NORDIC WORKING PAPERS

Measurements of emissions of EC, OC and other pollutants from residential wood combustion in the Nordic countries

Staffan Carlsson, Lennart Gustavsson, Mathias Johansson, Henrik Persson, Jes Sig Andersen, Max Bjerrum, Kim Sig Andersen and René Borch Hvidberg

<http://dx.doi.org/10.6027/NA2017-920>
NA2017:920
ISSN 2311-0562

This working paper has been published with financial support from the Nordic Council of Ministers. However, the contents of this working paper do not necessarily reflect the views, policies or recommendations of the Nordic Council of Ministers.

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Jes Sig Andersen, Kim Sig Andersen, René Borch Hvidberg (TI)

Abstract

Measurements of Elemental Carbon (EC), Organic Carbon (OC) and other pollutant emissions from residential biomass combustion representative of the Nordic countries

An experimental survey of emissions of BC and PM_{2,5} emissions from domestic biomass combustion in the Nordic countries has been made. Measurements have been made on ten central heating boilers and nine residential heaters. BC emissions expressed as Elemental Carbon (EC) varied between 0,5 and 35 mg/MJ for boilers and between 3 and 122 mg/MJ for heaters. PM_{2,5} emissions varied between 14 and 1975 mg/MJ for boilers and between 35 and 821 mg/MJ for heaters. As could be presumed, the emissions from modern boilers with advanced combustion technology were much lower than from older boilers with simpler technology.

Key words: Black Carbon, residential biomass combustion, emissions, Nordic countries

SP Sveriges Tekniska Forskningsinstitut
SP Technical Research Institute of Sweden

SP Report 2016:06
ISBN 978-91-88349-13-2
ISSN 0284-5172
Borås 2016

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Preface

This work has been conducted within the Nordic Council of Minister's project "Improved emission inventories of SLCP". SLCP means Short Lived Climate Pollutants, and include Black Carbon (i.e. "soot") from combustion sources. In this project, headed by IVL, Swedish Environmental Research Institute, all five Nordic countries work together in three areas: Background analysis, Measurements and Abatement measures. The Measurements part was as a result of the Background analysis focused on creating a significantly improved database for EC/OC emissions as well as PM_{2,5} emissions from domestic biomass combustion. The work was assigned to SP Technical Research Institute in cooperation with Teknologisk Institut, Danmark, and is reported here.

SP and TI want to express their appreciation of being chosen to execute this project. We also want to thank IVL for interesting and constructive discussions during the project. We do hope that the results will contribute to a better understanding of sources for Black Carbon emissions in the northern part of Europe (and we do want to contribute further to this knowledge building).

Summary

The aim of this project has primarily been to significantly expand the existing database for BC and PM_{2,5} emissions from domestic biomass combustion in the Nordic countries. The data achieved will form the basis for developing more accurate emission factors for these parameters. Measurements have been made on ten domestic central heating boilers as well as on nine residential heaters for installation in the living area. Within both groups, units for log wood fuel as well as for wood pellets were represented. Also, tests on one boiler for wood chip burning was made. The test objects were chosen in order to represent different technologies and ways of use, together forming a representative array of products in use and on the market. The measurements were made at standardized operating conditions, including both nominal heat load and part load and also in some cases fuels with different moisture contents.

Determination of Elemental Carbon (EC) and Organic Carbon (OC) was made by sampling on filters after dilution of the flue gases with air and subsequent thermal-optical analysis of the filters according to the NIOSH Protocol 870. Also, the sampling filters were analyzed with a Magee OT 21 Aethalometer for Black Carbon. PM_{2,5} was determined gravimetrically on the same filters before EC/OC analysis. In addition, unburnt gaseous hydrocarbons as TOC, methane and NMVOC as well as O₂, CO₂ and CO in the flue gases were continuously measured. For each test of a boiler or residential heater at a defined operating condition, 3-7 sampling periods of 30 min duration each on separate filters were used. This was made in order to track varying emission levels during the combustion cycle, and also expand the data base as much as possible.

The EC emissions from **residential heaters**, expressed as average values over each test at constant operating conditions varied between 3 and 122 mg/MJ. The mean value of all tests was 32 mg/MJ. The OC emissions varied between 6 and 441 mg/MJ, and the mean value was 76 mg/MJ. For **domestic central heating boilers**, the EC emissions as average values were between 0,5 and 35 mg/MJ; the mean value being 11 mg/MJ. The OC emissions varied between 6 and 776 mg/MJ, and the mean value was 96 mg/MJ. When boiler data are divided into modern log wood boilers, old log wood boilers, pellet boilers and a wood chips boiler, the mean values for EC emissions were 5, 24, 8 and 7 mg/MJ respectively, and for OC emissions they were 15, 290, 22 and 109 mg/MJ.

PM_{2,5} emissions were simultaneously determined gravimetrically, and the average values varied between 35 and 821 mg/MJ for residential heaters and between 14 and 1975 mg/MJ for boilers. The mean value for all residential heaters was 160 mg/MJ and 216 mg/MJ for all boilers. For log wood boilers, the mean values were 42 and 777 mg/MJ (modern and old boilers respectively), for pellet boilers 59 mg/MJ and for the wood chip boiler 260 mg/MJ.

As could be presumed, the emissions of PM_{2,5}, EC and OC from modern boilers with advanced combustion technology were much lower than from older boilers with simpler technology. The same relation also holds for emissions of TOC, CH₄ and NMVOC. When comparing emissions at nominal heat load and at part load, the PM_{2,5} and OC emissions at part load were much higher than at nominal load, while the EC emissions were at about the same level.

1 **Aim**

The aim of this project has been to measure emissions of PM 2.5, EC (Elemental Carbon), OC (Organic Carbon), BC (Black Carbon), CH₄, methane and NMVOC from domestic boilers and stoves fired with biomass fuels. The boilers and stoves were chosen to be representative for the existing stock in the Nordic countries. In this way, the results achieved will substantially enhance the database for emission factors for e.g. Black Carbon

2 **Background**

2.1 **Nordic work for improved emission factors for SLCP**

Short Lived Climate Pollutants (SLCP), is a group of substances that have a negative impact on climate, air quality and human health. The SLCP includes mainly aerosol particles, tropospheric ozone, methane, and hydrofluorocarbons. An important source for aerosol particles as well as methane is combustion, especially small-scale combustion of biomass. During the last years, the importance of Black Carbon (BC) in the emitted aerosol particles for climate change has been highlighted. This is especially the case for sensitive areas such as the Arctic. It is supposed that reduced emissions of BC will give a more rapid response in reducing the climate change in comparison to reducing CO₂ emissions due to the shorter life time of these species in the atmosphere.

In order to assess and understand the contribution from BC to a changing climate in the Arctic, the Nordic Council of Ministers in 2013 launched a project named “Improved emission inventories of SLCP”. In this project, all five Nordic countries work together in three areas: Background analysis, Measurements and Abatement measures. For the measurement part, a pre-study was conducted, and the proposed program was then in 2015 put out to tender. The procurement was won by SP in cooperation with DTI. The work has been conducted during 2015-2016, and the results are reported here.

2.2 **Domestic biomass combustion in the Nordic countries.**

In this paragraph, a very short description of the domestic use of biomass in the Nordic countries is given. For a more detailed description, see [2].

The stock of equipment for domestic biomass combustion differs much between the Nordic countries. Very shortly, the following types dominates in terms of activity (i.e. energy in fuel consumed) in the different countries:

Denmark:	wood log stoves (55 %), new boilers (15 %)
Finland:	wood chips boilers (17 %), log wood boilers (19 %), masonry heaters/ovens (35 %), sauna stoves (13 %)
Norway:	stoves with old technology (47 %), stoves with new technology (48 %)
Sweden:	log wood boilers (57 %), pellet boilers (17 %), log wood stoves (16 %)

3 Experimental

3.1 General

The investigation was made in close cooperation between SP Technical Research of Sweden and Teknologisk Institut, Denmark. The project involved measurements on central heating boilers fired with log wood or wood pellets as well as on wood and pellet-fired room heaters and stoves. It is relevant to divide the Nordic stock of heating devices into these two groups for two reasons; firstly the way of use of the two groups differ significantly and secondly, the test methods normally used therefore also differ. It was consequently decided that SP would cover the central heating boilers and that TI would cover the room heaters and stoves.

The choice of test objects as well as the choice of test methods are thoroughly described in the pre-study for this project [1]. For simplicity, short descriptions are also given below.

3.2 Test objects

In Table 1, the chosen types of test objects are listed.

Table 1: Types of boilers and stoves included in the measurement campaign

Notation	Type of boiler/stove
<i>Boilers</i>	
P1	Log wood boiler with inverse combustion and λ -probe. Ceramic grate.
P2	Log wood boiler I with inverse combustion and flue gas fan . Ceramic grate.
P3	Log wood boiler II with inverse combustion and flue gas fan. Ceramic grate. Different manufacturer than P2
P4	Log wood boiler with inverse combustion and natural draught. Ceramic grate.
P5	“Simple” log wood boiler made from cast iron, natural draught and upward combustion
P6	Old combination boiler (oil + wood), upward combustion
P7	Traditional pellet burner in an combination boiler
P8	Advanced pellet burner with λ -probe in boiler designed for pellet firing
P9	Pellet boiler with integrated grate burner with λ -probe, pilot flame
P10	Wood chip boiler with λ -probe, pilot flame
<i>Heaters</i>	
A0	Modern medium class wood stove
A1	Traditional simple stove (DIY stove)
A2	Modern popular wood stove
A3	State-of-the-art room heater
A4	Traditional Nordic cast iron stove
A5	Traditional Nordic tiled stove
A6	Traditional Nordic slow heat release appliance (masonry stove)
A7	Swedish type pellets stove (now obsolete)
A8	European type pellets stove
A9	Traditional Nordic sauna stove

3.3 Operating conditions and test methods

3.3.1 Operating conditions

The test program included in general a test at nominal heat load as well as a test on part load. However, the operational habits are different for different product types. This is valid primarily when comparing boilers and residential appliances, but it is also valid for different product types within the boiler and residential heater group.

The test program was in general based on the relevant European standards for solid fuel central heating boilers (EN 303-5) [2] and for room heaters for solid fuel (EN 16510) [3]. These methods are described in some detail in the pre-study for this project [1], and is therefore not further dealt with here.

3.3.1.1 Boilers

A log-wood boiler which is connected to an accumulator tank is normally only operated at its nominal output, loading the accumulator. But the use of an accumulator tank implies a start-up fuel batch each time the boiler is used. The combustion of this initial fuel batch is however **not** included in the common EN 303-5 test method, but is relevant from a BC emission data perspective. This is considered in 6.3 below.

For log-wood boilers which are not necessarily connected to an accumulator tank, operation at part loads are very likely. Therefore, such boilers were tested at both its nominal heat output as well as at a part load of 30 %. This level is both relevant both from a heat load point of view as well as being somewhat of lowest possible heat load.

Pellets and wood chips boilers are normally designed to operate within a heat output range of at least 30 – 100 %. Therefore, pellet and wood chip boilers were tested at both their nominal heat output as well as at a part load of 30 %.

In conclusion, boilers were tested at heat loads according to Table 2.

Table 2: Heat loads for testing of different boiler types

Boiler type	Boilers no	Test at nominal heat load	Test at 30 % part load
Log wood boiler for accumulator operation	P1, P2, P3, P4	Yes	No
Log wood boiler not necessarily for accumulator operation	P5, P6	Yes	Yes
Pellet boiler	P7, P8, P9	Yes	Yes
Wood chip boiler	P10	Yes	Yes

3.3.1.2 Residential heaters

All residential heaters were, according to EN 16510 [3], as a basis tested at their nominal heat output. However, residential appliances are normally heating the air in the living area without any distribution system. Operation of the appliance at part loads are therefore very frequent situations.

