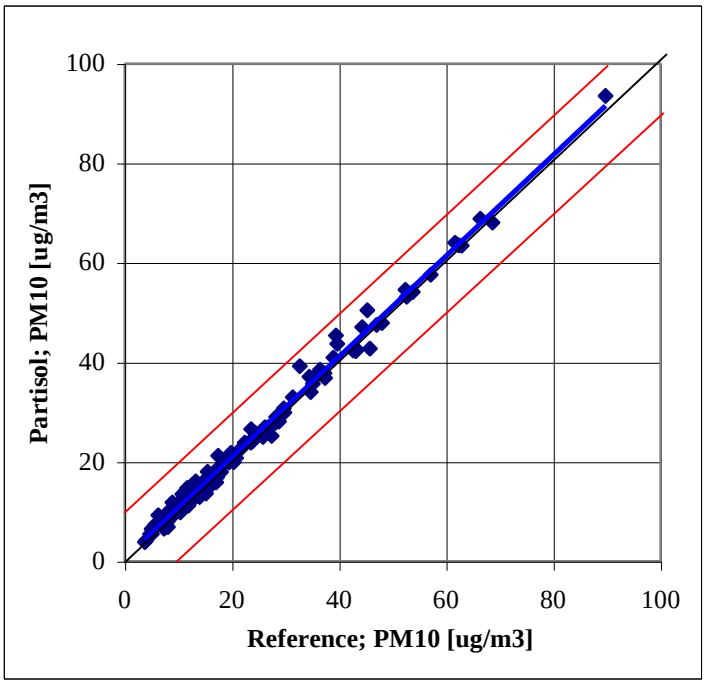

Figure 5. Partisol measurements compared with the reference data set.

Table 17 gives the factors to apply on the Partisol data in order to obtain reference equivalent measurements. The standard deviations in slope and intercept are also given.

Table 17. Slope, intercept, and their standard deviations.
*Reference = a * Partisol + b.*

Slope		Intercept µg/m^3	
a	St.dev. a	b	St. dev. b
0.98	0.01	-0.38	0.22

NILU sampler

Comparisons of the two samplers

Figure 6 compares the two NILU samplers with PM_{10} inlets. The Figure shows results that are close to the regression line and the 1:1 line. A few of the results deviate, however, from the regression line without being outliers in a statistical sense.

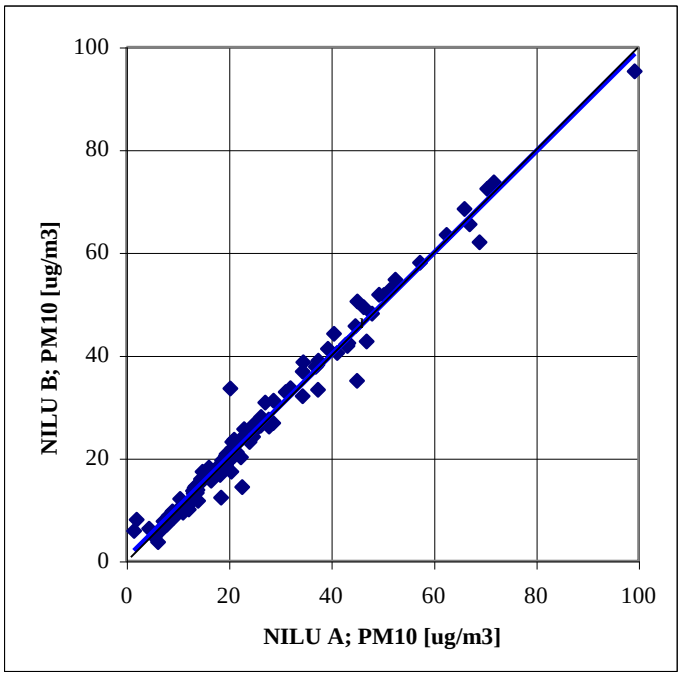

Figure 6. NILU sampler B compared with sampler A.

The measurement precision were estimated following the EN12341 standard giving the results presented in Table 18. The requirement in the standard is the 95 per cent confidence interval to be less than 5 µg PM_{10}/m^3. As seen from the Table the sampler results comply with this requirement.

Table 18. Precision of the NILU sampler, relating to the difference between parallel measurements.

Number of samples	Standard deviation µg/m^3	t $_{0.975}$	CL $_{95}$ upper limit µg/m^3
119	1.84	1.98	3.67

Table 19 gives the orthogonal regression of NILU sampler B versus sampler A. There were no significant differences between the samplers.

Table 19. Orthogonal regression of NILU sampler B with sampler A.
NILU B = a NILU A + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
119	0.99	0.96	1.02	0.49	-0.34	1.32	0.98

Comparisons of the samplers with the reference data set

The second part of the requirement in the EN12341 standard is that the calculated reference equivalence function is bounded within the limits of the y = x ± 10 acceptance envelope (red lines in Figure 7) when compared with the reference data (x), and that the variance coefficient R^2 is ≥ 0.95.

Figure 7 and Table 20 compare the NILU sampler results with the reference data set. As seen the requirement above have been met, and the sampler proven to be an equivalent to the reference samplers for PM_{10} levels lower than 100 µg/m^3.

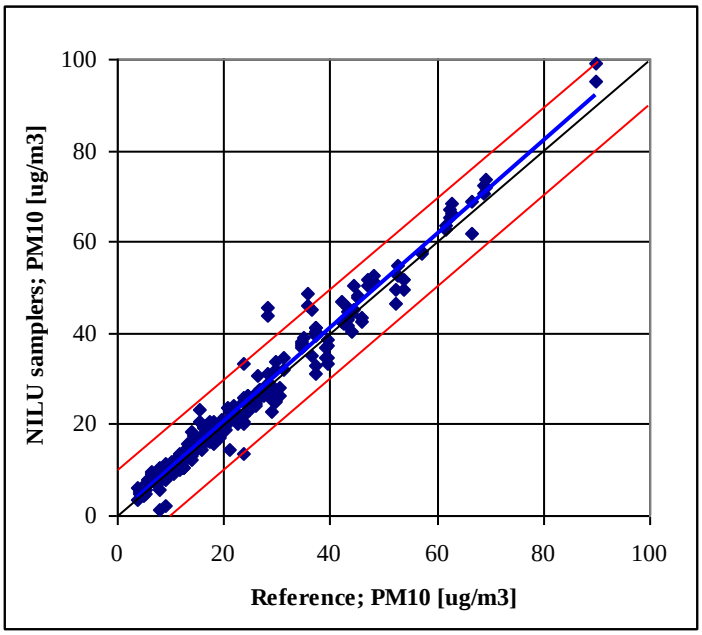

Figure 7. The two NILU samplers data compared to the PM_{10} reference data set.

The slope of the regression line in Table 20 is slightly higher than 1 while the intercept is not different from zero. The coefficient of determination (R^2) is close to 1 underpinning a good correspondence between the two data sets.

Table 20. Orthogonal regression of the NILU samplers data with the reference data set.

NILU = a Reference + b*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
239	1.04	1.02	1.07	-0.31	-0.85	0.23	0.98

Table 21 gives the factors to apply on the NILU data in order to obtain reference equivalent measurements. The standard deviations in slope and intercept are also given in the Table.

Table 21. Slope, intercept, and standard deviations.
*Reference = a * NILU + b.*

Slope		Intercept $\mu g/m^3$.	
a	St.dev. a	b	St. dev. B
0.96	0.01	0.29	0.26

Gent Sampler

Comparisons of the two samplers

The results from the two Gent samplers are presented in Figure 8. Corresponding measurements have generally very small differences, and the regression line is close to 1:1. The precision is quantified in Table 22.

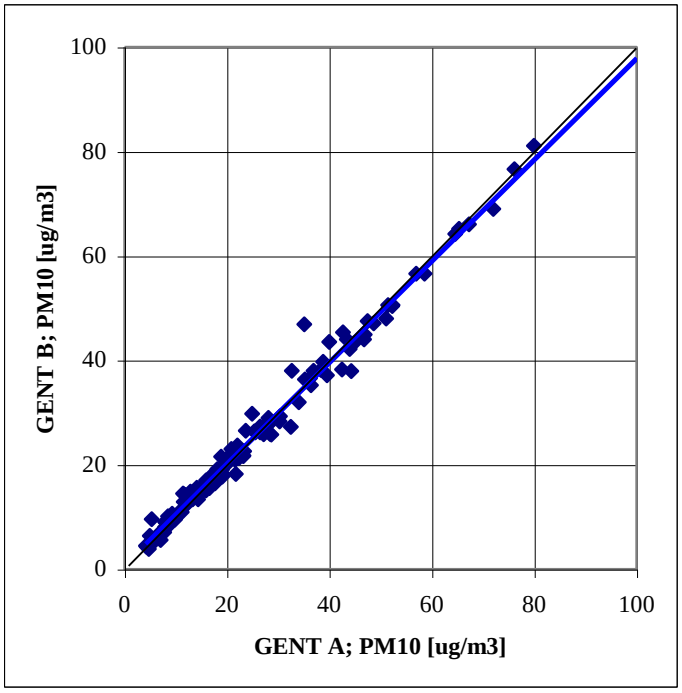

Figure 8. Gent sampler B compared with sampler A.

Table 22. Precision of the Gent sampler, relating to the difference between parallel measurements.

Number of samples	Standard deviation µg/m^3	t $_{0.975}$	CL $_{95}$ upper limit µg/m^3
121	1.46	1.98	2.90

The Gent sampler precision is good with an estimated confidence interval well below 5 ug/m^3 that is the upper limit in the EN12341 standard. Table 23 gives an orthogonal regression of samplers A and B.

Table 23. Othogonal regression of Gent sampler B with sampler A.
Gent B = a Gent A + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
121	0.98	0.96	0.99	0.58	0.18	0.97	0.99

Comparisons of the samplers with the reference data set
Figure 9 compares the Gent results with the reference data set.

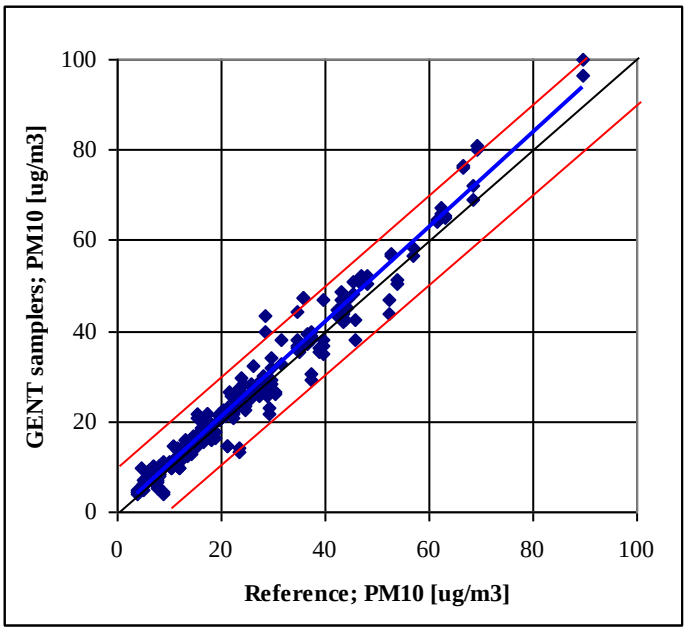

Figure 9. The two Gent samplers data compared to the PM_{10} reference data set.

The statistics on the regression of the Gent sampler data with the reference data is given in Table 24. The slope is 1.07 while the intercept is not significantly different from zero. The differences between the two data sets are small, as measured by the coefficient of determination (R^2).

Table 24. Orthogonal regression of the Gent samplers data with the reference data set.

Gent = a Reference + b*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
243	1.07	1.04	1.10	-0.56	-1.19	0.07	0.98

Table 25 gives the factors to apply on the Gent sampler data in order to obtain reference equivalent measurements. The standard deviations in slope and intercept are also given in the Table.

Table 25. Slope, intercept, and standard deviations.