In the EN standard series valid during 2015 for residential appliances, there was no test on part load for **log-wood fired conventional room heaters**. Therefore, the proposed test part loads in the revision of EN 16510 (endorsed in March 2016) were used.

The nominal heat load is not necessarily the maximum heat load. The maximum heat load would however be obtained e.g. if the fuel batch is large, the combustion air inlets are fully open and the fuel is dry. There are recent indications that these conditions may be advantageous for the formation and emission of Black Carbon during quite short time periods. Therefore such **high load tests** were made for two residential appliances.

Testing according to the Norwegian standard NS 3058 [4] differs from EN 16510 in that tests are made at four different heat loads, of which two are very low loads. The results from the four loads are then weighted together to form a “mean value” with some emphasis on low load operation. It was expressed that some estimation on the relation between BC emissions when tested according to the used EN-related scheme and the emissions when tested according to NS 3058 would be beneficial to have. Therefore, **measurements according to NS 3058** were also made for two residential appliances.

Slow heat release appliances are designed to be fully heated up during combustion of one or two fuel batches at high intensity, after which the accumulated heat is discharged to the surrounding air during a long time period, up to two days. This means only testing at high intensity combustion is relevant. This reasoning is also valid for sauna stoves.

Pellet stoves are on the other hand designed to adapt its heat output to a varying heat demand. Therefore, testing of pellet stoves were made both at nominal heat output and on a reduced heat output.

Table 3 summarizes the proposed heat loads for testing of the different residential appliances.

Table 3: Heat loads for testing of different residential appliances

Appliance type	Appliance no	Test at nominal heat load	Test at part load	Test at high load	Test acc. to NS 3058
Log-wood non-accumulating appliance	A1, A2	Yes	Yes	Yes	Yes
Log-wood non-accumulating appliance	A3, A4, A5	Yes	Yes	No	No
Slow heat release appliance	A6	Yes	No	No	No
Pellet stove	A7, A8	Yes	Yes	-	-
Sauna stove	A9	Yes	No	No	No

3.3.1.3 Fuels

The basic test fuels used mainly adhere to the descriptions in the relevant test methods in terms of moisture content, ash content and net calorific value. This implies for stoves use of hardwood (birch) in general and use of softwood (spruce) in case of NS3058 test work. For log wood boilers, birch wood was used. For pellet stoves and boilers, commercial 6 mm wood pellets was used.

In order to slightly decrease the influence of varying fuel moisture on test results, the acceptable interval for log wood moisture content was narrowed to 16 – 20 %. However, for a few log wood boilers and manually stoked residential appliances also tests with

more moist fuels were made since this was expected to influence the BC emissions significantly. This situation was deemed to be relevant, since that some installations, at least log wood boilers, are most probably fired with less dried fuel at some occasions.

It is also probable that log wood with a low moisture content is used at some times, especially in residential appliances. This is due to the fact that a small amount of fuel often is stored for some days in the vicinity of the appliance before being fired. Therefore, a few cases with more dry fuel were included in the test program.

In Table 4 the test fuel characteristics are summarized.

Table 4: Test fuels characteristics

Fuel type	Water content, %	Ash content, %	Net calorific value, dry basis, MJ/kg
Standard log wood (SLW)	16 - 20	$\leq 1,0$	>17
Moist log wood (MLW)	25 - 30	$\leq 1,0$	>17
Dry log wood (DLW)	10 - 14	$\leq 1,0$	>17
Wood pellets (WP)	≤ 12	$\leq 0,5$	>17
Standard wood chips (SWC)	20 - 30	$\leq 1,5$	>17
Moist wood chips (MWC)	40 - 50	$\leq 1,5$	>17

3.3.1.4 Additional issues in operating conditions

There are at least two other factors concerning the fuel that may influence PM 2.5, EC/OC and other emissions, namely, **how the ignition take place and, at part load, the size of the fuel batches used**. Normally the ignition is made by igniting a small amount of tiny wood pieces placed on paper so that small glowing bed is created (the scout method). Then the combustion chamber is filled up with a full fuel batch and the combustion process then approaches a relatively stable phase.

Alternatively, which has recently been shown to result in clearly lower emissions, the so called **top-down ignition method**, can be used. This means that a full fuel batch is placed in the combustion chamber; then paper and tiny wood pieces are placed on top of the batch and is ignited. Then the fire is slowly propagating down the fuel batch. To evaluate possible influence of the ignition method on EC/OC emissions, a full test cycle using the top-down ignition method was carried out for one residential appliance.

At part load, **the size of fuel batches** may influence the emission levels, both for log wood boilers and for residential heaters. A larger number of smaller batches can be expected to result in lower emissions than fewer larger batches containing the same energy amount. Also, it is probably in practice common to adapt the size of the fuel batches to the actual heat load. Therefore, to investigate the influence of fuel batch size on EC/OC emissions, a full test cycle for part load with small fuel batches was carried out for one log wood boiler.

3.3.1.5 Overview of test cases

Bringing all considerations of test object types, relevant fuel types and relevant operating conditions, two arrays of defined test cases for boilers as well as for residential heaters are identified. These arrays are shown in Table 5 and Table 6 respectively.

Table 5: Identification of test cases for boilers, showing boiler type, heat load and test fuel

Appliance	Test designation	Heat load	Test fuel
P1	P1NomSLW	Nominal	Standard log wood
	P1NomMLW	Nominal	Moist log wood
P2	P2NomSLW	Nominal	Standard log wood
	P2NomMLW	Nominal	Moist log wood
	P2NomDLW	Nominal	Dry log wood
P3	P3NomSLW	Nominal	Standard log wood
P4	P4NomSLW	Nominal	Standard log wood
	P4NomMLW	Nominal	Moist log wood
	P4NomDLW	Nominal	Dry log wood
P5	P5NomSLW	Nominal	Standard log wood
	P5NomMLW	Nominal	Moist log wood
	P5PartSLW	Part load	Standard log wood
	P5PartMLW	Part load	Moist log wood
P6	P6NomSLW	Nominal	Standard log wood
	P6PartSLW	Part load	Standard log wood
P7	P7NomWP	Nominal	Wood pellets
	P7PartWP	Part load	Wood pellets
P8	P8NomWP	Nominal	Wood pellets
	P8PartWP	Part load	Wood pellets
P9	P9NomWP	Nominal	Wood pellets
	P9PartWP	Part load	Wood pellets
P10	P9NomSWC	Nominal	Standard wood chips
	P9NomMWC	Nominal	Moist wood chips
	P9PartSWC	Part load	Standard wood chips
	P9PartMWC	Nominal	Moist wood chips

This means that the boiler test program included 25 tests on 10 different boilers, including six log-wood boilers, three pellet boilers and one wood chips boiler.

Table 6: Identification of test cases for residential appliances, showing appliance type, heat load and test fuels

Appliance	Test designation	Heat load	Test fuel
A1	A1NomSLW	Nominal	Standard wood log
	A1NomMLW	Nominal	Moist wood log
	A1PartSLW	Part load	Standard wood log
	A1HighSLW	High load	Standard wood log
	A1NomDLW	Nominal*	Dry wood log
	A1NS3058		NS3058
A2	A2NomSLW	Nominal	Standard wood log
	A2NomMLW	Nominal	Moist wood log
	A2PartSLW	Part load	Standard wood log
	A2HighSLW	High Load	Standard wood log
	A2NomDLW	Nominal*	Dry wood log
	A2NS3058	NS3028	NS3058
A3	A3NomSLW	Nominal	Standard wood log
	A3NomMLW	Nominal	Moist wood log
	A3PartSLW	Part load	Standard wood log
A4	A4NomSLW	Nominal	Standard wood log
	A4PartSLW	Part load	Standard wood log
A5	A5NomSLW	Nominal	Standard wood log
	A5PartSLW	Part load	Standard wood log
A6	A6NomSLW	Nominal	Standard wood log
	A6NomMLW	Nominal	Moist wood log

A7	A7NomWP	Nominal	Wood pellets
	A7PartWP	Part load	Wood pellets
A8	A8NomWP	Nominal	Wood pellets
	A8PartWP	Part load	Wood pellets
A9	A9NomSLW	Nominal	Standard wood log
	A9NomMLW	Nominal	Moist wood log

For residential appliances, this means that the test program included 27 tests on nine different residential appliances, including five log-wood stoves, one slow heat release appliance, one sauna stove and two pellet stoves. However, during the planning of the project, it turned out that the A7 pellets stove was no longer available on the market, neither was any other substitute using the principle of under-feeding of pellets. Instead the test results from the exploratory part of the test program (see 5.1) was included, in order to expand the data base. The stove used for the exploratory tests was a traditional wood stove, quite common in Denmark and Norway. The designation code of the stove to appear later on in the appendices, is A0.

3.3.2 Test cycles and sampling periods

Both for boilers and for residential appliances, the standard test methods includes a start-up and pre-test period to establish stable thermal conditions, but during which the emissions are not measured. It is known that significant amounts of e.g. particulate material can be emitted during this phase. Therefore, PM sampling was made on separate filters during this period to facilitate determination of EC/OC and PM 2,5 emissions to distinguish them from emissions during stable periods.

For manual log wood boilers, a test cycle of one ignition and pre-test period, followed by two consecutive test periods with one fuel batch each was used. These scheme was followed both at nominal heat output and at part load. The length of the test periods were required to be not less than 2 h. For automatic boilers, one test period after the ignition and pre-test period was carried out. The length of this was required to be not less than 6 h.

For residential appliances, in principle the same procedures were applied. Each test, apart from the tests at reduced heat output, consisted of three consecutive test periods, corresponding to the test pattern of the type test standard to CE-label. Tests at reduced heat output consisted of two consecutive test periods. The test to NS3058 consisted of three respectively four test periods depending on the capacity of combustion air inlets (the highest prescribed rate was not possible to achieve for one stove).

The test cycles for boilers and residential heaters respectively are principally shown in Figures 1 and 2 below.

Figure 1: Test cycle for log wood boiler.

Figure 2: Test cycle for log wood residential appliances. Note: Only three test periods were carried out.

Sampling for determination of PM 2.5 and EC/OC emissions was made during periods of 30 min. Sampling was repeated during each fuel batch. For tests at part load on residential heaters, two samples were taken during each fuel batch, except for one case (A2PartSLW, SB) where the fuel batch was split into two batches of each 50 %..

For log wood boilers, three samples were taken during each fuel batch. For automatic boilers, four samples were taken during each fuel batch. The sampling periods during each fuel batch were evenly distributed over the test period, the first beginning three minutes after loading.

In Table 7, the number of consecutive fuel batches as well as the number of samples for EC/OC determination for each batch are given.

Table 7: Number of samples for PM 2.5 and EC/OC during each fuel batch.

Type of heating device	Heat load	Number of consecutive test fuel batches	Number of EC/OC samples during each load
Log wood boiler for accumulator operation	Nominal load	2	3
Log wood boiler not necessarily for accumulator operation	Nominal load	2	2
d:o	Part load	2	2
Pellets or wood chips boiler	Nominal load	1	4
d:o	Part load	1	4
Log wood stove, not accumulating	Nominal load	3	1
d:o	Part load	1 (2)	2 (1)
Pellet stove	Nominal load	1	4
d:o	Part load	1	4
Slow heat release appliance or sauna stove	Nominal load	3	1
Sauna stove	Nominal load	3	1
d:o	Part load	3	1

4 Sampling and analysis

Below the main equipment and procedures for creating defined test conditions, sampling and analysis are briefly described. This is divided into

- test rigs
- sampling for determination of EC/OC and PM 2,5
- determination of gaseous compounds
- analysis of EC, OC and BC
- determination of PM 2,5 emissions.

4.1 Test rigs

The test rigs can functionally be divided into two parts, i.e. 1) a test stand for creating well defined operating conditions for the object to be tested and 2) an equipment for creating standardized conditions for sampling of flue gases for determination of EC/OC and PM 2,5.

4.1.1 Dilution tunnel

Sampling for determination of EC/OC and PM 2,5 was made in diluted flue gases. This was accomplished by using a full flow dilution tunnel with specifications according to the Norwegian Standard NS 3058. In this, the flue gases from the chimney are led into a duct and diluted with a fresh air. The reasons for this are both to arrive at readily analysable levels of the relevant compounds and also to include possible semivolatile compounds, which will condense when the temperature is lowered by the dilution, in the determination.

The dilution tunnel is described in detail in [4], and a schematic is shown in Fig. 3.

Fig. 3: Schematic of the full flow dilution tunnel for flue gases

The main characteristics of the tunnel operation and sampling during a test are:

- Gas velocity in the tunnel, m/s
- Probe diameter, mm
- Filter temperature, °C
- Volume flow through the PM 2,5 cyclone

Table 8 shows the main operational data used for boilers and stoves respectively.

Table 8: Operational data for flue gas dilution and sampling

Parameter	Boilers	Stoves
Gas velocity in tunnel, m/s	6,0	3,5
Probe diameter, mm	6	10
Filter temperature, °C	< 35	< 35
Volume flow through the PM 2,5 cyclone, m ³ /h	1	1

4.1.2 Log wood and pellet boilers

Testing of log wood and pellets boilers were made on a test stand according to EN 303-5 [2]. In this, the boiler including chimney is placed on a balance in order to be able to follow the fuel consumption during the test. The boiler is attached to a cooling rig, where the heated boiler water is cooled and returned to the boiler. By continuously controlling the boiler water flow through the cooling rig, the heat load (e.g. nominal load or 30 % part load) as well as the flow and return temperatures can be set at desired values.

Figure 4 shows a schematic of the boiler test rig according to EN 303-5.

Fig. 4: Schematic of the test rig used for boilers

4.1.3 Room heaters and tiled stoves

Testing of stoves was made according to the specifications of EN 13240. Also in this case it means that the test object is placed on a platform scale. Figure 5 shows a schematic of the test stand used for residential heaters.

Fig. 5: Schematic of test stand used for residential heaters.

4.2 Sampling for determination of EC/OC and PM_{2,5}

Sampling for determination of EC/OC and BC was made on plane filters with a diameter of 90 mm. The sampling setup and procedure was in the Exploratory phase (see below) made according to [5]. This means that double, parallel filter lines are used, where a quartz primary filter is used in one line and a PTFE primary filter in the other. A quartz back-up filter follows both the quartz and the PTFE primary filters. The principle is shown in Fig. 6.

Fig. 6: Sampling set-up for EC/OC and PM 2,5 in Exploratory phase.

In the Exploratory phase, an investigation was made on the feasibility of this setup for the current purpose and if using double filter lines is necessary for relevant determination of EC/OC. The results are reported in 5.1. Based on these it was decided for the project to use a single filter line with quartz filters, e.g. only the lower sampling line in Figure 6 was used (see also 7.2)

4.3 Determination of PM 2,5 content

The PM 2,5 content of the flue gases was determined gravimetrically by weighing the primary quartz filter before and after sampling and relating it to the flue gas flow sampled. The filter handling procedure was as follows:

- The filter was stored in an desiccator for min. 12 h before weighing in
- The filter was weighed in immediately before start of the sampling
- After end of the sampling, the filter was stored in an desiccator until it was deemed to be completely dry (i.e. several days)
- The filter was weighed again to determine the mass increase
- A 47 mm central part was punched out from the 90 mm filters
- The 47 mm filter was placed in a Petri dish until transport to the analysis.

For a discussion on this procedure, see 7.5.

The primary measurement result is the weight gain of the filter, which is then transformed into the unit mg/MJ according to Appendix 1.

4.4 Determination of gaseous compounds

Determination of TOC, methane and NMOC as well as CO, CO₂ and O₂ (for boilers only) was made in the undiluted flue gases in the chimney.

4.4.1 TOC, methane and NMVOC

The emissions of unburnt hydrocarbons were measured by the following FID instruments:

- JUM 3-300A alt. Sick/Maihak 3006 (boilers at SP)
- Mess- und Analysetechnik THERMO-FID (residential heaters at TI)

Both instruments are continuous FID analysers which are calibrated with propane and which produce results expressed in ppm propane equivalents. They have both a “methane cutter”, which means that also the methane content as well as the NMVOC (Non-Methane Volatile Organic Compounds) content of the flue gas can be determined simultaneously. The methane cutter was a JUM (boilers at SP), at TI it was integrated in the instrument.

4.4.2 O₂, CO₂ and CO

The contents of O₂, CO₂, and CO in the flue gases were determined by the following continuous instruments:

- O₂ paramagnetic analyser M&C PMA 10 (boilers at SP)
- CO₂ infrared analyser X-stream (boilers at SP)
- CO infrared analyser X-stream (boilers at SP)
- CO₂ infrared analyser ABB AO2020 (residential heaters at TI)
- CO infrared analyser ABB AO2020 (residential heaters at TI)

4.5 Analysis of EC, OC and BC

Analysis of EC (elemental carbon) and OC (organic carbon) was made by a analyser operating according to a thermal-optical measurement principle. The analysis was performed by Sunset Laboratory Inc., which also is the manufacturer of the analyser.

Analysis of BC was made by an Magee OT 21 aethalometer. This was made at SP for all filters before they were sent to Sunset Laboratory Inc. for EC/OC analysis.

4.5.1 Thermal-optical analysis of EC/OC

The EC/OC analysis was made according to the NIOSH Protocol 870 (6), which is illustrated in Fig 7.

Fig. 7: Scheme of EC/OC analysis according to NIOSH Protocol 870 [6]

At the laboratory, a 1,5 cm² piece of the sample filter is punched out and used for the analysis. The sample is pyrolyzed in an oxygen-free helium atmosphere where the temperature is raised in steps from 25 °C to 870 °C. During this procedure, the carbon-containing compounds are vaporized (OC). The helium with its OC contents are then oxidized to CO₂, and then methanized to CH₄ in two consecutive steps. The carbon content, corresponding to the OC content, are finally measured by a FID (Flame Ionization Detector).

When this first step is completed, the sample oven is cooled down to 550°C, and 2% of oxygen is added to the helium atmosphere. The temperature of the oven is then raised in steps from 550 °C to 870 °C, whereby the EC on the filter is oxidized to CO₂. The CO₂-containing gas is again sent to the methanator to form CH₄, which is then quantified in the FID, corresponding to the EC content on the filter.

The results are reported in the unit µg/cm², and has then to be recalculated considering the flue gas flows, filter area relations etc. to arrive at the emissions expressed in mg/MJ. The method for this is exemplified in Appendix 1.

4.5.2 Aethalometer analysis of BC and UVPM

The aethalometer uses an optical analysis to quantify the content of Black Carbon directly on the filter used without any laboratory analysis. Therefore, the results can be obtained in principle immediately. Black Carbon is defined by the “blackness” of the sample material. It is claimed that the aethalometer BC measurement is closely proportional to filter-based EC measurements. It is widely used for ambient air measurements etc. It was now included in the measurement programme to study and evaluate the possibilities to use it for measurement in diluted flue gases.

Fig 8: Principle of aethalometer

In the aethalometer, the sample is originally collected on a quartz fiber filter tape which does not move during sampling. The optical analysis is running continuously until the sample spot has reached a certain density; then the filter tape is moving into a new sample spot and analysis is starting over again. In the application used in this project, the filters used for thermo-optical analysis according to the NIOSH scheme were analyzed in the aethalometer before going to the lab.

The optical method used for analysis is a measurement of the attenuation of a light beam transmitted through the sample collected on the filter. The optical attenuation calculated by the instrument is said to be linearly proportional to the amount of BC in the filter deposit.

The attenuation principle is also used in the UV spectral region. In this region, a large number of different organic compounds absorb radiation. This is in contrast to the visible region, where only Black Carbon absorbs. Also, different compounds have strongly different absorption characteristics, which makes specific measurements of these compounds impossible. Therefore, the parameter “UVPM” (=UV-absorbing Particulate Material) is defined as an UV-absorbing material that absorb UV photons with the same efficiency as Black Carbon does at the UV wavelength in question. Hence a measure of the total mass of UV-absorbing organic compounds can be achieved. BUT, and this is to be underlined, is NOT a real physical or chemical compound, only a physical measure of a group of compounds.

5 Results

5.1 Exploratory tests

5.1.1 Experimental

The aim of this initial part was to safeguard that the measurements would be made in such a way that they would:

- produce samples which could be analyzed
- provide some insight of the repeatability of the results
- evaluate if it is possible to simplify the sampling in terms of number of filter holders
- safeguard that the choice of filter material for analysis is relevant

In order to cover different combustion and sampling conditions, the exploratory tests were made on two types of heating devices:

1. A manually fired log-wood boiler with ordinary combustion quality (SP)
2. A manually fired room heater with ordinary combustion quality (TI)

For both devices tests were made at nominal heat output as well as on part load.

The exploratory tests were divided into two campaigns. In the first campaign, one test scheme on nominal heat load were carried out for each of the boiler and the room heater. The sampling flow rate as well as the probe size were calculated so as to achieve a deposition on the filters in the middle of the interval recommended by Sunset Labs. After this test run, all filters were sent to the lab for analysis and for evaluation. No further test runs were made until the results for EC/OC have been received from the laboratory.

At the laboratory, analysis samples from each filter were punched out and analyzed according to Sunset Lab's routines. For one filter each from the boiler and room heater respectively, five analysis samples, representative for 20 % of the filter area each (one for the central part and four for the outer segments), were punched out and analyzed. This was made to evaluate the uniformity of EC/OC distribution across the filter area.

The analysis data for the different filters in each test set (primary quartz, back-up quartz, primary PTFE and back-up PTFE) were then used to evaluate if the use of parallel lines with double filter holders should be necessary to achieve relevant EC/OC data.

Following the evaluation of the first campaign, a second campaign for quality assurance was carried out. The purpose of this was to verify the sampling and analysis procedure, but also to generate data for an estimate of the repeatability. For that reason, two identical test schemes on nominal heat load were made on the same boiler and room heater respectively as in the first campaign. Therefore, in total three identical test schemes were carried out.

In each test, three consecutive samplings for PM_{2,5} and EC/OC determination were made. At nominal heat load, the three samplings are made at three consecutive fuel loadings. At part load, for the boilers three samplings were made during the same combustion period, i.e. the same fuel loading and for the stoves in general two samplings.

5.1.2 Results

The main EC/OC analysis results from Exploratory tests of the boiler are reported in Appendix 2.

For test #1, analysis results for the primary quartz filter, the back-up quartz filter and the back-up PTFE filter are given. For each sampling period, the quotient between the contents of OC on back-up filter for quartz and PTFE respectively was calculated (BU ratio Q/T). The quotients between back-up and primary filter contents of EC and OC respectively (BU/Pri EC and BU Pri OC) were also calculated.

Looking at the values of BU ratio Q/T for OC from test #1 for boiler P1 and stove A2, the mean values are 0,80 and 0,70 respectively. This means that 80 % and 70 % of the OC trapped on the back-up filter behind a primary PTFE filter is also trapped on the back-up filter behind a quartz filter.

For the exploratory tests on room heaters, the main EC/OC analysis results are reported in Appendix 2.