*Reference = a * Gent + b.*

Slope		Intercept $\mu g/m^3$	
a	St.dev. a	b	St. dev. b
0.94	0.01	0.53	0.30

IVL sampler

Comparisons of the two samplers

An excellent correspondence between the two IVL PM_{10} samplers is seen in Figure 10. The regression line coincides almost 1:1.

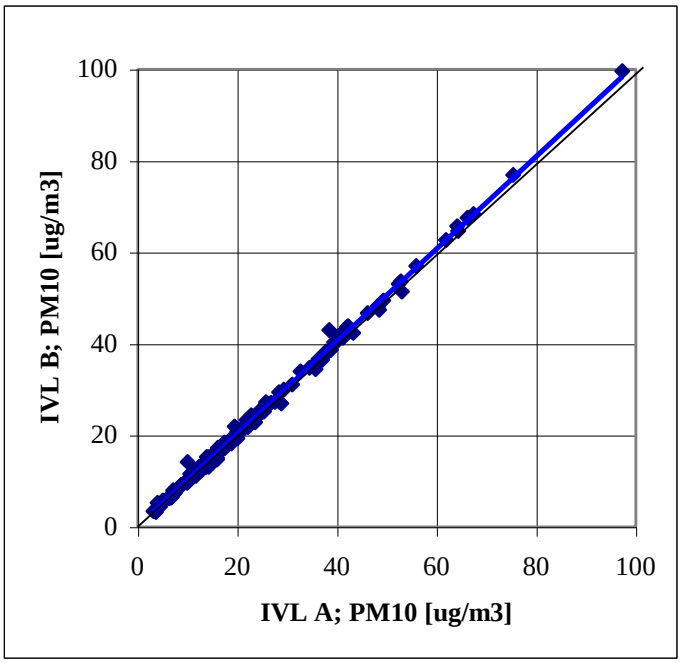

Figure 10. IVL sampler B compared with sampler A.

Table 26 gives the corresponding precision estimates. The good precision is reflected in the estimate of a low 95 per cent confidence limit as seen from Table 26 and Table 27 that gives the orthogonal regression of IVL sampler B versus sampler A. The IVL precision as expressed by the width of the confidence interval for parallel measurements is very good as seen from the two Tables, and the variance coefficient is 1.00.

Table 26. Precision of the IVL sampler, relating to the difference between parallel measurements.

Number of samples	Standard deviation µg/m^3	$t_{0.975}$	CL_{95} upper limit µg/m^3
127	0.68	1.98	1.33

Table 27. Orthogonal regression of IVL sampler B with sampler A.
IVL B = a IVL A + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
127	1.01	1.00	1.02	0.09	-0.15	0.32	1.00

Comparisons of the samplers with the reference data set

Figure 11 compares the average of the two IVL samplers with the reference data set. All measurements are within the envelope described above,

and the sampler satisfies the requirement in the EN12341 standard as an equivalent sampler for PM_{10} concentrations less than 100 ug/m^3.

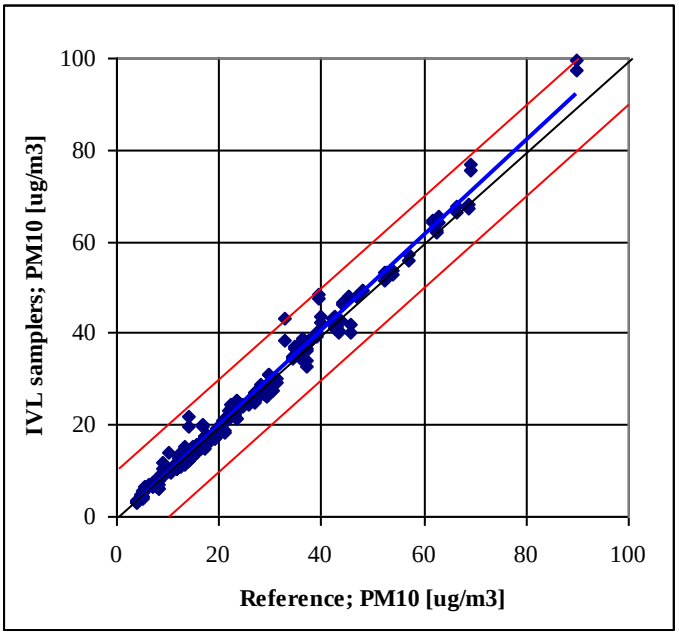

Figure 11 The two IVL samplers data compared to the PM_{10} reference data set.

Table 28 presents the statistics from the orthogonal regression.

Table 28. Orthogonal regression of the IVL samplers data with the reference data set.

IVL = a Reference + b*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
254	1.05	1.02	1.07	-1.16	-1.64	-0.67	0.99

The slope and the intercept are somewhat different from one and zero respectively, but the differences between the regression line and the measurements in the Figure were in general very low as indicated by the coefficient of determination (R^2).

Table 29 gives the factors to apply on the IVL data in order to obtain reference equivalent measurements. The standard deviations in slope and intercept are also given in the Table.

Table 29. Slope, intercept, and standard deviations.
*Reference = a * IVL + b.*

Slope		Intercept µg/m^3	
a	St.dev. a	b	St. dev. b
0.96	0.01	1.11	0.23

Eberline monitor

Comparisons of the two monitors

Figure 12. shows good correspondence between the two monitors in this comparison. The precision is good and the regression line is almost equal to the 1: 1 line. There were no outliers in the data.

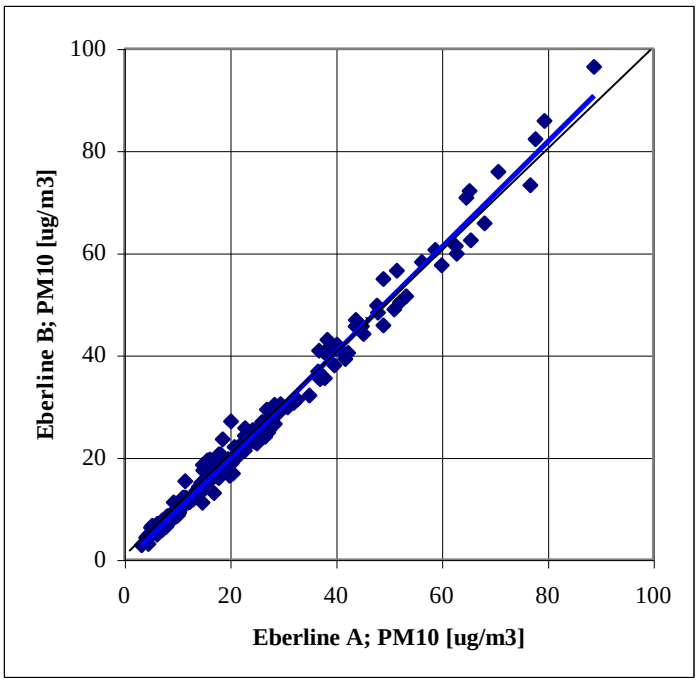

Figure 12. Eberline monitor B compared with monitor A using 23-hour averages.

The EN12341 standard does not deal with automated methods, as mentioned above, but a precision estimate when following this procedure gave the result presented in Table 30. The 95 % confidence limit is well below the upper limit of the standard, i.e. 5 µg/m^3. Table 31 gives the orthogonal regression of the Eberline A daily averages with the corresponding monitor B data. It is seen that there is a small, but significant difference between the two monitors.

Table 30. Precision of the Eberline monitors, relating to the difference between parallel measurements.

Number of samples	Standard deviation µg/m^3	t $_{0.975}$	CL $_{95}$ upper limit µg/m^3
209	1.41	1.97	2.78

Table 31. Orthogonal regression of daily averages from Eberline B monitor with corresponding monitor A data.

Eberline B = a Eberline A + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
209	1.04	1.01	1.06	-0.75	-1.25	-0.25	0.99

Comparisons of the monitors with the reference data set

Figure 13 presents the comparison between the two Eberline monitors and the reference data. As seen the regression line is quite close to the 1:1 line, and very well within the envelope y = x ±10 ug/m3

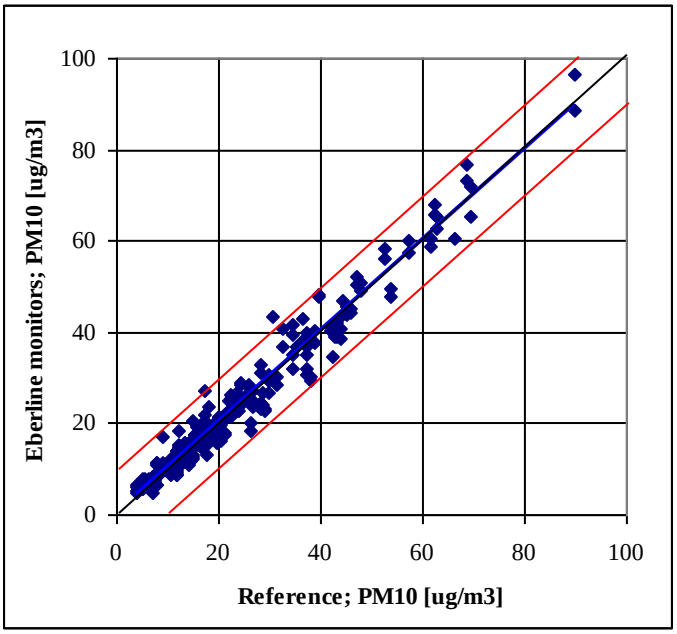

Figure 13. The two Eberline monitors data compared to the PM_{10} reference data set using 23 hour Eberline monitor averages.

Regression between the Eberline and the reference data have been summarized in Table 32. The Table reveals an excellent correspondance and that neither slope nor intercept were significanly different from 1 and 0 respectively.

Table 32. Orthogonal regression of the Eberline monitors daily averages with the reference data set.

Eberline = a Reference + b*

Num. samples	Slope			Intercept μg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
236	1.01	0.98	1.04	0.06	-0.58	0.70	0.98

Table 33 gives the factors to apply on the Eberline averages in order to obtain reference equivalent measurements. The Table additionally gives standard deviations in slope and intercept .

Table 33. Slope, intercept, and standard.

*Reference = a * Eberline + b*

Slope		Intercept $\mu g/m^3$	
a	St.dev. a	b	St. dev. b
0.99	0.01	-0.06	0.32

The Eberline monitor results were stratified on relative humidity as seen in Figure 14. Only winter data have been used in the Figure since few summer data were available. Days with differences larger than 5 $\mu g/m^3$ between the two monitors were excluded from the Figure. The circles represent the averages of the results in intervals of ten per cent width, e.g. for RH 50 – 60 per cent. The number of data in the two lowest intervals in the Figure is well below 10 and the averages are rather uncertain, while the three highest intervals contain 22-16 data. The Figure suggests none, or only a small, dependence of monitor results on the relative humidity during winter conditions when compared with the reference data set.

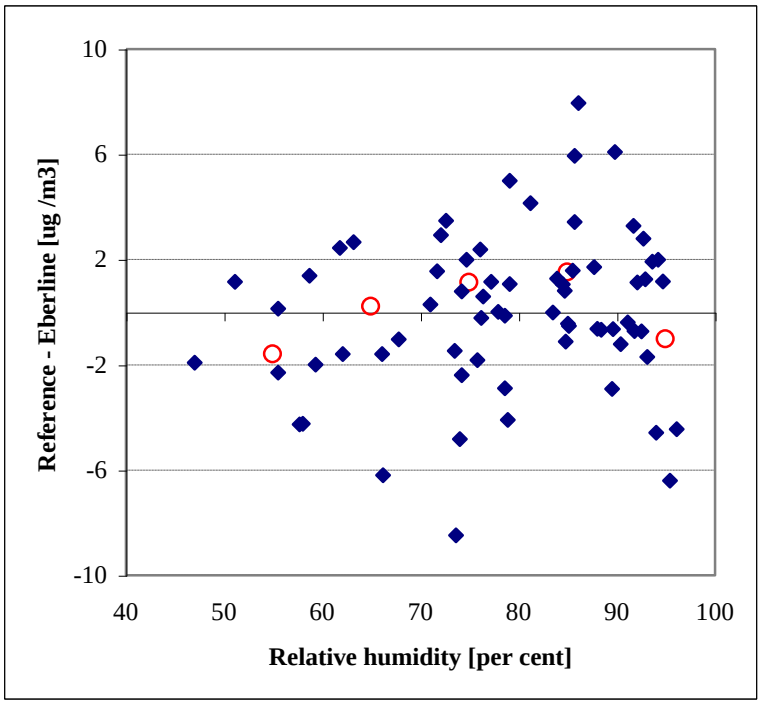

Figure 14 The difference between the reference data and the average of the two Eberline monitors, and the averages in ten percent intervals (circles).

It is however, important to bear in mind that the conditioning of the KFG and high-volume filters before and after exposure can also give a loss or gain of water and a loss of semi-volatile substances during wintertime.

TEOM monitor

Comparisons of the two monitors with nafion driers

Figure 15 compares the two identical TEOM monitors 1 and 2 that are equipped with nafion driers. TEOM 1 give lower PM_{10} concentrations than TEOM 2; the reason for this was not identified. The precision in the measurements was, however, good and there were no outliers in the measurements.

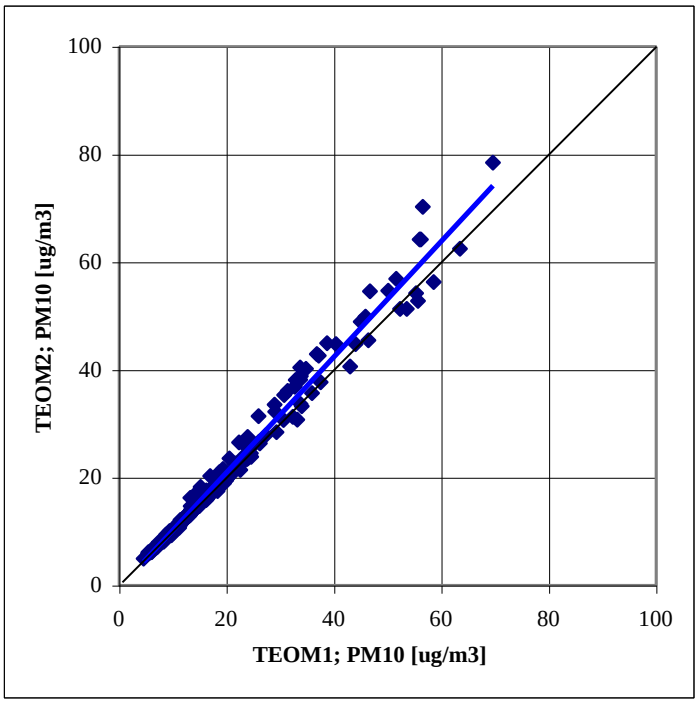

Figure 15. TEOM monitor 2 compared with monitor 1 using 23-hour averages. Both monitors were equipped with nafion driers.

The tests described in EN12341 standard do not deal with the equivalence of automated methods such as TEOM monitors to reference instrumentation. The procedure given for estimating the precision, when applied on the TEOM 1 and 2 results, gave however a good precision for the two monitors that would have been in compliance with the standard, as seen in Table 34. The requirement is that the CL_{95} does not exceed 5 µg PM_{10}/m^3.

Table 34. Precision of the TEOM monitors with nafion driers, relating to the difference between parallel measurements.

Number of samples	Standard deviation µg/m^3	$t_{0.975}$ µg/m^3	CL_{95} upper limit µg/m^3
198	1.65	1.97	3.26

Table 35 further compares the two TEOM monitors with nafion driers by orthogonal regression of the daily averages. The regression line is significantly different from a 1 : 1 line.

Table 35. Orthogonal regression of daily averages from TEOM 2 with corresponding TEOM 1 data.

TEOM 2 = a TEOM 1 + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
198	1.08	1.04	1.13	-0.84	-1.53	-0.15	0.99

Comparisons of the monitor without nafion drier

The third TEOM in the comparison was a standard type TEOM operating at a higher temperature (50 °C). Figure 16 compares the standard type TEOM 3 with TEOM2 with nafion drier. Table 36 and Table 37 give the results of orthogonal regressions between the standard monitor and the nafion drier type.

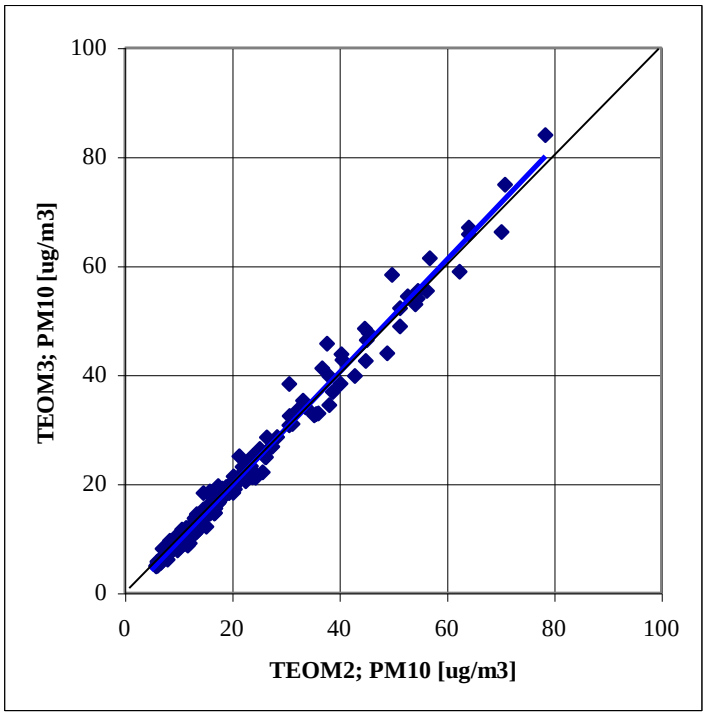

Figure 16. TEOM monitor 3 compared with monitor 2 using 23-hour averages. TEOM 3 was a traditional TEOM running at 50 °C while TEOM 2 was equipped with nafion drier at 30 °C.

Table 36. Orthogonal regression of daily averages from standard type TEOM 3 with corresponding nafion drier type TEOM 1 data.

TEOM 3 = a TEOM 1 + b.*

Num. samples	Slope			Intercept µg/m3		R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95}	

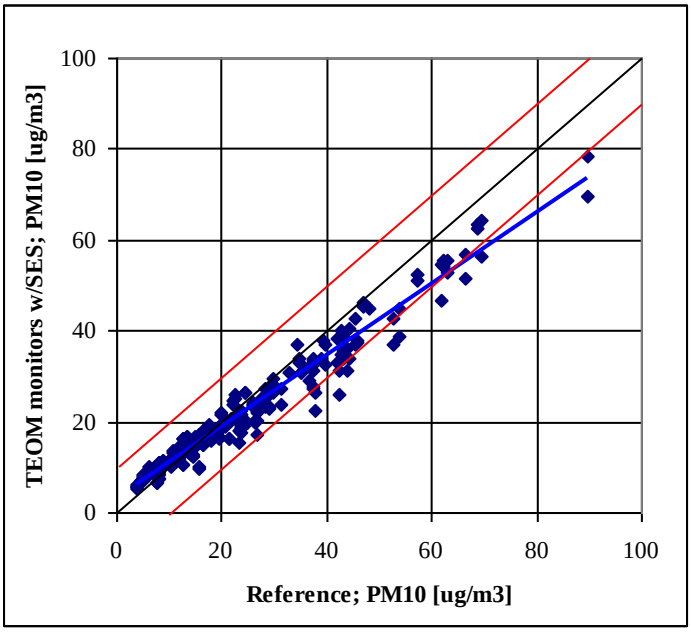

Figure 17. The two nafion drier type TEOM monitors data compared to the PM_{10} reference data set using 23 hour TEOM monitor averages.

Table 38. Orthogonal regression of the nafion drier type TEOM monitors daily averages with the reference data set.

TEOM1 and 2 = a Reference + b*

Num. samples	Slope			Intercept µg/m3			R^2
	A	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
223	0.80	0.77	0.83	2.74	2.17	3.31	0.98

Table 39 gives the factors to apply on nafion type TEOM averages in order to obtain reference equivalent measurements. The Table additionally gives standard deviations in slope and intercept .

Table 39. Slope, intercept, and standard deviations.

*Reference = a * TEOM (nafion drier) + b.*

Slope		Intercept µg/m^3	
a	St.dev. a	b	St. dev. b
1.25	0.02	-3.43	0.42

Comparisons of the monitor without nafion drier with reference data set

Table 40 give a comparison of the standard type TEOM against the reference data.

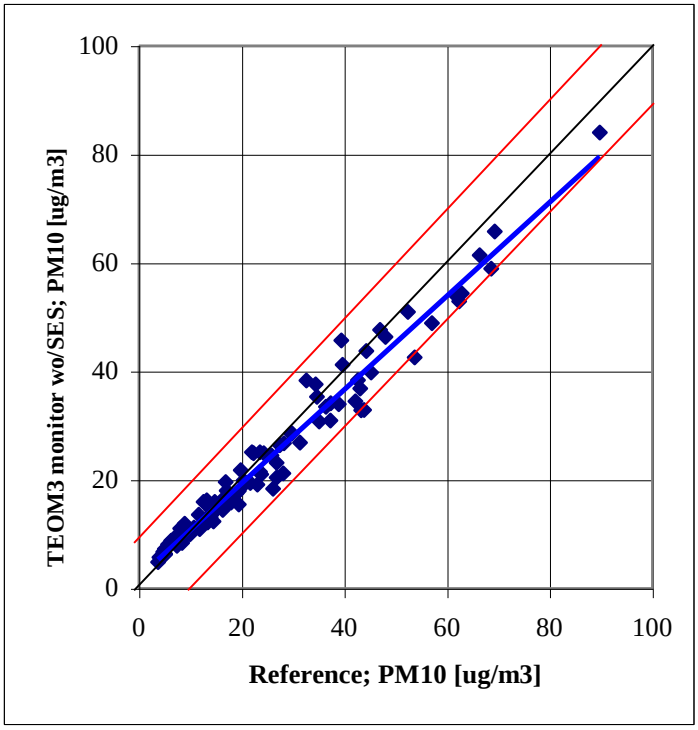

Figure 18. Comparison between the standard type TEOM monitor data and the reference data set.

Table 40. Orthogonal regression of standard type TEOM monitor data with the reference data set.

TEOM3 = a Reference + b*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
93	0.88	0.84	0.91	1.70	0.97	2.43	0.98

Table 41 gives the factors to apply on the standard TEOM averages in order to obtain reference equivalent measurements. The Table additionally gives standard deviations in slope and intercept .

Table 41. Slope, intercept, and standard deviations.

*Reference = a * TEOM 3 (standard type) + b.*

Slope		Intercept $\mu g/m^3$	
a	St.dev. a	b	St. dev. b
1.14	0.023	-1.94	0.46

It is seen that factors are needed to make the TEOM results in good correspondence with the reference instruments. The standard type TEOM deviated less than the nafion drier type from the reference data set in this comparison. The Figures above reveal that the second part of the requirement in EN12341 is violated, but the standard does, as mentioned above, not include automated methods.

The two Figures below compare the difference between the reference data and daily averages (23 hour means) of the TEOM data with relative humidity. The Figures are based on winter data only since much less summer data became available in this comparison. The Figures show that the difference between the reference data and the TEOM monitors increases somewhat with increasing relative humidity. The circles represent the averages of relative humidity in intervals of width ten percent, e.g. for RH 50 – 60 per cent. Number of data in the intervals (circles) in the Figures are between 15 and 26 for the three highest RH intervals, but lower than 10 for the two lowest intervals. Days with differences larger than 5 ug/m3 between the two identical TEOM monitors were not included in Figure 19 in order to exclude the largest TEOM-TEOM differences.