The following conclusions were drawn from the achieved results:

1. **The used values of nozzle size, suction rate and dilution factor in the dilution tunnel resulted in that no filter was clogged during the sampling, and that the filters could be analyzed for both EC/OC and BC.** No analysis results were outside or even close to be outside the range of analysis.
2. **The collected particles on the filters were evenly distributed across the filter area, so that the analysis of EC/OC only has to be carried out for one punched-out sample.** This is a conclusion after the triple punches from analyzing the filters Q2, Q7, 2 and 16 (Appendix 5). The conformity for the three values from each filter is very good.
3. **A single test line is enough to achieve relevant results, this means that only one line with double filter holders for primary filter and back-up filter in quartz is enough.** This conclusion is drawn from the fact that the EC/OC data for the back-up filters behind the Quartz and Teflon primary filters respectively are more or less the same (data for BU Teflon and BU Quartz respectively in Appendices 2a and 2d). The EC/OC analysis is not possible to perform on the primary filter from the second sample line due to the fact that PTFE melts at much lower temperature than used for the analysis. The conclusion must therefore be based on the contents from the back-up filters.
4. **The OC values reported should be based on the sum of contents on the primary filter and the back-up filter. This sum should be multiplied by a constant correction factor, due to the different retention of OC on backup filters behind quartz and teflon filters respectively.** This correction factor should be derived from the complete data set for OC achieved during the project. This means that both the primary filter and the back-up filter need to be analyzed for OC and EC. The EC data reported could though be based on the primary filter content only.

The derivation of the correction factor for OC is described in 6.2.

5.2 Boilers

In this paragraph, the results from testing of boilers are reported, and the possible influence of boiler type and operating conditions is shown. More detailed results, are reported for each test in Appendix 3.

5.2.1 Overview of emission data

In Table 9, emission values for all measured parameters are shown for all tests. The numbers given are mean values over all sampling periods in each test, including the ignition period. Separate values for each sampling period are reported in Appendix 3.

Table 9: Summary of emission data for boilers as mean values over all sampling periods in each test

Test designation	PM2,5 mg/MJ	EC mg/MJ	OC mg/ MJ	BC mg/MJ	UVP mg/MJ	CO mg/M J	TOC mgC/M J	CH4 mgC/M J	NMVOC mgC/MJ
P1NomSLW	24	1,8	12, 5	0 ¹	1,2 ¹	233	31	3	28
P1NomMLW	46	1,6	7,8	0,7	1,5	178	14	2	12
P2NomSLW ²	29,1	3,25	9,9	0,2 ¹	2,7 ¹	1077	69	8	62
P2NomMLW	49,6	4,4	13, 2	1,9 ¹	3,7 ¹	1335	142	13	129
P2NomDLW	32,2	3,3	11, 5	0 ¹	2,5 ¹	754	56	6	50
P3NomSLW	44,7	7,6	18, 5	1,7 ¹	4,9 ¹	2036	149	24	125
P4NomSLW	36,0	14,7	13, 7	0 ¹	1,0 ¹	1516	136	17	119
P4NomMLW	32,1	4,8	14, 0	2,7 ¹	4,9 ¹	1894	214	27	187
P4NomDLW	89,3	9,2	36, 0	3,0 ¹	5,3 ¹	3160	293	46	248
P5NomSLW	319,6	18,7	>96	Too dark	Too dark	3578	499	77	422
P5NomMLW	524,3	>30,6	>14 3	Too dark	Too dark	4748	>262 ³	>21 ³	>241 ³
P5PartSLW	1137,6	15,4	>42 5	Too dark	Too dark	8978	>514 ³	>26 ³	>487 ³
P5PartMLW ⁴	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
P6NomSLW	317,0	27,2	138	17,0 ¹	56,3 ¹	2963	443	35	408
P6PartSLW	1975,2	35,1	776	8,5 ¹	128,9 ¹	6408	1373	194	1179
P6PartSLW small batches	373,3	21,2	181	21,5 ¹	35,8 ¹	3437	880	56	824
P7NomWP	56,6	14,2	11, 2	0,9 ¹	2,2 ¹	631	23	3	20
P7PartWP	181,5	17,0	53, 7	9,4	21,2	1225	55	5	50
P8NomWP	37,5	1,8	11, 8	0	2,0	134	8	1	8
P8PartWP	14,0	6,8	15, 2	0	2,7	250	9	0	8
P9NomWP	15,0	0,5	5,6	0,2	1,5	120	14	1	14

P9PartWP	88,0	6,1	33,9	2,9	12,7	2273	212	20	193
P10NomSWC	48,2	0,9	20,4	0	1,0	366	45	3	42
P10NomMW C ⁵	61,2	5,5	17,3	3,6 ¹	10,3 ¹	1894	100	15	85
P10PartSWC	226,9	6,8	98,2	0	27,9	4479	602	48	555
P10PartMWC ⁵	717,9	13,8	367	Too dark	Too dark	5839	901	60	840

¹ One or more test are "too dark", see appendix Z for results on each test.

² Mean value from all three exploatory tests.

³ Measurement only part of time (se appendix Z). Actual value higher.

⁴ Test not performed due to bad combustion already during P5PartSLW.

⁵ Not possible to ignite on moist wood chips. Values from ignition taken from P10PartSWC.

In Table 10, the mean values of emissions for all tests for each boiler are shown.

Table 10: Mean values of emissions for all tests for each boiler

Boiler	PM2,5 mg/MJ	EC mg/MJ	OC mg/MJ	BC mg/MJ	UVP mg/MJ	CO mg/MJ	TOC mgC/MJ	CH4 mgC/MJ	NMVOC mgC/MJ
P1	35,0	1,73	10,2	0,3 ¹	1,3 ¹	275,38	22	2,27	20
P2	33,8	3,5	10,8	0,7 ¹	3,0 ¹	1050,1	81	8,5	72
P3	44,7	5,2	18,5	1,7 ¹	4,9 ¹	2036,6	149	8,7	140
P4	52,5	9,6	21,3	1,9 ¹	3,6 ¹	2190,1	214	29,9	181
P5	667,3	21,1	213,9	--	--	5819,1	>425	54,2	367
P6	888,5	27,8	365,0	16 ¹	74 ¹	4269,5	899	95,0	804
P7	119,0	15,6	32,5	5,2 ¹	11,7 ¹	928,0	39	4,0	35
P8	25,7	4,3	13,8	0	2,4	191,8	8	1,1	9
P9	31,9	2,9	18,2	1,4 ¹	7,1 ¹	1196,3	113	10,2	103
P10	259,6	6,5	109,3	1,2 ¹	13 ¹	3346,9	41	34,8	422

¹ One or more test are "too dark"

5.2.2 Influence of boiler type on EC/OC and PM 2,5 emissions

In Fig. 9-11, the EC, OC and PM 2,5 emissions for all boilers as mean values of all tests for each boiler are shown.

Fig. 9: EC emissions from each boiler as mean values for all tests

Fig. 10: OC emissions from each boiler as mean values for all tests

Fig. 11: PM 2,5 emissions from each boiler as mean values for all tests

The boilers P1 – P4 are all boilers with modern combustion technology. i.e. with inverse combustion, ceramic insulated combustion chamber, in three cases a flue gas fan and in one case also a λ probe for excess air control. Boiler P5 is a boiler with simple combustion technology, and boiler P6 is an old combination boiler, i.e. a boiler intended for both wood and fuel oil firing. None of these two boilers have any of the features listed above.

It is very obvious that the EC, OC and PM 2,5 emissions are much lower for the modern boilers P1- P4 than for the old-type boilers (P5, P6). This is also the case for TOC, CH₄, NMVOC and CO emissions. However, the difference between modern and old-type boilers are much less for EC emissions than for OC and PM 2,5 emissions.

It is also quite obvious that the values for EC, OC, TOC, CH₄ and NMVOC are higher for boiler P4 which has no flue gas fan. This probably reflects the more stable combustion process in P1, P2 and P3 which all have flue gas fans. For PM 2,5 emissions, there is no such difference.

Another interesting comparison can be made between modern log wood boilers and pellet fired boilers (P7 – P9). In this, a distinction should be made between P7, which is an old combination boiler, in which a separate pellet burner is installed, and P8 and P9. P7 is therefore not designed for pellet firing, but this type of pellet boiler installation is the most common in Sweden. P8 and P9 on the other hand are boilers which are designed for pellet firing; P8 having a separate advanced pellet burner installed and P9 having an grate designed for pellet combustion integrated.

The EC emissions from the two advanced pellet boilers are at the same magnitude at nominal heat output as for modern log wood boilers. For the pellet fired combination boiler, the EC emissions are significantly higher than for the modern log wood boilers. This is not the case for OC emission though. Concerning PM 2,5 emissions, they are of the same order for the advanced pellet boilers as for the modern log wood boilers. PM 2,5 emissions for the pellet fired combination boiler are slightly higher, especially at part load.

Looking at TOC, CH₄, NMVOC and CO emissions, about the same relations can be seen. They are lower for the two advanced pellet boilers than for modern log wood boilers; the

pellet fired combination boiler on the contrary emitting more than the two advanced boilers.

The wood chips boiler (P10) showed slightly lower EC emissions than the modern log wood boilers, but OC emissions were slightly higher. PM 2,5, TOC, CH₄, NMVOC and CO emissions were of the same magnitude.

5.2.3 Influence of heat load on EC/OC and PM 2,5 emissions

For the modern boilers P1 – P4, intended for connection to an accumulator tank, no tests at part load were made since this is not relevant. For boiler P6, and for the pellet and wood chips fired boilers P7 – P10 though, such tests were made. In Figs 12-14 the emissions of EC, OC and PM 2,5 respectively are shown for both nominal and part load.

Fig. 12: EC emissions at different heat loads for each boiler

Fig. 13: OC emissions at different heat loads for each boiler

Fig. 14: PM 2,5 emissions at different heat loads for each boiler

In general, the emissions of PM 2,5, OC, unburnt hydrocarbons and CO are much higher at part load than at nominal load.. However, the EC emission do not differ much between part load and nominal load for the log wood boiler and for the combination boiler with a traditional pellet burner. For the advanced pellet boilers and for the wood chips boiler, all emissions were significantly higher at part load than at nominal heat load, though at low absolute levels.

For the combination boiler fired with log wood (P6), it is also interesting that the emissions at the part load test with small fuel batches were low, in the same level as those at nominal heat load.

5.2.4 Influence of fuel moisture on EC/OC and PM 2,5 emissions

Measurements on boilers P1, P2, P4 (modern log wood boilers) and P5 (“old” log wood boiler) were made both with fuel of standard moisture content and with fuel with high moisture content. For boilers P2 and P4, measurements were also made with “extra dry” fuel. In Figures 15-17, the EC, OC and PM 2,5 emissions are shown for all tests with different fuel moisture contents.

Fig. 15: EC emissions for different fuel moisture contents for each boiler

Fig. 16: OC emissions for different fuel moisture contents for each boiler

Fig. 17: PM 2,5 emissions for different fuel moisture contents for each boiler

The influence of fuel moisture content on EC, OC and PM 2,5 emissions was very weak, if any. Only in the case of “extra dry” fuel in boiler P4 (with natural draught), the emissions differ being significantly higher, not lower. Also for TOC, CH₄, NMVOC and CO emissions, the influence of fuel moisture content was weak; also here with the exception of higher emissions from P4 when fired with “extra dry” fuel.

For the wood chips fired boiler, measurements were made both with fuel with standard moisture content and with fuel with high moisture content. In this case, all emission values were two to five times higher with “moist” fuel than with standard fuel at nominal heat output, and also higher at part load, though not that much.