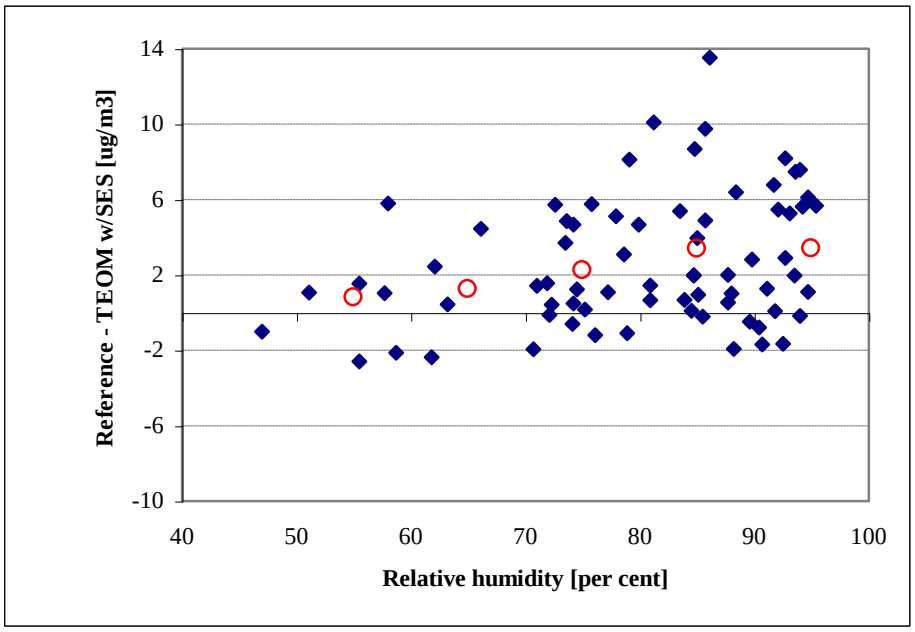

Figure 19. PM_{10} difference between the reference data set and two nafion drier type TEOM monitors as a function of relative humidity.

TEOM data are the average of two monitor daily averages when the difference between the TEOM results are less than 5 ug/m3. Circles give the averages of results within 10 % RH classes.

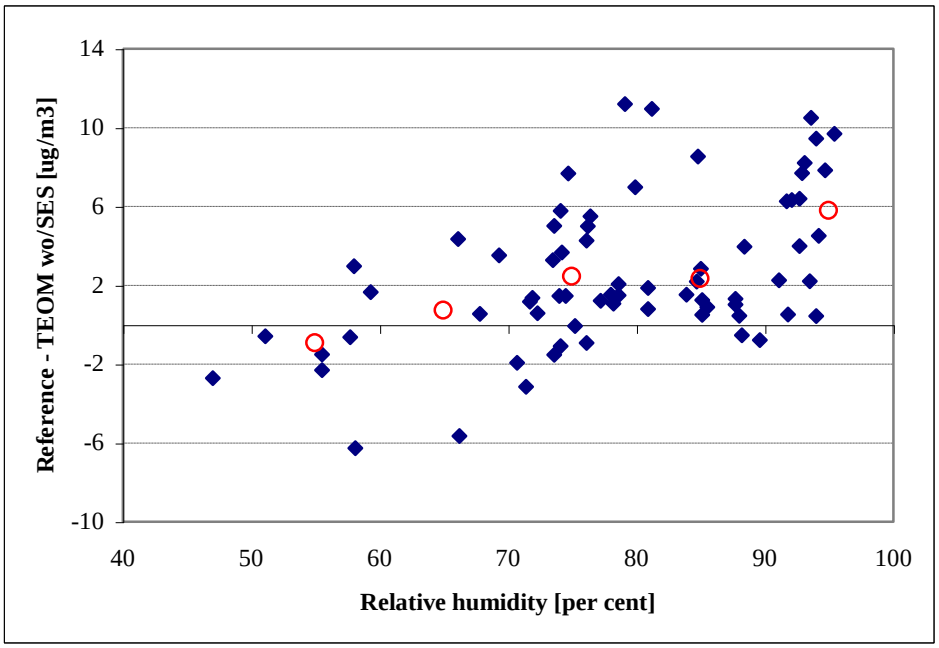

Figure 20. PM_{10} difference between reference data set and standard type TEOM3 as a function of relative humidity. Circles give the averages of results within 10 % RH class.

The loss of water vapour is thought to contribute, together with semi-volatile organics and to some extent ammonium nitrate, to the differences between the reference data and the TEOM data. The results do not suggest large differences with respect to loss of water comparing standard type and nafion drier type TEOM under winter conditions.

As stressed earlier in the report the conditioning of the KFG and High-volume filters before and after exposure can also give a loss or gain of water and of semi-volatile substances, particularly during wintertime.

GRIMM monitor

Comparisons of the two monitors

The GRIMM monitor measured both the PM2.5 and the PM_{10} concentrations, only the latter concentrations have been reported and compared with the other data below. It should be noted that no temperature or pressure corrections have been applied on the GRIMM data, while the reference data have been recalculated, as in the other comparisons above, to 273 deg. K and 1013 hPa.

The results revealed that the monitors operated clearly with large errors at the first two measurement sites, Helsfyr and Sofienberg. These data have therefore been rejected. The measurements from the Elverum site are described below.

Figure 21 compares daily averages (i.e. 23 hour averages) of the two identical GRIMM monitor PM_{10} concentrations from the wood burning site (Elverum). The comparability between the monitors was very good at this site.

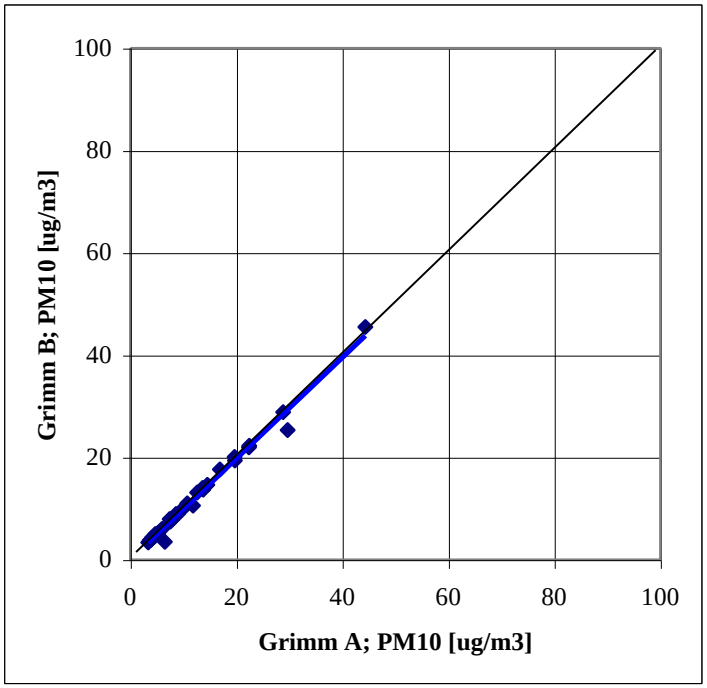

Figure 21. GRIMM monitor A compared with monitor B using 23 hour averages. Data from Elverum wood burning site only.

Table 42 gives the GRIMM precision for the Elverum data estimated following the EN12341 standard. The standard does not deal with automated methods, as mentioned above, but a precision estimate was nevertheless calculated as for the other monitors. The precision is very good as seen from the Table. Table 43 gives the orthogonal regression of the GRIMM A daily averages with the corresponding monitor B data. The good correspondence between the monitors is reflected in narrow confidence limits for slope and intercept.

Table 42. Precision of the GRIMM PM_{10} monitors, relating to the difference between parallel measurements.

Number of samples	Standard deviation $\mu g/m^3$	$t_{0.975}$	CL_{95} upper limit $\mu g/m^3$
35	0.69	2.03	1.39

Table 43. Orthogonal regression of daily averages from GRIMM monitor B with corresponding monitor A data.

GRIMM B = a GRIMM A + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
35	0.99	0.93	1.05	-0.03	-0.60	0.54	0.99

Comparisons of monitors with the reference data set

Figure 22 presents the Elverum GRIMM data sets compared to the reference data. The GRIMM data are the averages of the two monitor results as far as possible. Since the data capture for the monitors was very low, but the precision in the measurements was very high, data with only one operational monitor have also been included in the Figure. About half of the GRIMM data is from one of the two monitors alone, with a total number of 32 data in the Figure.

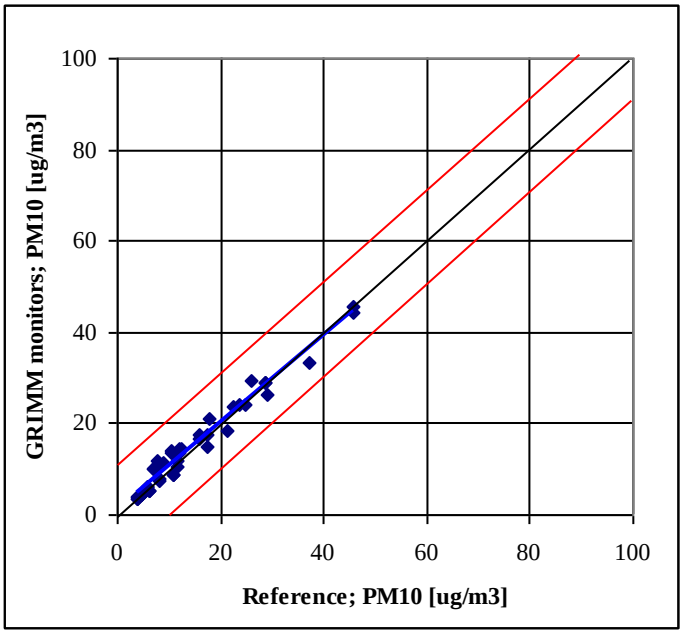

Figure 22. The two GRIMM monitors data compared to the PM_{10} reference data set using 23 hour GRIMM monitor averages. Data from Elverum wood burning site only.

The monitors gave PM_{10} results that compared well with the reference data set. The daily average concentrations were, however, rather low at this site, and they never exceeded 50 µg PM_{10}/m^3 in contrast to the samplers and monitors above where concentrations up 80 – 100 µg PM_{10}/m^3 could be compared.

Table 44 gives the results of a linear regression of the reference data from the wood-burning site at Elverum with the monitor data in Figure 22. The confidence intervals for slope and intercept are wider than for the other samplers and monitors above due to a lower number of data.

Table 44. Orthogonal regression of the GRIMM monitors daily averages (*) from wood-burning site with the reference data set.

GRIMM = a Reference + b*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
50	0.97	0.92	1.01	0.95	0.19	1.71	0.98

(*) with exceptions as explained above

The factors to be applied on the GRIMM data from the wood-burning site in order to obtain results equivalent to the reference set are given in the Table 45.

Table 45. Slope, intercept, and standard deviations (all concentrations below 50 µg/m³).

*Reference = a * GRIMM daily average + b.*

Slope		Intercept µg/m³	
a	St.dev. a	b	St. dev. b
1.03	0.02	-0.98	0.41

ADAM monitor

Comparisons of the monitor and filter data

The measurements consist of both monitor data and PM_{10} mass collected on a filter that was weighed. Figure 23 presents the ADAM monitor results compared to the ADAM filter data. The ADAM monitor was operated only during the winter period at the city background site at Sofienberg, and the number of data available is therefore small compared to the other monitors and samplers.

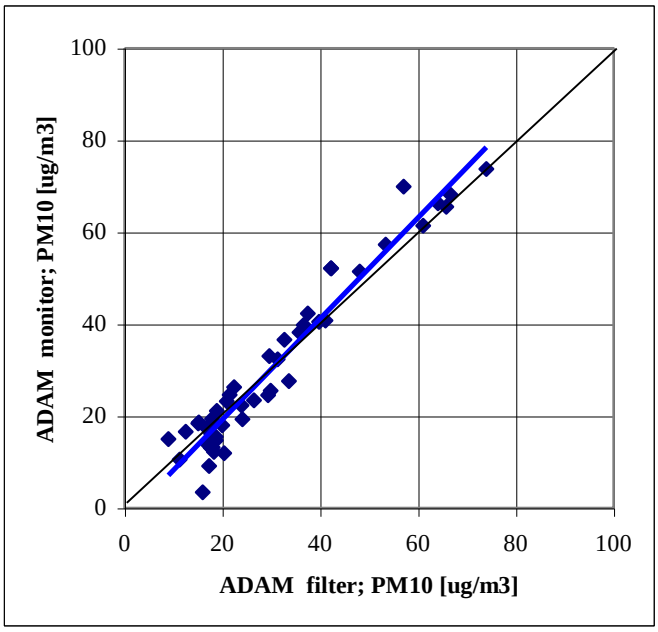

Figure 23. 24 hour averages of the ADAM monitor results compared to the 24 hour ADAM filter results.