5.2.5 Correlations between EC/OC data and PM 2,5 data

It is interesting to investigate if there is any correlation between EC/OC contents and PM 2,5 content in the flue gases when comparing different test objects as well as different operating conditions. In Fig. 18, EC emissions for all sampling periods for boilers are showed versus the corresponding emissions of PM 2,5. In Fig 19, this relation is shown for only the sampling periods where the PM 2,5 content was below 300 mg/MJ. This exclusion means that results from all tests of the “old type boilers”, i.e. P5 and P6, as well as the test of the wood chips boiler at part load with moist wood chips are excluded.

Fig. 18: Emission of EC (mg/MJ) vs emissions of PM 2,5 (mg/MJ) for all sampling periods for log wood boilers, pellet boilers and a wood chips boiler..

Fig. 19: Emission of EC (mg/MJ) vs emissions of PM 2,5 (mg/MJ) for all sampling periods for modern log wood boilers, pellet boilers and a wood chips boiler (excl. part load with moist wood chips)

Fig 19 shows a very weak dependence of EC emissions on PM 2,5 levels, thereby indicating that EC constitute a small portion of the particulate emissions.

In Fig. 20, OC emissions for all sampling periods for boilers are showed versus the corresponding emissions of PM 2,5. In Fig. 21, the same relation is shown for only the sampling periods where the PM 2,5 content was below 100 mg/MJ.

Fig 20: Emission of OC (mg/MJ) vs emissions of PM 2,5 (mg/MJ) for all sampling periods for log wood boilers, pellet boilers and a wood chips boiler.

Fig. 21: Emission of OC (mg/MJ) vs emissions of PM 2,5 (mg/MJ) for all sampling periods for modern log wood boilers, pellet boilers and a wood chips boiler (excl. part load with moist wood chips)

In contrast to EC emissions, the OC emissions show a clear dependence on PM 2,5 levels, indicating that OC contribute to about 50 % of the particulate emissions.

5.2.6 Correlations between EC/OC data and BC data from OT 21

In Fig. 22, BC values from the OT21 aethalometer are shown vs EC values from thermal-optical analysis. Also, Fig. 23 shows UVPM values from the aethalometer vs OC values from thermal-optical analysis. In both figures, tests where any data were outside measurement range have been excluded. Also, one outlier (EC and OC) has been excluded.

Fig.22: BC emissions from aethalometer analysis vs EC values from thermal-optical analysis

Fig.23: UVPM values from aethalometer analysis vs OC values from thermal-optical analysis

It can be seen that there is to some extent a qualitative correlation between BC and EC and between UVPM and OC values. However, the quantitative compliance is not very good. That means that measurement of BC with the aethalometer in this configuration is not a good representation of EC values from thermo-optical analysis. This is also the case for UVPM values as representation of OC values. However, the possibilities of the aethalometer for emission measurements is very interesting to investigate further.

5.2.7 Correlations between EC/OC data and gaseous hydrocarbon emissions

In Fig. 24, EC emissions for all sampling periods for boilers are showed versus the corresponding emissions of TOC. In Fig. 23, the same relation is shown for only the sampling periods where the TOC content was below 300 mg/MJ.

Fig. 24: Emission of EC (mg/MJ) vs emissions of TOC (mg/MJ) for all sampling periods for log wood boilers, pellet boilers and a wood chips boiler.

Fig. 25: Emission of EC (mg/MJ) vs emissions of TOC (mg/MJ) for all sampling periods for modern log wood boilers, pellet boilers and a wood chips boiler (excl. part load with moist wood chips)

When looking at Fig. 25, it can be seen that there is no correlation at all between EC and TOC for TOC values below 300 mg/MJ. Nearly all EC values are below 10 mg/MJ and most of them below 5 mg/MJ, irrespective of the TOC value in the interval up to 300 mg/MJ.

In Fig. 26, the OC emissions for all sampling periods for boilers are showed versus the corresponding emissions of TOC. In Fig 27, the same relation is shown for only the sampling periods where the TOC content was below 300 mg/MJ

Fig. 26: Emission of OC (mg/MJ) vs emissions of TOC (mg/MJ) for all sampling periods for log wood boilers, pellet boilers and a wood chips boiler.

Fig. 27: Emission of OC (mg/MJ) vs emissions of TOC (mg/MJ) for all sampling periods for modern log wood boilers, pellet boilers and a wood chips boiler (excl. part load with moist wood chips)

5.3 Residential heaters

In this paragraph, the results from testing of boilers are reported, and the possible influence of boiler type and operating conditions is shown. More detailed results, are reported for each test in Appendix 4.

The stoves were tested according to the EN13240 test pattern, meaning the refuelling interval must be no less than 45 minutes. In general, for the regular wood stoves, the refueling interval was between 45-55 minutes.

PM sampling, and hence also EC and OC sampling, was carried out using a fixed sampling period of 30 minutes, starting three minutes after loading the stove and ending 33 minutes after loading the stove. The sampling pump was controlled by a timer, safeguarding uniform sampling periods.

In recalculation the emissions of PM, EC and OC to emission factors expressed in mg/MJ, one need to compensate for the wood consumption taking place outside the sampling period. This is fairly complicated, because the calorific value degrades over the test from 100% CV in the beginning to very low values at the time the fire is nearly out.

From the illustration below, it is clearly seen that the wood consumption is quite significant for the first three minutes before starting PM sampling, approximately 0,25 kg. After the end of the PM sampling period, after a lapse of a few minutes, the fire is out at the same time the char remaining has a mass of approximately 0,23 kg, however carrying only limited calorific value, estimated 5-10% of the initial CV. Among the regular wood stoves there is a minor variation in terms of refueling interval and the ability to maintain the fire.

For reasons of simplicity, and in order to facilitate bulk recalculation of emission factors, it was decided to apply a common fuel mass correction factor of 0,80 (or 80%) to represent the proportion of firewood consumed during the PM sampling period. For obvious reasons, this approach introduces a minor error (estimated 3-6%) in the recalculation of PM, EC and OC to emission factors, but still this error is insignificant, compared to the substantial error factors descending from using quartz filters for gravimetric PM determination, and that of the uncertainty factor related to EC determination by the NIOSH method (see 6.1, 6.2 and 7.6)

5.3.1 Overview of emission data

In Table 11, a total summary of the mean emission values for residential heaters over each test is given.

Table 11: Summary of emission data as mean values over all sampling periods in each test for residential heaters

Test designation	PM _{2,5} , mg/MJ	EC, mg/MJ	OC, mg/MJ	BC, mg/MJ	UVP, mg/MJ	CO, mg/MJ	OGC, mgC/MJ	CH ₄ , mgC/MJ	NMVOC mgC/MJ
A0NomSLW Ap1	53,1	8,2	23,7	n.a.	n.a.	1977	207	90	117
A0PartSLW Ap2	102,7	14,6	28,9	n.a.	n.a.	2512	177	81	96
A0NomSLW Ap3	133,9	8,9	54,7	n.a.	24,11	3046	259	134	125
A0Part SLW Ap4	105,5	19,8	73,9	n.a.	36,9	3048	256	128	128
A0Nom SLW Ap5	47,1	8,6	41,8	n.a.	26,2	2740	260	120	140
A0PartSLW Ap6	35,7	21,6	52,3	n.a.	14,2	3117	213	117	96
A1NomSLW Ap7	88,6	43,4	11,8	14,1	21,1	961	43	29	14
A1NomMLW Ap8	820,7	17,6	440,7	3,4	47,3	3839	890	207	683
A1NomSLW Ap9	97,3	39,6	9,6	11,5	19,5	1253	68	49	19
A1PartSLW Ap10	94,5	6,5	13,5	2,3	7,9	1777	43	37	6
A1NomDLW Ap11	150,5	19,4	7,9	5,1	13,5	931	44	33	11
A1HighDLW Ap12	287	104,5	43,7	36,6	24,3	2223	128	51	77
A1HighSLW Ap13	208,0	n.a.	n.a.	n.a.	n.a.	785	22	19	3

A1NS3058 Ap14	346,8	31,3	167,4	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
A2NomSLW Ap15	60,0	23	16,9	7,6	23,8	1077	60	27	33
A2NomSLW+ TD Ap16	62,5	13,8	28,7	13,5	15,2	1043	63	26	37
A2NomMLW Ap17	347,9	9	90,9	n.a.	37,4	3078	326	90	236
A2PartSLW+ BUAp18	148,5	23,4	51,4	11,0	35,0	2199	178	87	91
A2PartSLW+ SB Ap19	458,3	12,1	191,3	n.a.	46,6	3084	525	138	387
A2NomDLW Ap20	72,2	9,7	23,4	n.a.	11,0	656	38	21	17
A2HighSLW Ap21	131,4	43,7	48,9	19,1	32,2	1628	130	35	95
A2NS3058 Ap22	346,8	120,0	180,0	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
A3NomSLW Ap23	106,5	2,9	16,5	n.a.	n.a.	919	48	23	25
A3NomMLW Ap24	99,9	4,4	24,6	n.a.	8,4	1490	108	45	63
A3PartSLW Ap25	74,1	3,5	6,7	n.a.	0,9	1387	7	6	1
A4NomSLW Ap26	146,6	12,9	48,2	3,8	24,2	1156	154	37	117
A4PartSLW Ap27	329,7	14,7	127,3	n.a.	30,4	2194	390	105	285
A5NomSLW Ap28	197,9	122,1	56,8	n.a.	n.a.	3145	328	125	203
A5PartSLW Ap29	285,2	109,9	74,6	n.a.	4,3	2750	289	153	136
A6NomSLW Ap30	82,0	22,3	27,1	n.a.	n.a.	1585	164	46	118
A6NomMLW Ap31	78,0	7,2	22,5	1,1	7,6	1175	103	36	67
A7NomWP	The A7 stove is no longer available on the market, neither is any equivalent								
A7PartWP	The A7 stove is no longer available on the market, neither is any equivalent								
A8NomWP Ap32	100,0	9,8	10,9	n.a.	6,8	189	4	1	3
A8PartWP Ap33	152,9	7,3	23,9	n.a.	6,8	447	14	2	12
A9NomSLW Ap34	104,0	17,9	51,6	19,6	21,7	1405	107	32	75
A9NomMLW Ap35	120,3	28,5	51,4	17,0	n.a.	2030	219	60	159

In Table 12, the mean values of emissions for all tests for each residential heater are shown.

Table 12: Mean values of emissions for all tests for each residential heater

Stove	PM _{2,5} , mg/MJ	EC, mg/MJ	OC, mg/MJ	BC, mg/MJ	UVP,mg, mg/MJ	CO, mg/MJ	OGC, mg/MJ	CH ₄ , mg/MJ	NMVOC mgC/MJ
A0	79,7	13,6	45,9	n.a.	25,4	2740	229	111	117
A1	261,7	38,5	98,2	12,2	22,3	1681	177	61	116
A2	213,8	31,9	75,0	12,8	28,7	1824	189	61	128
A3	93,5	3,6	15,9	n.a.	4,7	1265	54	25	29
A4	238,2	13,8	87,8	3,8	27,3	1675	272	71	201
A5	241,6	116,0	65,7	n.a.	4,3	2948	309	139	170
A6	80,0	14,8	24,8	1,1	7,6	1380	134	41	93
A8	126,5	8,6	17,4	n.a.	6,8	318	9	2	7
A9	112,2	51,5	23,2	18,3	21,7	1718	163	46	117

5.3.2 Influence of heater type on EC/OC and PM 2,5 emissions

Fig 28: PM 2,5 emissions from each heater as mean values for all tests

Fig. 28 shows the average PM 2,5 emissions, including all test scenarios SLW, MLW, DLW, HIGH and PART. Since MLW is a major driver of PM_{2,5} emissions and since only 5 out of 9 stoves had the MLW test, this leads to a bias on the average PM emissions which is quite influential for the A1 and A2 stoves but of little influence for the A3, A6 and A9 stoves. It's reasonable to assume that variation in impact is due to the duration of the smoldering combustion phase after reloading, which could be linked to the properties of the air system. Moderns stoves tend to have quite limited capacity opposed to the older stoves.

However, all the stoves display average PM emissions below the Guidebook emission factor for energy efficient stoves at 370 mg/MJ and 6 out of 9 stove have average PM emissions below or close to the Guidebook emission factor for eco-labelled stoves at 93 mg/MJ.

Fig. 29: EC and OC emissions as average values for all tests for each heater

Fig. 29 illustrates that for most stoves OC emissions are generally higher than the EC emissions. Stoves A5 and A9 forms an exemption from the general rule. This is most likely due to their very basic air systems where most or all of the combustion air enters as primary air, leading to substantial soot formation. Please notice that stoves A5 and A9 has the highest EC emissions among all 8 stoves.

Fig. 30: OC emissions values from each heater with standard fuel moisture content

Fig. 30 shows mean OC emissions among all test scenarios (SLW, MLW, DLW, HIGH and PART). The batch fired A3 state of the art stove and the continuously burning A8 pellets stove are performing significantly better than the remaining stoves. Please notice not all stoves had the MLW test. MLW is a strong driver of OC emissions.

Fig. 31: EC emissions values from each heater with standard fuel moisture content

Fig 31 shows mean EC emissions among all test scenarios (SLW, MLW, DLW, HIGH and PART). The A5 slow heat release appliance A5 and the A9 sauna stove working on primary air supply solely or predominantly, displays significantly higher EC emissions than the remaining stoves. The A3 state of the art stove displays the best EC emission.

5.3.3 Influence of heat load on EC/OC and PM 2,5 emissions

Fig 32: PM 2,5 emissions at nominal and part loads for each heater

Except for the A3 stove, part load PM 2,5 emissions are higher than the nominal load emissions. Expectedly, the A3 should also display higher emissions during part load. When it doesn't in this case, it may be the result of several factors. First of all it is difficult to steer an automatically controlled stove like the A3 into reduced heat output, because the valve is beyond control. Secondly there was difference in fuel moisture and thirdly there is the element of repeatability which plays a role too.

For the modern stoves A0-A3 generally, the part load emission is one third higher than the nominal load emission. For the elder stoves A4-A5 the part load emission is roughly two thirds higher than the nominal load emission.

Fig. 33: PM 2,5 emissions when overloading the stove

Overloading the stove with firewood is leading to higher PM 2,5 emissions. On average the PM2,5 emission is doubled (x 2,5) among stoves A1 and A2 tested. Those stoves are prone to react to firewood overload because of their optimized air systems. For the benefit of reducing heat output to the room, many modern stoves have downsized air capacity, when comparing to 10-15 year old stoves.

Fig. 34: OC emissions at nominal and part loads for each stove

Age counts. The newer stoves A0-A3 displays on average significant lower levels of OC emissions than the elder stoves A4 and A5. This is due to enhanced start-up properties in

general among the newer stoves. In general, part load OC is higher than nominal load OC emission. The state of the art batch fired stove A3 has similar OC emitting properties as the continuously burning A8 pellets stove.

Fig 35: EC emissions at nominal and part loads for each heater

Apart from the historic A5 tiled stove and to some extent the A9 sauna stove, all stoves display fairly low EC emissions. There is no clear dependency between heat load and the magnitude of the EC emissions. On 50% of the stoves, the part load and nominal load EC emissions are of the same magnitude. There are reasons to suspect Primary Air supply as a driver of EC emissions, as A5, A6 and A9 stoves are operating solely or predominantly on primary air supply.

5.3.4 Influence of fuel quality on EC/OC and PM 2,5 emissions

Fig 36: PM 2,5 emissions for different fuel moisture contents for each heater

It's quite clear that moist firewood has devastating impact on the PM_{2,5} emissions among A1 and A2 stoves, whereas stoves A3, A5 and A9 are much more robust against moist firewood. There are reasons to believe that the discrepancy is mainly due to limited capacity of the air systems among many modern stoves. As to DLW, only limited excess PM_{2,5} emission is observed.

Fig 37: OC emissions for different fuel moisture contents for each heater

Like with PM, it's quite clear that moist firewood is devastating to emissions of OC among stoves A1 and A2. It's reasonable to assume that there is a similar relation between firewood quality and emissions of OC, as with PM.

Fig. 38: EC emissions for different fuel moisture contents for each heater

It's quite remarkable that emissions of EC reacts differently to firewood quality than PM_{2,5} and OC. Among stoves A1 and A2 where all three firewood qualities were tested, the moist firewood EC emissions are in fact less than half of the normal dry EC

emissions, and among A3, A6 and A9 stoves, the EC emissions are of the same magnitude, independent of the firewood quality. In terms of emission levels, all stoves, except for A5 and A9 and to some extent the A1 stove displays fairly low EC emissions, all below the Guidebook BC emission factor of 26 mg/MJ. It is reasonable to blame the relatively simple air systems of the A5 and the A9 stoves for the higher EC emissions.

5.3.4 Influence of ignition method on EC/OC and PM 2,5 emissions

For the A2 stove, which is the average stove wide spread in Sweden, emission measurements was carried out as well under traditional Bottom-Up ignition, as under Top-Down ignition, in recent years recommended by environmental agencies in Scandinavia. In Fig. 34, the emissions of PM 2,5, EC, OC, BC and UVPM at Bottom-Up ignition and Top-Down ignition are compared.

Fig. 39: Emissions of PM 2,5, EC, OC, BC and UVPM at Bottom-Up ignition and Top-Down ignition from the A2 stove.

It's clearly seen that the emissions of PM, EC, OC and BC are of approximately the same order of magnitude (perhaps with OC as an exemption), when the tests are carried out by an skilled operator in a well-controlled test lab environment. However, it is the general assumption, that among average stove users, the Top-Down ignition method is beneficial as it leads to lower emissions, because the fire is never excessively roared up, meaning that the pyrolysis gases can be burnt in the order they are released from the firewood.

5.3.5 Magnitude of emissions during ignition

Fig. 40: PM 2,5, OC and EC emissions for consecutive fuel loads

Fig. 40 shows the emissions of PM 2,5, EC and OC for the consecutive fuel loads separately. As expected the emissions are higher during ignition, than during the subsequent repeated burn cycles. For the A2 stove, the PM ratio is approximately 2,5. For EC the ratio is approximately 4 and for OC the ratio is approximately 7.

Earlier it was a general assumption that the PM ratio was even higher, but recent years of technological development among wood stoves has led to stoves having better firebox insulation and better air systems, both effective to rapid start-up sequences yielding a robust fire and higher firebox temperatures sooner for the benefit of the emissions.

5.3.6 Correlations between EC/OC data and PM 2,5 data

Fig 41: Correlation between OC and PM for residential heaters

There seems to be a fair correlation between OC and PM. This is not totally unexpected, since OC is normally dominating the carbonaceous fractions from biomass burning.

In contrast, Fig. 42 shows that there is a very poor correlation between EC and PM. This is not totally unexpected, since PM is a composite of the solid fractions and the condensable fractions, whereas EC is just a fraction of the solid matter (the remaining solid matter like ashes counts in the inorganic carbonaceous fraction which was not included in this measurement campaign)

Fig 42: Correlation between EC and PM for residential heaters

5.3.7 Correlations between EC/OC data and BC data from OT 21

Fig. 43: Correlation between OC and UVPM data for residential heaters

There seems to be some relation between UVPM and OC, however the correlation factor is not sufficiently robust to securely deem a correlation.

Fig. 44: Correlation between EC and BC data for residential heaters

Fig. 44 shows that there is a clear correlation between emissions of BC and EC. However, in absolute figures BC counts for approximately one third of the EC emission. This is a very questionable result, since BC should reasonably show higher levels than EC.

5.3.8 Correlations between EC/OC data and gaseous hydrocarbon emissions

Fig. 45: Correlation between EC and OGC for residential heaters

Fig. 45 shows clearly that there is no correlation between EC and OGC.

Fig. 46: Correlation between OC and OGC for residential heaters

In contrast, Fig. 44 shows that there seems to be a good correlation between emissions of OC and emissions of OGC. A possible explanation on any discrepancy between OGC and OC is that the FID (OGC measurement principle) detects all carbon atoms passing by, whereas OC only captures those compositions being able to form particles in the Full Flow Dilution Tunnel.

5.3.9 Relation between PM emissions by NS3058 test engine and EN13240 test engine

Both of the A1 and A2 stoves were subjected to the full scope of testing incl. particle test to NS3058. Figs 47 and 48 show PM emissions for all the tests of A1 and A2.

Fig. 47: Magnitude of NS PM emission compared with the proxy type test PM emission, A1 stove

Fig. 48: Magnitude of NS PM emission compared with the proxy type test PM emission, A2 stove

As expected, the NS test displays higher PM emissions than the EN test at nominal heat output does. The NS emissions are by large four times as high as the normal emissions during the NOM SLW test. For stove A1, the NS emissions are however outnumbered by the moist firewood test MLW, but for stove A2 they are about the same.

Without doubt, part of the difference is due to various sampling periods. The NS samples over the full burn cycle, whereas the European test only samples over 30 minutes, leaving out the first three minutes and whatever remains after a lapse of 33 minutes. But there are reasons to suspect also different firewood type and amount to be main responsible for the difference shown here. The NS uses an “artificial” firebed of spruce with a huge surface and mass calculated as a function of the firebox volume, whereas the European test uses hardwood (here birch) where the mass is defined by the manufacturer corresponding to nominal heat output and a cycle time of no less than 45 minutes.

6 Uncertainty

In the following, we describe representation of uncertainty for measurements of PM, EC and OC. We focus on the new contributors to uncertainty, notably the NIOSH analysis method carrying uncertainty for EC and OC results, and loss of filter material causing PM uncertainty.

As described in detail in 7.6, quartz filters are less suitable for sampling of PM in conjunction with analysis for EC and OC, because they are likely to lose material during handling and use.

In case joint sampling for combined determination of PM, EC and OC is used, the impact of loss of filter material can become quite influential, because precise EC and OC determination relies on controlling the PM dissipation, or layer thickness, to a minimum, preferable within the boundaries of a Grey shading scale having White and Black as the extremes.

In all measurements a back filter was used to secure proper dissipation of PM material, in the eventuality that the front filter had burst for some reason. No filters did burst however, so there was very little, if any at all PM and EC on the back filters. As for OC there can be a certain dissipation on the back filter, due to some VOC's not yet having reached state of equilibrium, as discussed elsewhere in this report.

6.1 PM uncertainty, general

As described in details in 7.6, the absolute PM uncertainty is 1,03 mg within 95% confidence interval, when using quartz filters for PM determination. This leads to a relation shown in Table 13 below;

Table 13: Relative uncertainty for PM determination for different filter weight differences

dw (mg)	1	2	4	6	8	10
U rel. (%)	103	51	26	17	13	10

Depending on the lab practice applied, the resulting PM uncertainty can be 1,03 mg if both filters were weighed at a the same time. However, if both filter were weighed individually, the resulting PM uncertainty becomes the root sum of two times 1,03 square, which equals 1,46 mg.

6.2 EC and OC uncertainty, general

As for EC and OC we have transformed the uncertainties stated in the analysis reports from SUNSET laboratories, illustrated in the example below.