Table 46 corresponds with the Figure above and gives the linear regression between the monitor and the filter data. As seen both from the Figure and Table, the regression line deviated from a 1 : 1 line.

Table 46. Orthogonal regression egression of daily averages of ADAM monitor data with corresponding filter data.

ADAM monitor = a ADAM filter + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
47	1.14	1.04	1.23	-3.95	-7.18	-0.71	0.97

Comparisons of the monitor data with the reference data

The ADAM monitor data compared fairly well with corresponding data from the reference data set. The regression line, in Figure 24, has a slope that is not significantly different from 1 as seen from Table 47, but the intercept is different from zero. This highly limited data set indicated a fairly good correspondence between the monitor data and the reference data.

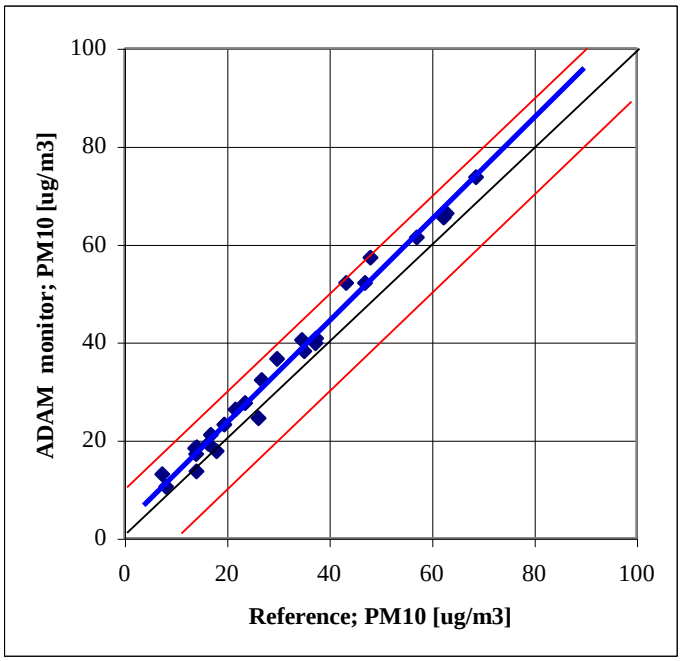

Figure 24. 24 hour averages of the ADAM monitor compared to the PM_{10} reference data set.

Table 47. Orthogonal regression of daily averages of ADAM monitor data with reference data.

ADAM monitor = a Reference + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
26	1.05	1.00	1.10	2.55	0.81	4.29	0.99

In order to obtain results that are equivalent with the reference data in this limited data set the factors in Table 48 should be applied.

Table 48. Slope, intercept, and standard deviations .

*Reference = a * ADAM monitor daily average + b.*

Slope		Intercept µg/m^3	
a	St.dev. a	b	St. dev. b
0.96	0.02	-2.44	0.85

Comparisons of the sampler data with the reference data

The ADAM sampler data compared fairly well with corresponding data from the reference data set. The regression line, in Figure 25, has a slope that is not significantly different from 1 as seen from Table 49, but the

intercept is different from zero. This highly limited data set indicated a fairly good correspondence between the filter data and the reference data.

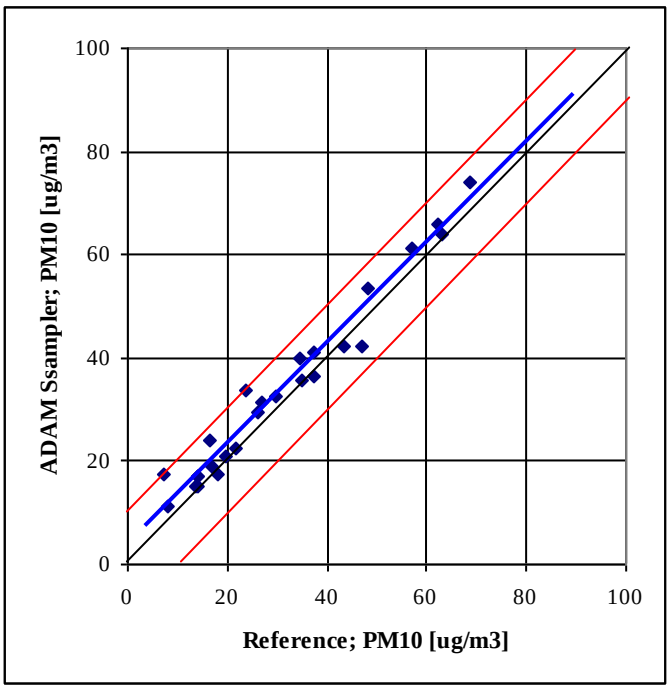

Figure 25. 24 hour averages of the ADAM sampler compared to the PM_{10} reference data set. PM_{10} concentrations in $\mu g/m^3$.

Table 49. Orthogonal regression of daily averages of ADAM sampler data with reference data.

ADAM sampler = a Reference + b.*

Num. samples	Slope			Intercept µg/m3			R^2
	a	CL_{95} low	CL_{95} high	b	CL_{95} low	CL_{95} high	
26	0.99	0.93	1.05	3.27	0.81	5.73	0.98

In order to obtain results that are equivalent with the reference data in this limited data set the factors in Table 50 should be applied.

Table 50. Slope, intercept, and standard deviations.

*Reference = a * ADAM sampler daily average + b.*

Slope		Intercept $\mu g/m^3$	
a	St.dev. a	b	St. dev. b
1.01	0.03	-3.31	1.30

Comparison of all samplers and monitors

The reference methods in this study were the Low-volume sampler, the KFG with PM_{10} inlet and the High volume PM_{10} sampler. These two methods do not give a correct PM_{10} amount in a strict sense since semi-volatile substances both can be gained and lost during the sampling and the filter treatment. The object of a comparison of this kind is, however, to see to which extent a candidate sampler or monitor is able to reproduce the reference methods given the pollution levels. The three candidate samplers all complied with the requirements to an equivalent method for the pollution levels and composition at the sites, as shown earlier in this report.

In order to further compare all samplers' and monitors' performances with one common data set, a data set consisting of all days with all equipments in operation and with valid data was selected. The exceptions in this common data set are the traditional TEOM type (3) that had an extended period with missing data, and the GRIMM and ADAM monitor data were not included in this comparison due to the small amount of valid data.

The results show that the arithmetic averages always were larger than their corresponding medians because the data were lognormal rather than normal distributed. The candidate samplers had all higher 99 percentiles and maximum concentrations than the reference samplers. The Eberline monitor had results similar to the candidate samplers while the TEOM monitors underestimated the highest concentrations. The Partisol sampler seemed to have results somewhat closer to the highest reference data than the candidate instruments.

Since all filter samplers slightly overestimated the PM_{10} concentrations at the levels in this comparison, except for the lowest ones, they will overestimate the number of data exceeding a high percentile. This was also the case for the Eberline monitors in this comparison.
The differences between the two reference samplers have been commented earlier in this report. The results showed that the KFG obtained higher PM_{10} percentiles than the High Volume sampler for most of the values selected.

This comparison is given with more details in Appendix A.

4.3 TSP measurement results

The highest measured concentrations of TSP occurred at the roadside sampling site at Helsfyr. The maximum concentration reached 214 µg TSP/m^3 in February 2001 with the PM_{10} concentration at 90 ug/m3. The ratio between TSP and PM_{10} concentrations varied between 1.5 and 3.6 at this site. The lowest TSP concentrations were found at the wood-burning site in Elverum where the loading in June approximated those of PM_{10} with concentrations below 20 µg /m^3. Although the concentrations here were higher during the winter period, the TSP to PM_{10} ratio was generally about 1. The background city site at Sofienberg had quite low concentrations of PM_{10} during the summer (i.e. the autumn) period with TSP below 40 µg /m^3, and TSP/PM_{10} ratios near 1. The November and December measurements of TSP were, however, generally higher, as were the TSP/PM_{10} ratio.

Figure 26 to Figure 29 present the concentrations of the two reference methods for PM_{10} and their equivalent Partisol sampler together with the TSP concentrations during all measurement periods.

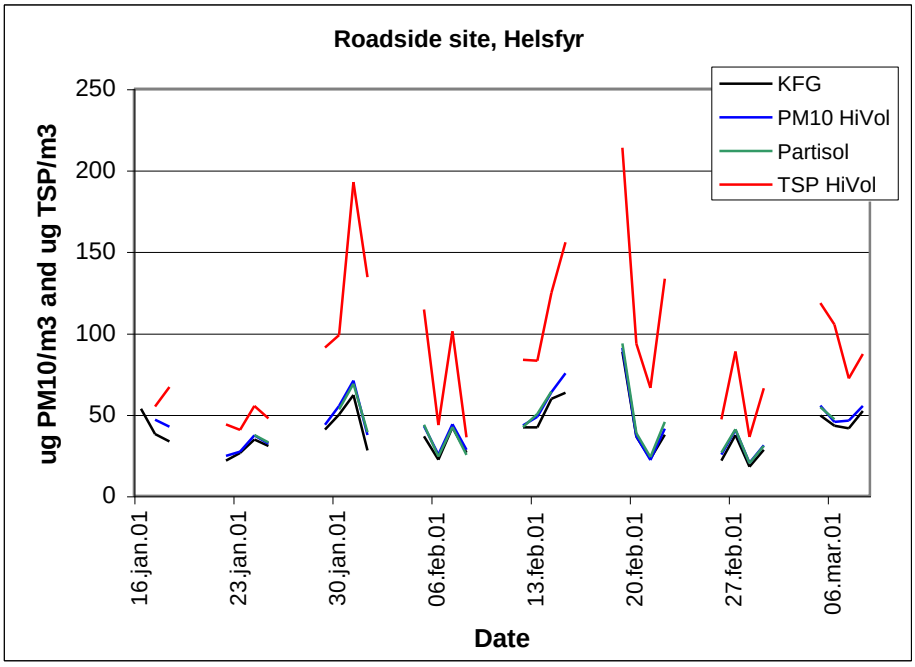

Figure 26. TSP and PM_{10} concentrations at the roadside site.

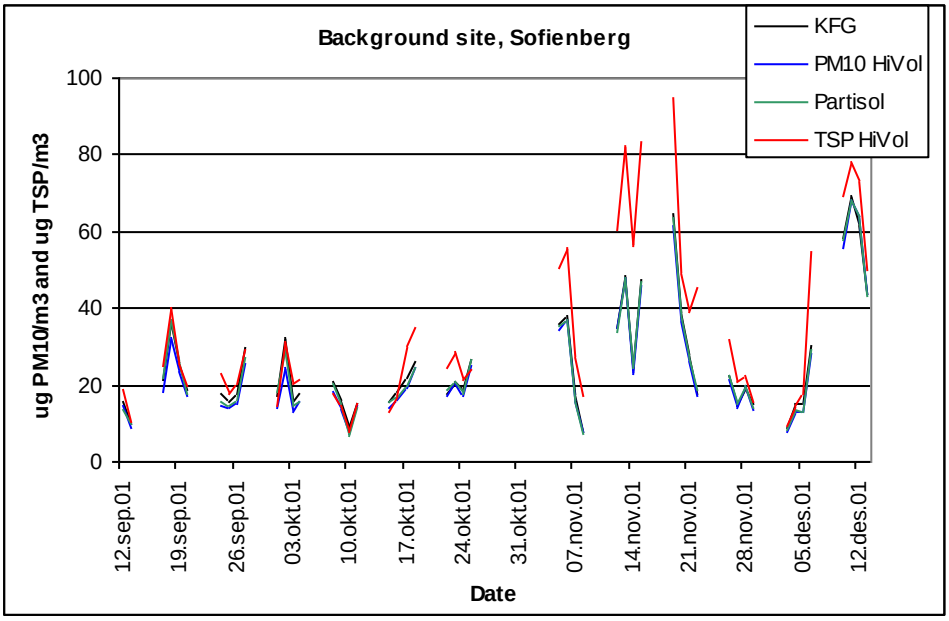

Figure 27. TSP and PM_{10} concentrations at the city background site

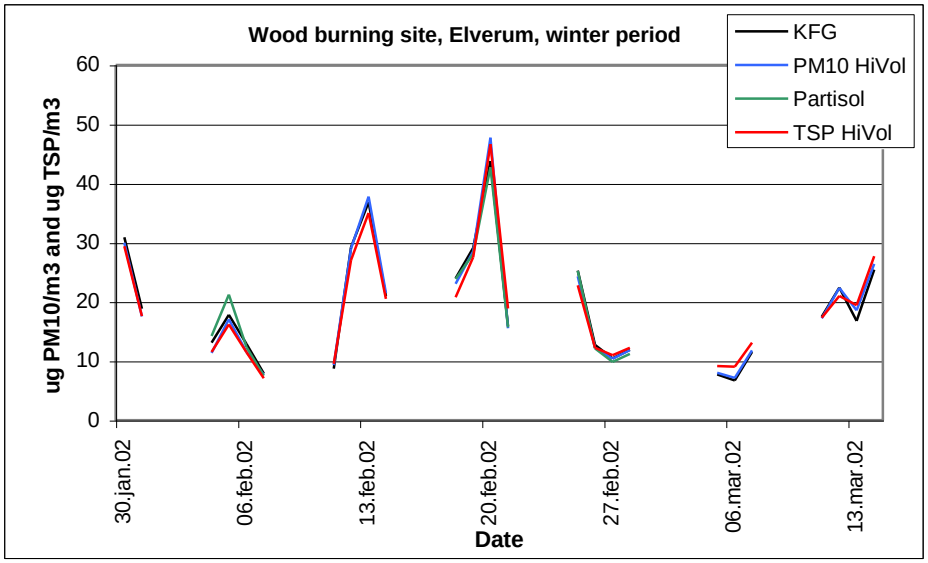

Figure 28. TSP and PM_{10} concentrations at the wood-burning site during winter.

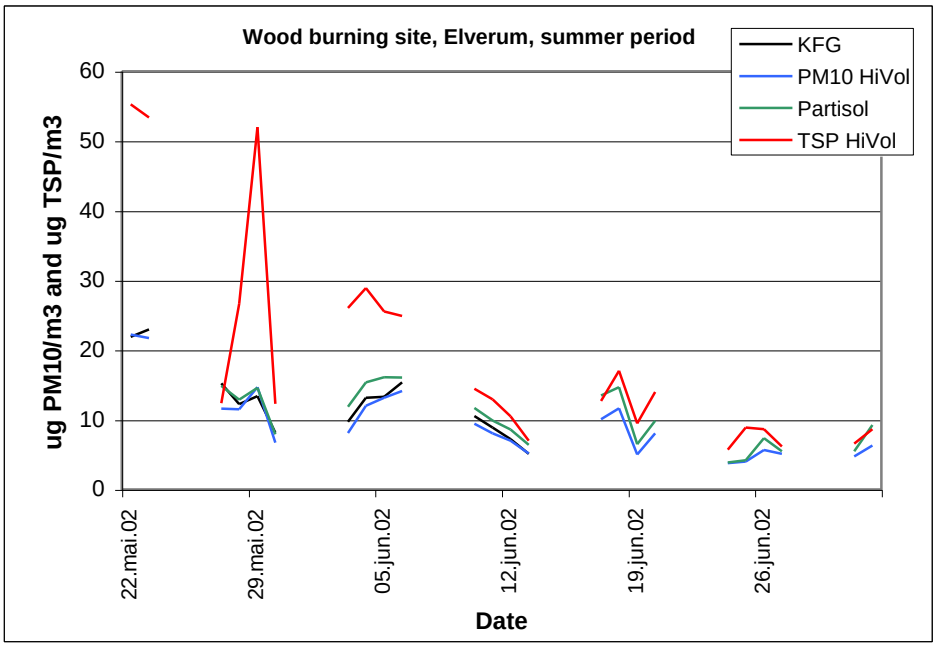

Figure 29. TSP and PM_{10} concentrations at the wood-burning site during summer.

4.4 Aerosol analysis results

Aerosol sources and chemistry

The sources to airborne particles in cities in general are numerous, the most important being exhaust from diesel and gasoline- powered cars with and without catalysts, debris from tires, road and soil dust, burning of wood in fireplaces and stoves, vegetative detritus and pollen, and gaseous pollutants from distant and near sources giving sulphates, nitrates and secondary organic compounds.

A large fraction of the particulate matter consists of organic substances. The soluble inorganic compounds collected at the city background site in Oslo made up for only 25 per cent in average of the total PM_{10} mass, the remaining substance consisting of organic compounds, elementary carbon, water-insoluble inorganic compounds, and water. The relative contributions of these groups have not been investigated in this study.

Ammonia is often related to agricultural activities, but in cities and near major roads vehicle catalysts will be a major source (Baum et al., 2001, Huai et al.,2003). There are yet many unanswered questions with respect to vehicle ammonia emissions, type and characteristics of the individual catalyst seems important, and the way a vehicle is driven does not alone explain the emissions. It seems, however, clear that aggressive driving increases ammonia emissions. Reduced sulphur contents in fuels may

also have increased the ammonia emissions since sulphur-containing substances poison the catalyst's reaction sites. Vegetation and soil surface can be both a sink and a source to ammonia depending upon many factors including surface wetness and relative humidity (Wyers et al., 1998, Neftel et al., 1998). Ammonia will react with acidic substances, e.g. to form ammonium sulphate or ammonium nitrate, and is readily dissolved in non-alkaline water films.

The main source to sodium, chloride and magnesium is marine salts. The magnesium concentrations during winter at the roadside site were generally much lower than expected from sea-water composition. The reason for this is the highway de-icing by snowfall with marine salts that were richer in sodium chloride than sea-water is. The content of sodium chloride was 99.3 % (Rieber Salt, 2002) with small concentrations of other components. Particularly the magnesium concentrations were low, less than 0.1 % compared to 3.9 % in sea water. This is probably due to the large difference in solubility between sodium and magnesium chlorides, which becomes important as water evaporates during the production of solid marine salts.

The potassium concentrations in particulate matter will during winter have contributions from wood burning in stoves, and in summer from vegetation. Calcium is correlated to ammonia at highways and is related to traffic, road and soil dust. Non-catalyst gasoline engines (Schauer et al., 2002) and diesel engines (Wang et al., 2003), and to a less extent catalyst engines, emit crust metals such as calcium, aluminium, iron, and zinc in their exhaust besides other elements and organic and elemental carbon.

Roadside measurements at Helsfyr, Oslo

The three secondary pollution components ammonium, sulphate, and nitrate collected on the particle filter were well correlated through the measurement period. The nitrate concentrations were generally lower than those of the other two components. The sums of nitrate and sulphate concentrations were always lower than the ammonium concentrations and the nitrate and sulphate were to a large extent bound to metal ions. The ratio ammonium to the sum of sulphate and nitrate varied from 0 to 75 per cent on equivalent basis, with a median at 57 per cent.

The gaseous ammonia concentrations were usually, but not always, higher than the particulate ammonium concentrations. Both the ammonia maximum and the median at Helsfyr were higher than the corresponding statistics from the two other sites, supporting that the ammonia main source is related to vehicles. The maximum nitric acid concentration at Helsfyr on the other hand, was lower than the corresponding maxima

seen at two other sites, which could fit with the high ammonia concentrations in Figure 30.

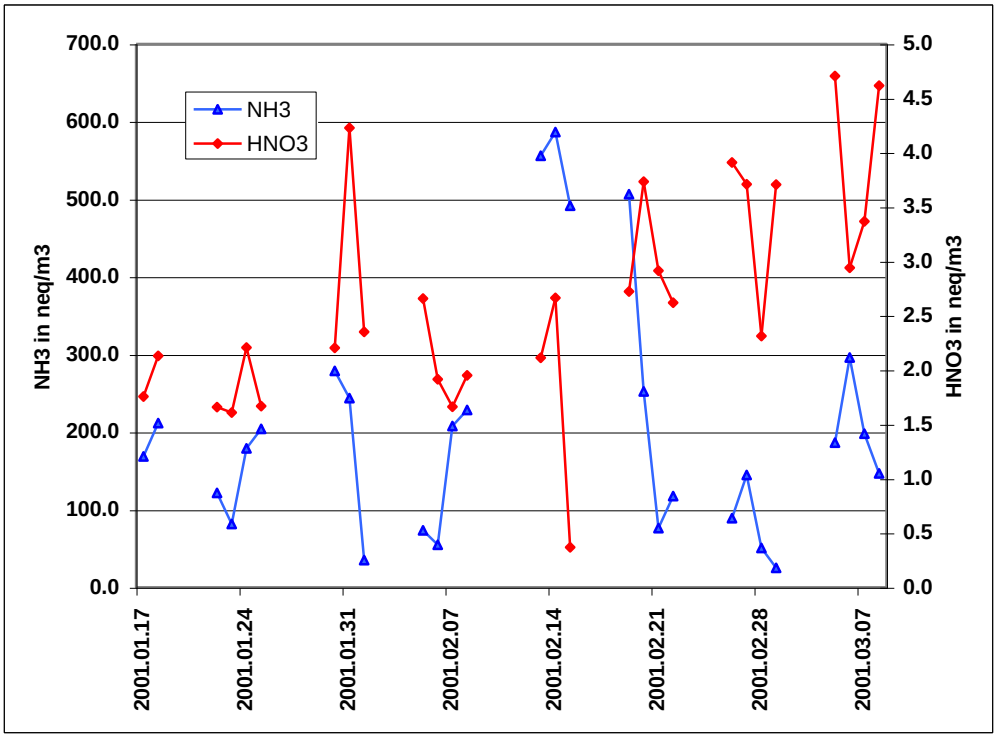

Figure 30. Daily averages concentrations of gaseous NH_3 and HNO_3 measured at the roadside site at Helsfyr.

Assuming that ammonium sulphate is preferred to ammonium nitrate, there were three days only with NH_4NO_3 concentrations larger than zero with a maximum at 2.2 µg NH_4NO_3/m3 the 30th January. This ammonium nitrate concentration did, however, not explain the difference between reference measurement and the TEOM monitor data. Other semi-volatile substances, most likely water and organic compounds contributed therefore to the reference aerosol mass.

De-icing of the highway near the site and sea salt carried from more distant sources both contributed to the concentrations of sodium, chloride, magnesium and other elements at the site. Estimation of the relative PM_{10} mass contributions, based on the average concentrations of the elements, gave 8 and 3 µg/m^3 for de-icing and sea spray respectively during the measurements at Helsfyr. The day-to-day variation was large with a maximum de-icing contribution well above 20 µg/m^3 the 7th February when the HiVol sampler measured 44.1 µg PM_{10}/m^3.

The contributions of marine salts to potassium and calcium concentrations were small at Helsfyr, and the main sources are different ones as

explained above. The three sea salt components are well correlated, and their time series are different from the anthropogenic compounds above reflecting the different sources. The chloride concentrations were close to those expected from sea salts, little or no chloride seems to have been lost as hydrogen chloride.

Background city measurements at Sofienberg, Oslo

The concentrations of all components were generally lower at this background site than at the roadside site at Helsfyr. The difference were small for nitric acid that frequently had concentrations at the detection limit at all sites, and also for potassium. The largest differences are as expected seen for the traffic related compounds, the marine salts and ammonia. The sodium concentrations were nearly ten times higher at the roadside site than at this city background site, and the ammonia level the double. Calcium, considered to have traffic-related sources, also had a lower median concentration at Sofienberg than at the roadside site, but had a higher maximum due to an inversion that occurred at the end of the measurement period.