Table 14: Example of uncertainties for EC and OC analysis results

Sample ID	OC(ug/sq cm)	OC unc	EC(ug/sq cm)	EC unc
136 (Front)	21,81	1,19	12,45	0,72
137 (Back)	3,66	0,28	0,03	0,10
138 (Front)	21,56	1,18	9,94	0,60
139 (Back)	3,23	0,26	0,01	0,10
140 (Front)	16,61	0,93	14,86	0,84
141 (Back)	3,26	0,26	0,02	0,10
153 (Back)	1,02	0,15	0,00	0,10
154 (Front)	2,24	0,21	0,01	0,10
155 (Back)	0,89	0,14	0,00	0,10
156 (Front)	1,83	0,19	0,20	0,11
163 (Back)	0,66	0,13	0,00	0,10
164 (Front)	1,71	0,19	0,09	0,10
165 (Back)	1,75	0,19	0,04	0,10
166 (Front)	6,16	0,41	0,39	0,12
167 (Back)	0,84	0,14	0,16	0,11
168 (Front)	1,92	0,20	0,31	0,12

Below Limit of Detection: 0.05 µg/cm²

Below Limit of Quantification: 0.15 µg/cm²

It is very clear that the back filters in practice carry no EC, which does lead to huge uncertainties on the back filters.

According to SUNSET laboratories, uncertainty on EC and OC results are calculated by the equation:

$$Unc = (Conc * 0.05 + 0.1) \mu g$$

We find it justifiable, in terms of uncertainty calculation, to disregard the back filters for EC and for OC. However, for calculation of the OC emissions, both the contents of the front and of the back filter were taken into consideration.

The EC and OC uncertainty was recalculated to relative uncertainty and applied to the calculated emission factors and illustrated by symmetrical error-bars on top of the bar charts shown in 6.3 below.

6.3 Stove uncertainty, DTI

In total, DTI used 232 filters, corresponding to 116 tests of each a pair of filters. Out of those 232 filters, 30 filters displayed negative mass increment, when weighed for PM determination. In the end 97 valid results ended up being reported. DTI did weigh the filters individually, leading to extended uncertainty of 1,46 mg for the PM measurements. Since quite a few of the filter loads in milligrams were numerically quite low, notably all the back filters and some of the front filters, this will inevitably lead to substantial uncertainty. This is illustrated below, clearly demonstrating that for PM emissions lower than 250 mg/MJ the results are in practice useless due to uncertainty being several times higher than the result.

Fig 49: PM results for residential heaters based on the sum of front and back filters including calculated uncertainties

Since the higher uncertainties are caused by the back filters, more robust PM results could be obtained from calculation PM emission solely based on the content of the front filter. The error introduced by disregarding any (diminutive) PM load on the back filter, is more than compensated for, by only having to take into consideration the uncertainty of 1,03 mg applied to a much higher absolute front filter load value. This is shown in Fig. 50 below.

Fig. 50: PM results for residential heaters based on the front filter including calculated uncertainties

By regarding the difference among Fig. 49 and Fig 50, it becomes apparent that in the latter case, taking only into consideration the front filter PM load and only the related uncertainty of 1,03 mg, the limit of reasonable accurate PM results has been lowered from approximately 250 mg/MJ to now approximately 100 mg/MJ.

In Figs. 51 and 52 below, EC and OC results for residential heaters including calculated uncertainties according to 6.2 are shown.

Fig. 51: EC results for residential heaters including calculated uncertainties

Fig. 52: OC results for residential heaters including calculated uncertainties

6.4 Boiler uncertainty

The corresponding uncertainty calculations have been made for PM, EC and OC determinations on all boiler tests. The results are shown in Figs. 53-54 below.

Fig. 53: EC results for central heating boilers including calculated uncertainties

Fig. 54: OC results for central heating boilers including calculated uncertainties

7 Discussion

7.1 Relevance of results for emission factors (EF)

7.1.1 Boilers

For boilers, there is little earlier data before this survey on EC and OC emissions. The judgment on whether the results are as expected therefore has to be made on other emission parameters, primarily PM 2,5, CO and TOC emissions. It is convenient to group the boilers into four groups, namely:

- Modern log wood boilers, P1 – P4 (MLB)
- Traditional log wood boilers, P5 – P6 (TLB)
- Pellet-fired boilers, P7 – P9 (PFB)
- Wood chips boiler, P10 (WCB)

Looking at emissions at nominal heat load with fuel of standard moisture content, calculating the mean values of the interesting emissions for these groups render values in Table 15.

Table 15: Mean values of PM2,5, CO, TOC, EC and OC emissions for boiler groups

Boiler group	PM 2,5 (mg/MJ), mean value	CO (mg/MJ), mean value	TOC (mg/MJ), mean value	EC (mg/MJ), mean value	OC (mg/MJ), mean value
Modern log wood	33	1215	96	6,8	15
Traditional log wood	319	3270	1731	23	290
Pellet fired	36	295	295	5,5	22
Wood chips	48	366	366	0,9	109

The obvious conclusion is that traditional log wood boilers (TLB) emit much more of all these emissions than what other boilers groups do. This is what could be expected. Modern log wood boilers, pellet-fired boilers and the wood chips boiler (MLB, PFB and WCB) emit all about the same amounts of PM 2,5, TOC and EC and therefore can be grouped together. It should be noted that the data included refers to nominal heat load with fuel of standard moisture content.

Comparing the relations between emissions of EC and PM 2,5, CO and TOC from TLB and the other boiler groups, one arrives at the values in Table 16.

Table 16: Relation between emissions from traditional log wood boilers and mean value of emissions from modern log wood boilers, pellet fired boilers and wood chip boiler.

	PM 2,5	CO	TOC	EC	OC
Ratio TLB / (MLB, PFB, WCB)	8	5	7	5	6

It is seen that the relation of EC emissions is about of the same order as for PM 2,5, CO and TOC emissions.

The data above for modern log wood boilers to a quite large extent reflects common use of these boilers; i.e. operating connected to an accumulator tank and using wood that has been dried outside but under cover for some months during summer. Looking at the influence of fuel moisture on emission levels, see Figs 12-14, one sees that a higher fuel moisture content does **not** increase the PM 2,5, EC and OC emissions very much for the modern log wood boilers P1 – P4. For the traditional boiler P5 though, the difference is larger; emissions with moist log wood being 60 -90 % higher than with standard log wood.

These facts are very sensible when looking at the combustion conditions in the different boiler types. In a modern log wood boiler only a small part of the fuel load are taking part in the combustion at any given time; the rest gradually approaching the combustion zone by gravity. During this process, the fuel is continuously dried and then volatilized. The conditions are held almost constant at feasible temperatures in the primary combustion zone; thereby generating reasonably low emissions even with moist fuel. On the other hand, in a traditional boiler as P5, the full fuel load is burning at the same time, meaning that in the beginning of the combustion cycle the total amount of fuel must be dried, then volatilized and finally the char is combusted. This in turn means that combustion temperatures etc. is varying wide over the fuel cycle, as well as the air excess and mixing of air and combustible gases. Significantly higher emissions at last during large parts of the cycle are the natural results.

Concerning the influence of heat load, measurements were made at part load on the pellet fired boilers, on the wood chips boiler and also on the old combination boiler for log wood firing (P6). Generally speaking, the emissions of EC, OC and PM 2,5 from the pellet boilers were significantly higher at part load than at nominal heat load. In Table 17, the ratios of emissions at part load to emissions at nominal load are given for boilers P6 – P10.

Table 17: Ratios of emissions of PM2,5, CO, TOC, EC, and OC at part load to the emissions at nominal load for boilers P6 to P10.

	PM 2,5	CO	TOC	EC	OC
P6	6,2	2,1	3,1	1,3	5,6
P6, small batches	1,2	1,2	2,0	0,78	1,4
P7	3,2	1,9	1,9	1,2	4,8
P8	0,4	1,9	1,1	3,8	1,3
P9	5,9	18,9	15,1	12,2	6,1
P10	4,7	12,2	13,4	7,5	4,8

Generally, the emissions at part load are significantly higher than at nominal load, normally between two and fifteen times higher. Both pellet fired boilers and wood chips boilers are normally not connected to an accumulator tank and are therefore operated directly against the momentary heat demand of the house. **This means that the “real-life emissions” for these boiler types are probably more close to the part load values than to the nominal load numbers.**

Concerning the old combination boiler (P6), it is important to note that operating it with small batches at part load (i.e. shorter time between fuel loadings) causes the emissions to decrease quite dramatically. This is also a reflection of approaching more constant combustion conditions. This type of operation however requires the user to be more or less present at home during firing, which may not be the case for many today.

7.1.2 Stoves

7.1.2.1 Grouping of stove technologies

Among the stoves tested, the stoves A0, A1 and A2 all are modern stoves, basically having the same kind of air system with primary, secondary and tertiary air supply, and they can be regarded as a group in terms of expected emission levels. The A3 stove having automatic valve control and downdraft combustion in a dual firebox is a class of its own. The A4 stove is an elder stove and on top of that, it is made of cast iron, so it is also a class of its own.

The A5 and A6 are share quite a few characteristics in common, even though in age there are centuries between them. Both are masonry/tiled type of stoves having a comparatively basic air system and a long flue way before exit of the stove. They can be regarded as a group.

Being a continuously fed pellets stove, the A8 is a class of its own, and also the sauna stove A9 having a small firebox and a very basic air system is a class of its own. It has been quite common to distinguish and to categorize stoves by age alone, but it is not as simple as that. Many other parameters like insulation of the firebox and the nature of the air system and more also are quite influential on which levels of emissions could be expected. This is illustrated by the curve below, showing the development in emissions measured by NS3058 over a decade at DTI.

Fig 55: Particle emissions according to NS 3058 for stoves tested at DTI during 2002 - 2014

Clearly, the trend displays declining emissions, but in particular for recent years, there is some spread among the PM emission reported. This can be explained by the intended use of the stove, e.g. if it must comply with the Swan level emission of 4, soon going down to 3 g/kg or if it is mainly intended for sales in Norway, where the limit is 10 g/kg.

7.1.2.2 The emission potential of a stove

Any stove can produce any emissions; it just depends on the number of misuse conditions occurring simultaneously. However, in general, modern stoves are more forgiving because of better air systems, better firebox insulation and easier valve control. On top of this comes stoves having various degree of automation onboard, which enables them to adapt to and to control the combustion parameters depending on the quality of the combustion at any time.

Fig 56: Principal spectrum of emissions for different uses of a stove

Fig. 56 above illustrates the principle of type tests being frequently close to the optimum, thus enabling comparison of stoves without influence of any ‘disturbing’ parameters. Live conditions typically represent a wider scale of emissions. A skilled user can at best replicate the results of the test lab, if he bothers to read and understand the user’s manual, but an average user not having the best understanding for wood combustion, can easily produce twice the emissions, perhaps even more. The illustration is purely principal; please do not pay any attention on the actual figures on the x- and y-scales.

Below a summary of the most influential parameters affecting stove emissions is given. This is summarized in Table 18.

1. Moist firewood

It is very important that fuel moisture is controlled within a range of typically 12-20%. Firewood having higher moisture content is hard to ignite, and it takes longer time to establish a robust fire with sufficiently high temperature to secure complete combustion of the pyrolysis gases. The current measurement campaign has added new information to understanding the importance of using well-seasoned firewood. There are strong indications that old stoves having higher capacity in the air system, is better able of burning wet firewood, than modern stoves having limited capacity. As well the amount of air as the ability to supply plenty of primary air seems beneficial to burning wet firewood. As to the drier firewood, it has become apparent that moderately over-dried firewood (8-12%) does not have any significant adverse influence on the emissions. It is generally recognized that extremely dry firewood (0-5%), e.g. waste wood from industrial manufacture of windows or floors, is too dry to burn properly and is likely to cause excessive emissions of soot.