During the autumn period September-October the components had little co-variation except for the usual correspondence between the three marine salt components, and between the three secondary anthropogenic particulate components ammonium, sulphate, and nitrate. The winter period November – December had two inversion periods, 16th –20th November, and from the 9th December. The highest concentrations of all components except for nitric acid occurred during the inversion periods. Nitric acid had the lowest concentrations during the inversions. The particulate components ammonium, sulphate, and nitrate reached their maximum during the second inversion period, but were rather low during the first one.

Potassium was well correlated with excess calcium during winter, behaving like a primary pollutant and being linked to wood burning in stoves. The correspondence between calcium and potassium was weak during autumn.

The ratio of sodium to magnesium concentrations corresponded reasonable well to that expected from a sea salt composition, and were generally lower than at the roadside site; salt from road de-icing was less important at the background site than at the roadside site. In the autumn period the median of the chloride that had been converted to hydrochloric acid, as calculated from the chloride and sodium content in sea water was 40 per cent. The loss during winter was much lower.

Ammonium nitrate had a rather small influence on the monitor data during the autumn period. The maximum concentrations may have been reached the 27th September and 25th October with about 2 ug NH_4NO_3/m^3. During the winter period at the 13th to 14th December ammonium nitrate was most likely present; estimates suggest 5- 6 ug NH_4NO_3/m^3 this day during an inversion.

Measurements at Elverum wood burning site

Concentrations of all components except for nitric acid and potassium were lower at Elverum than at the two other sites.

The ammonium concentrations on the particle filter were in nearly all cases less than the sum of sulphate and nitrate concentrations in neq/m3. Ammonium, sulphate, and nitrate in particles were generally well correlated with each other, but about half of the sulphate and nitrate were chemically bound to metals on a molecular basis The three components above were, like at the two other sites, not correlated with gaseous ammonia.

The excess potassium concentrations at Elverum were slightly higher than the winter measurements in Oslo. The major source to excess potassium during winter in this country is thought to be wood burning, e.g. Schauer et al. (2001) measured the potassium fraction in the fine particle emissions from burning pine to 3 per cent. It is, however, well known that the potassium fraction in particles varies with wood and stove types. Figure 31 presents the minimum temperature at Elverum during winter and the corresponding potassium concentrations (in neq/m^3). Its seen that the potassium concentrations were relatively high when the minimum temperatures were low, and vice versa. The maximum potassium concentration, 9 neq/m^3 (0.34 μg K/m^3), occurred at the coldest measurement day.

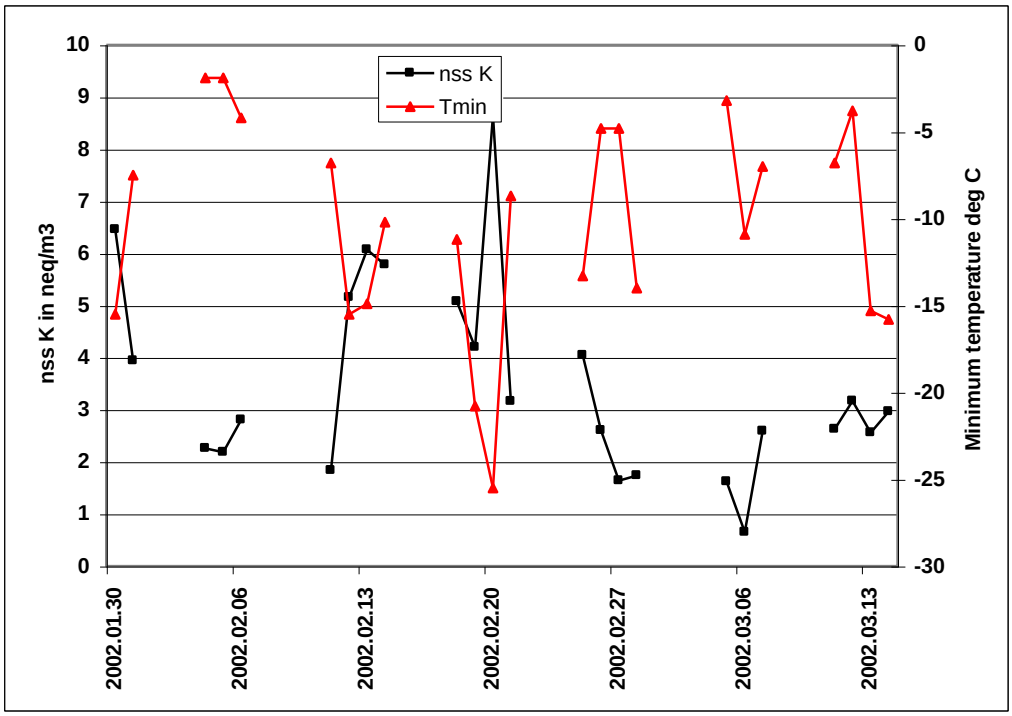

Figure 31. Daily excess potassium averages and minimum temperature at the site in Elverum during the winter period. Potassium concentrations are given in nanoequialents/m^3 and the minimum temperature in ^oC.

The ammonium nitrate result that occured 7th March apparently is an example of a relatively high concentration, 3 ug NH_4NO_3/m^3 together with a correspondingly low ammonium sulphate.

The chloride concentrations due to sea spray strongly decreases with the distance from sea, and are low at Elverum during summer. The median of chloride lost as hydrochloric acid during winter was 40 per cent, and 85 per cent during summer. The minimum lost chloride during summer was 60 per cent.

Ammonium nitrate in PM_{10}

The ammonium nitrate contribution to the PM_{10} mass was mostly negligible as Figure 32 shows. Elevated concentrations were seen on a few occasions only, the highest concentration at 6 µg NH_4NO_3 /m^3 being measured at the Oslo city background site in December 2001. The NH_4NO_3 estimates were based on the assumption that ammonium nitrate will be formed only when no sulphuric acid is present. The ammonium nitrate may have been transported to the sampling site from distant locations, but may also have been generated locally to the extent that nitric acid has been available.

The measured nitric acid concentrations during the measurement period were, however, negligible and much lower than the particulate nitrate concentrations. The measured ammonia concentrations were usually in a very large excess to the measured nitric acid concentrations.

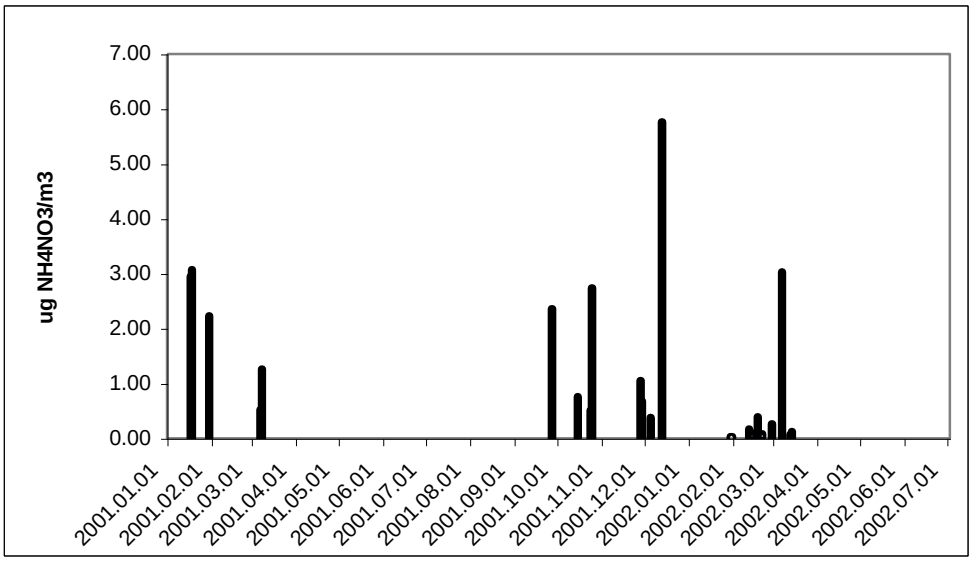

Figure 32. Estimated ammonium nitrate in PM_{10} during the measurements.

The ammonium nitrate concentrations were too small to give significant changes in the regressions of automatic methods with heated inlets to the reference methods, which were given earlier in this report.

It should be kept in mind that the equilibrium between particulate ammonium nitrate and the two gaseous components will change even for the reference methods, and ammonium nitrate lost, when filters are conditioned after exposure before weighing in the laboratory.

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Appendix A. Comparison of all samplers and monitors

Comparison of all samplers and monitors

The reference methods in this study were the Low-volume sampler, the KFG with PM_{10} inlet and the High-volume PM_{10} sampler. These two methods do not give a correct PM_{10} amount in a strict sense since semi-volatile substances both can be gained and lost during the sampling and the filter treatment. The object of a comparison of this kind is, however, only to see to which extent a candidate sampler or monitor is able to reproduce the reference methods given the pollution levels at the sites.

The Partisol Plus Model 2025 has previously proven to be an equivalent method to the gravimetric reference method (Charron et al., 2004). The other three samplers complied with the requirements to an equivalent method for the pollution levels and composition at the sites, as shown earlier in this report.

In order to further compare all sampler's and monitor's performances with one common data set, a data set consisting of all days with all equipments in operation and with valid data was selected. The exception in this common data set is the traditional TEOM type (3) that had an extended period with missing data. The common data set contains 75 corresponding measurements from all devices, but 64 data only for the traditional TEOM. The GRIMM and ADAM monitor data were not included in this comparison due to the small amount of valid data.

Simple statistics have been calculated and are presented in Table 51 and Table 52 and in Figure 33 - Figure 1.

Table 51 gives the percentiles, arithmetic averages, and the minimum and maximum concentrations obtained with each single method in the common data set.

With respect to the central tendency in the data set Table 51 shows that the averages always were larger than their corresponding medians due to skew distributions that are lognormal rather than normal. The differences between the maximum concentrations and the 99 percentile were about 15 $\mu g/m^3$ for the reference samplers and about 20 $\mu g/m^3$ for some of the other samplers. It is also important that all candidate filter samplers had higher 99 percentiles and maximum concentrations than the reference samplers. Figure 33 – Figure 35 presents some of the percentiles. The Figure shows that the candidate filter samplers underestimated the smallest concentrations and the largest were overestimated when compared to the reference data. The opposite was the case for the TEOM monitors while the Eberline results resembled the filter samplers'.

It is useful to inspect more closely the differences between the statistics from each sampler and monitor and the reference set as presented in Table 52. Except for the 1 to 5 per cent of the lowest concentrations (and the 25 per centile for the IVL sampler) all candidate filter samplers obtained higher percentiles than the reference data set. The monitors tended to give slightly higher concentrations than the reference set for the 5 per cent lowest data.

Table 51. Percentiles, arithmetic average, minimum and maximum of the reference samplers, KFG and the High volume PM_{10} sampler, and other samplers and monitors. All data are 23 hour averages sampled from 13 H one day to 12 H the next day. The data set contains 75 corresponding measurements with no missing data, except for TEOM 3 that have 64 measurements. Units are μg PM_{10}/m^3.

	KFG	HIVOL PM_{10}	PARTISOL	NILU A	NILU B	GENT A	GENT B	IVL A	IVL B	EBER 1H	EBER 2H	TEOM 1H	TEOM 2H	TEOM 3H
min	5.1	5.2	6.4	1.5	4.7	4.9	5.1	4.0	5.2	6.1	6.7	6.6	6.8	6.4
1 %	6.7	6.3	6.5	4.5	5.6	5.4	6.1	6.1	6.2	6.4	6.7	6.9	7.2	7.3
5 %	8.5	7.7	8.1	8.5	8.3	8.3	8.2	7.4	7.8	8.6	8.4	9.7	9.8	9.0
10 %	10.5	9.7	11.4	11.1	10.5	10.3	10.7	10.2	10.7	10.4	9.9	10.2	10.6	11.0
25 %	14.8	13.0	14.5	14.2	14.4	14.4	14.9	12.9	13.1	14.3	13.6	14.1	14.0	14.4
50 %	18.7	17.2	19.9	19.6	20.4	20.5	21.3	18.5	19.0	19.1	18.1	18.4	18.2	18.9
average	25.7	25.1	26.1	26.3	26.8	26.9	26.8	25.2	25.7	25.7	25.6	22.9	23.6	24.5
75 %	35.3	34.5	34.8	35.9	36.6	35.2	37.5	36.1	36.2	35.8	33.7	30.0	31.0	33.0
90 %	46.2	48.2	49.2	48.8	51.3	51.3	49.5	48.2	48.1	47.8	49.2	38.3	44.1	46.5
95 %	60.1	61.9	63.4	63.6	64.1	64.8	64.5	62.6	63.2	61.6	61.2	53.3	53.2	53.8
99 %	74.3	74.0	74.7	78.0	78.3	79.3	76.2	75.2	76.5	79.9	79.3	65.1	66.6	68.2
max	88.5	90.9	93.5	99.3	95.2	100.0	96.7	97.3	99.6	88.8	96.4	69.7	78.4	84.0

The Partisol and the IVL samplers obtained 90 to 99 percentiles between 0.5 to 2.5 $\mu g/m^3$ higher than the reference. The NILU and Gent samplers obtained somewhat higher corresponding percentile values; 1.5 to 4.0, and 2.0 to 5.0 $\mu g/m^3$ respectively.

Since all filter samplers slightly overestimated the PM_{10} concentrations at the levels in this comparison, except for the lowest ones, they will overestimate the number of data exceeding a high percentile. This was also the case for the Eberline monitors in this comparison while the TEOM monitors need a correction.

Table 52. Differences in corresponding statistics from each sampler and monitor and the statistics from the reference set. The data set is identical to that in Table 51. Units are μg PM_{10}/m^3.

	PARTI SOL	NILU A	NILU B	GENT A	GENT B	IVL A	IVL B	EBER 1H	EBER 2H	TEOM 1H	TEOM 2H	TEOM 3H
min	1.2	-3.7	-0.5	-0.3	-0.1	-1.2	0.0	0.9	1.5	1.4	1.6	1.2
1 %	0.0	-2.0	-0.9	-1.1	-0.4	-0.4	-0.3	-0.1	0.2	0.4	0.7	0.8
5 %	0.0	0.4	0.2	0.2	0.1	-0.7	-0.3	0.5	0.3	1.6	1.7	0.9
10 %	1.3	1.0	0.4	0.2	0.6	0.1	0.6	0.3	-0.2	0.1	0.5	0.9
25 %	0.6	0.3	0.5	0.5	1.0	-1.0	-0.8	0.4	-0.3	0.2	0.1	0.5
50 %	1.9	1.6	2.4	2.5	3.3	0.5	1.0	1.1	0.1	0.4	0.2	0.9
average	0.7	0.9	1.4	1.5	1.4	-0.2	0.3	0.3	0.2	-2.5	-1.8	-0.9
75 %	0.0	1.1	1.8	0.4	2.7	1.3	1.4	1.0	-1.1	-4.8	-3.8	-1.8
90 %	2.0	1.6	4.1	4.1	2.3	1.0	0.9	0.6	2.0	-8.9	-3.1	-0.7
95 %	2.4	2.6	3.1	3.8	3.5	1.6	2.2	0.6	0.2	-7.7	-7.8	-7.2
99 %	0.5	3.8	4.1	5.1	2.0	1.0	2.3	5.7	5.1	-9.1	-7.6	-6.0
max	3.8	9.6	5.5	10.3	7.0	7.6	9.9	-0.9	6.7	-20.0	-11.3	-5.7

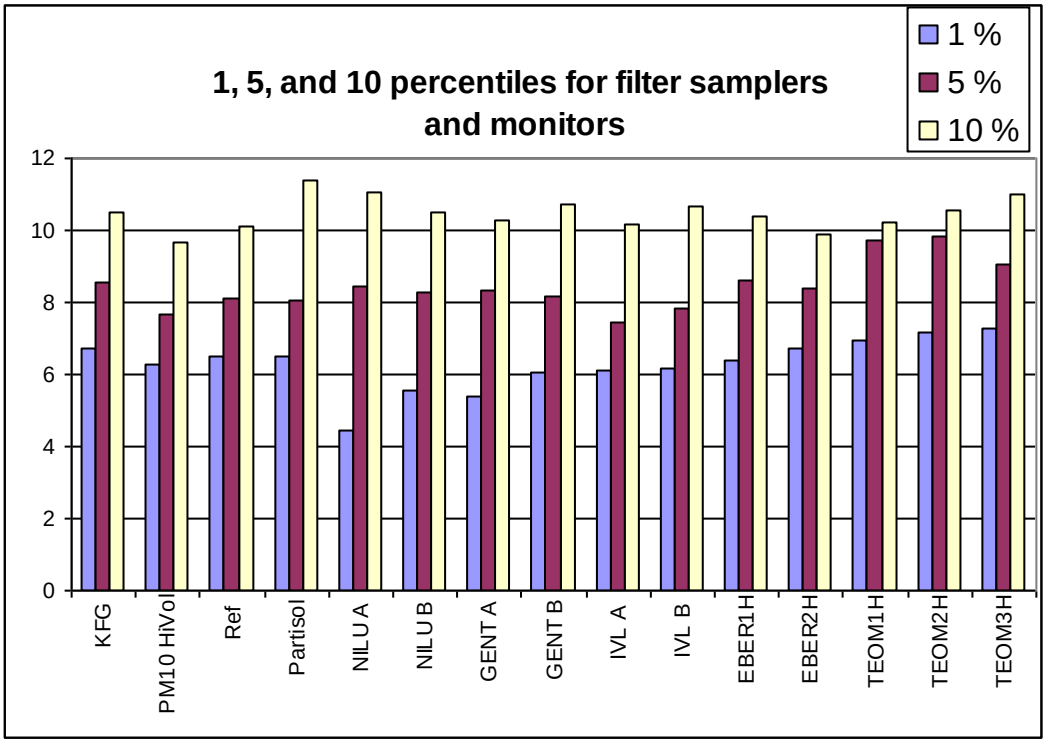

Figure 33. 1, 5, and 10 percentiles for filter samplers and monitors

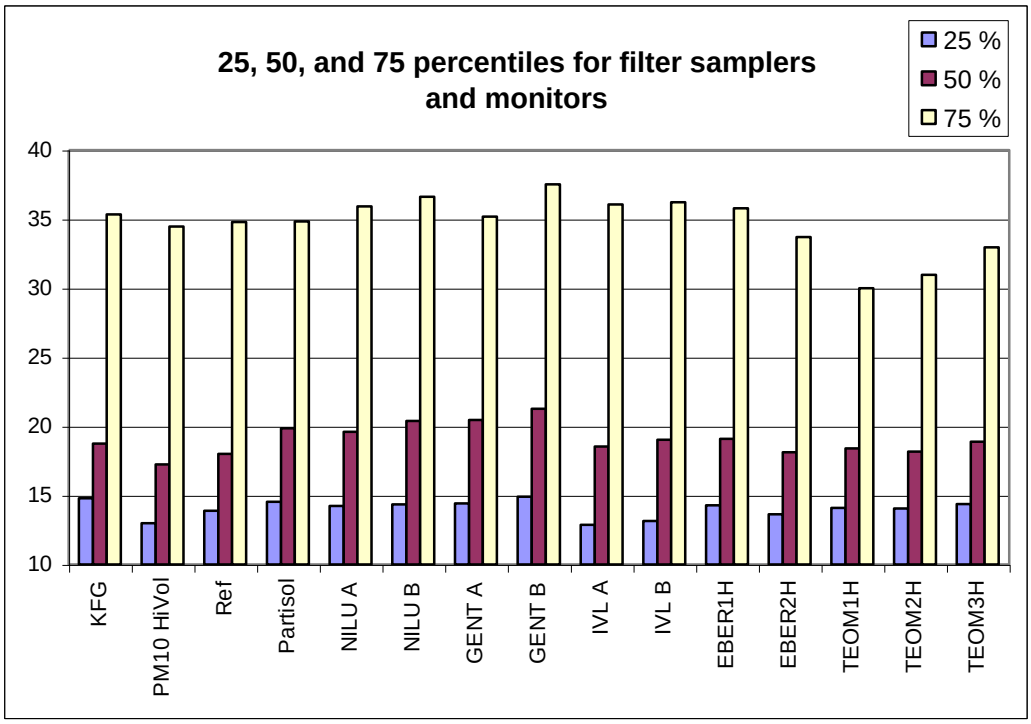

Figure 34. 25, 50, and 75 percentiles for filter samplers and monitors

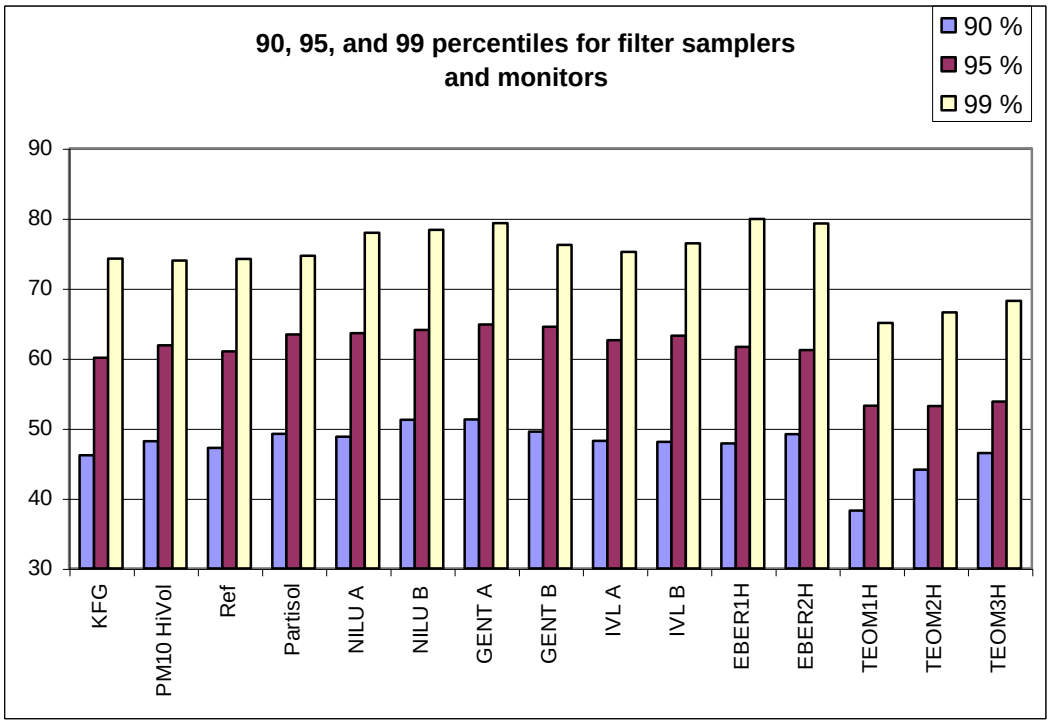

Figure 35. 90, 95, and 99 percentiles for filter samplers and monitors

The differences between the two reference samplers were commented in Section Reference samplers above. The Table and Figures above show

that the KFG obtained higher PM_{10} percentiles than the High Volume sampler for most of the values selected. The high volume sampler had, however, 90- and 95-percentiles that were 2 $\mu g/m^3$ higher than the KFG.

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ABSTRACT

This report presents the results from a PM_{10} field intercomparison exercise performed in Norway. The exercise included several sampelrs and automatic monitors as candidate instruments as well as the Andersen high volume and Kleinfiltergerát (KFG / Low volume) samplers as reference instruments. The exercise was performed according to the European standard EN12341.

NORWEGIAN TITLE

Sammenlignende målinger av svevestøv med PM10 måleinstrumenter

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Intercomparison	PM_{10}	EN12341

ABSTRACT (in Norwegian)

Denne rapporten presenterer resultatene fra en sammenlignende feltmåling av PM_{10}-instrumenter utført i Norge. Sammenligningen omfattet flere prøvetakere og automatiske instrumenter som kandidater samt en Anderesen Høyvolum og en KFG lavvolum prøvetaker som referanseinstrumenter. Sammenligningen ble utført i henhold til den europeiske standarden EN12341.

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