2. Overloading the stove

Modern stoves are optimized for moderate to low heat generation. Typically, a modern stove is tested to 5 kW nominal heat output, corresponding to a burn rate of approximately 1 kg firewood per hour. In modern, well insulated dwellings this is sufficient to maintain comfort temperature. Over recent years, it has become apparent that stove manufacturers are downsizing the capacity of the air system. At the NS3058 particle test, earlier most stoves were able to enter the higher most burn rate 3 ($>1,9$ kg/h) and burn rate 4 ($>2,8$ kg/h). Nowadays it is quite common that the uppermost burn rate attainable is burn rate 2 ($>1,25$ kg/h).

So if or when, the modern optimized stove ends up in a dwelling with higher heat demand, it's likely that the user will repeatedly enter more firewood, that the stove was optimized for. This is leading to shortage of combustion air, which in turn is leading to higher emissions.

Fig. 57: Heavily overloaded stove seen in a private dwelling in the fall 2015

The opposite, underloading, is hardly an issue, except for those cases, where the user only loads one wood log per refueling. It's commonly recognized among professionals that just one log doesn't burn properly. Better enter two logs of half the size.

3. Smoldering combustion

Smoldering combustion occurs if the air valve is throttled while there are still unburnt firewood. This might happen if the stove was oversized and the user attempts to control heat output by throttling the valve rather than reducing the fuel load, or it could occur in case the user attempt to fire overnight, by stuffing the stove and closing the air valve. Smoldering combustion inevitably leads to significant increased emissions.

4. Increased draft

At the type test, stoves are tested at a fixed flue draft of -12 Pa. This is a kind of worst-case scenario mimicking a pretty poor chimney. In real life, most chimneys builds up

higher flue draft, in fact 20 Pa or more is not unusual, if only the chimney is 3-4 m high. Higher flue draft is helpful to good combustion quality, and can to some degree make up for other misery.

The combination of wet firewood and a poor chimney is bad for the combustion quality, as the chimney and the stove are mutually dependent on each other and the good results relies on release of adequate residual heat into the chimney.

In the eventuality of too high flue draft the burn rate is often accelerated towards higher heat output. If excessively high flue draft is a permanent problem, it can be dealt with by installing a flue draft regulator in the chimney. Conversely, if the flue draft is generally too low, it can be dealt with by installing an flue gas fan on the top of the chimney.

5. Maintenance of the stove

A well-maintained stove is prerequisite to remain the good combustion quality. Gaskets and firebox insulation material deteriorates due to tear and wear, and has to be replaced regularly, depending on the intensity of the use of the stove. A leaking door will allow plenty of false air to enter into the firebox and counter act controlling the combustion with the valve system. Broken or missing firebox insulation bricks leads to colder combustion, in turn leading to higher emissions.

6. Installation error

If the entry of the connecting fluepipe into the chimney is leaking, fluedraft is lost, potentially leading to reduced combustion quality.

Table 18: Summary of selected variables and their influence on combustion quality

Factor	General influence on emissions
Moist firewood	Increases emissions
Moderate over dry firewood	Neutral
Overloading by 150% or more	Increases emissions
Underloading (part load)	Increases emissions
Smoldering combustion	Increases emissions
Increased flue draft	Reducing emissions
Poor stove maintenance	Increases emissions
Installation error	Increasing emissions

Therefore, in order to sum up this chapter, it is obvious that there is no single condition such as ‘real life operating conditions’, on the contrary there are any number of combinations including properties of the stove, properties of the house and its chimney, variation in climatic conditions (even within Scandinavia), variation in stove type, variable quality of the firewood used and perhaps most important of all, the insight of the user. Several of those variables has been taken into consideration in this project, such as the quality of the firewood (DLW, SLW, MLW) overloading the stove and various valve settings (the NS tests), so these results are deemed adequately diverse, to allow composition of emission factors for air quality inventories.

In terms of reducing emissions from domestic wood burning, there is an increasing awareness of the stove, the chimney and the user acting as an interconnected system, where the resulting emission depend on all three components. This new understanding should lead policy makers to consider, whether posing increasingly stricter emission

limits on the stoves is the most effective way to deal with the air quality issue. In this regard, it is counterproductive that national building regulations these years seem to let go of absolute requirements to the chimneys in terms of height and temperature class, to replace these by the so-called functional requirements, which are so much harder to make sure works. As for the abilities of the users, there are evidence that the burn right campaigns repeatedly launched by the Nordic environmental protection agencies, does have an impact on having stove users abandoning old habits and taken on new and better ways of using the stove. This work must carry on, on regular intervals.

7.2 Derivation of correction factor for OC data

7.2.1 OC distribution between filters in general

In brief, there are principally three alternative correction models for OC results, in terms of filter configurations [7].

1. Use the dual line T-Q and Q-Q sampling train according to Figure 6. Subtract OC value for T-Q backup filter from OC value of Q-Q primary filter (to allow individual assessment of particulate phase and vapor phase OC).
2. Use a single line Q-Q sampling train. Subtract OC value for backup filter from OC value for primary filter, either individually or as an average over several samples.
3. Use a single line Q-Q sampling train. Report uncompensated OC values from the primary filters only.

In Table 19 below, the analysis results for OC and EC on the primary (Front Q-Q and T-Q respectively) and back-up (Back Q-Q and T-Q respectively) filters are shown.

Table 19: Test results, DTI wood stove Exploratory test (raw data in $\mu\text{g}/\text{cm}^2$)

Test #	Filter #	Filter location	EC	OC
1. test run	8	Front Q-Q	1,4	12,3
2. test run	10	Front Q-Q	1,2	7,9
3. test run	12	Front Q-Q	1,8	7,0
4. test run	14	Front Q-Q	1,0	11,9
5. test run	16	Front Q-Q	4,3	6,3
1. test run	9	Back Q-Q	0,0	2,9
2. test run	11	Back Q-Q	0,0	2,5
3. test run	13	Back Q-Q	0,0	1,8
4. test run	15	Back Q-Q	0,1	2,7
5. test run	17	Back Q-Q	0,0	1,5
1. test run	1	Back T-Q	0,0	3,7
2. test run	2	Back T-Q	0,0	3,2
3. test run	3	Back T-Q	0,0	2,6
4. test run	4	Back T-Q	0,1	4,2
5. test run	5	Back T-Q	0,0	2,8

In order to assess the OC retention share after the Teflon front filter, respectively the quartz front filter, the data is arranged as below. Then the share of retention calculated vs the OC content of the Q-Q front filter is calculated (the Teflon front filter just acts as a blocker and is not fit for thermos-optical analysis). The ratios are shown in Table 20 and 21.

Table 20: OC share Back/Front on Q-Q configuration

	OC Front Q	OC Back Q	Back/Front %
1. test run	12,3	2,9	24%
2. test run	7,9	2,5	32%
3. test run	7,0	1,8	26%
4. test run	11,9	2,7	23%
5. test run	6,3	1,5	24%
		Average	26%

Table 21: OC share Back/Front on T-Q configuration

	OC Front Q	OC Back Q behind T	Back/Front %
1. test run	12,3	3,7	30%
2. test run	7,9	3,2	41%
3. test run	7,0	2,6	38%
4. test run	11,9	4,2	35%
5. test run	6,3	2,8	45%
		Average	38%

It is clearly demonstrated that although of approximately the same order of magnitude, still the OC deposition behind teflon is approximately 30 % higher than behind quartz. This observation adapts to the theory of Turpin et al [7]:

“If condensable organics had not yet reached equilibrium in the vicinity of the quartz front filter, the front filter would be depleting the concentration reaching the backup filter. This would result in higher loadings on TQ backups than on QQ backups, as observed. This logic suggests that a TQ backup is a better estimate of adsorbed vapor on a quartz filter than a QQ backup.”

When we subject our data to the three alternative correction models listed above, the results for the five tests are as in Fig 58.

Fig. 58: Results of correction of OC measurements according to three models by Turpin et al [7]

The conclusion is that correcting values according to any of the two first models yields not to differing results.

After this preliminary analysis, it was concluded that for all measurements in the project, also the backup filter should be analyzed for EC and OC in order to build up an extensive database for OC distribution. This database would then be used as a basis for elaborating a constant correction factor taking care of the OC distribution between filters. Below, this elaboration is described.

7.2.1 Derivation of OC correction factor

According to theory, OC dissipation on the backup filter is an indicator of organics passing the front filter in gaseous phase or under transition from gaseous to liquid phase. Looking at the OC-OC data, it soon becomes apparent that the OC dissipation on the backup filter is guided by some boundaries. The data is analyzed focusing at the same time on the magnitude of the OC dissipation on the front filter and the relative difference between the back and front filter OC dissipation.

First, the mean values of OC for each stove is calculated for front and back filters respectively, based on the raw data from Sunset Labs, expressed in $\mu\text{g}/\text{cm}^2$. Regarding the individual sets of OC-OC data, it is apparent that the OC concentration on the backup filter is seldom less than 1 – 2 $\mu\text{g}/\text{cm}^2$ regardless of even quite low concentrations of OC on the front filter. In case of “clean burning stoves” like A3 and A8, the OC-OC back-to-front ratio can easily amount to 60 – 80 % when the magnitudes are at the same time very low. This suggests that there is always a certain portion of vapor phase OC present, also for the clean burning stoves having low absolute OC emissions.

In the opposite end of the scale, OC concentration of the backup filter was never exceeding 6 $\mu\text{g}/\text{cm}^2$ regardless how high the front filter OC concentration was. In case of

less clean burning stoves, the OC-OC back-to-front ratio can be as low as 5-10 % when the magnitudes are quite high.

In Table 22, mean values for OC on front and backup filters respectively for each stove are shown. Fig. 59 illustrates these values graphically.

Table 22: Mean values for OC on front and backup filters respectively for each stove

Heater	Mean Front OC ($\mu\text{g}/\text{cm}^2$)	Mean Back OC ($\mu\text{g}/\text{cm}^2$)
A0	11,1	2,3
A1	13,8	2
A2	20	3,6
A3	2,2	1,1
	26,3	2,7
A5	34,3	2,6
A6	36,4	3,3
A8	2,5	1,5
A9	13	2,2

Fig. 59: Mean values for OC on front and backup filters respectively for each stove

Given that there is a minimum value for the backup filter OC concentration, and given also that there is also an upper value which was not exceeded, it is reasonable to assume that across all stoves tested there is a non-linear relation between backup and front filter OC. Fig 60 shows a best-fit for this kind of relation.

Fig 60: Best-fit logarithmic relation between mean backup and front filter OC contents for each stove

The R-square value for the best-fit of 0,73 indicates that there is some kind of systematic relation between front and backup filter OC concentrations, and that is justifiable to assume that the nature of this is logarithmically rather than linear. If one remove the outlying pair of data (20; 3,6), the curve fit becomes even better with an R-square value of 0,88, se Fig. 61.

Fig 61: Best-fit logarithmic relation between mean backup and front filter OC contents for each stove, excluding one outlier

In order to allow further propagation towards recommending a constant OC correction factor, the subset of regular wood stoves is studied, assuming that they are a relatively

uniform group, sharing the same basic features regarding the overall working principle. Table 23 shows the data for this subset.

Table 23: Mean values for OC on front and backup filters respectively for stoves A0, A1, A2 and A4

Heater	Mean Front OC ($\mu\text{g}/\text{cm}^2$)	Mean Back OC ($\mu\text{g}/\text{cm}^2$)	OC-OC relative (%)
A0	11,1	2,3	21%
A1	13,8	2	14%
A2	20	3,6	18%
A4	26,3	2,7	10%
Mean	17,8	2,65	15 %

If one for e.g. cost reasons wants to skip analysis of the backup filter, it is therefore reasonably accurate to downscale the front filter OC concentration from the NIOSH 870 to 85 % of the analysis result. However, this was not the case within this project since for all tests both the front and the backup filters were analyzed for OC.

Coming back to the different OC retention between Teflon and quartz filters described in 7.2.1, the calculated retentions of 26 % and 38 % for Q-Q and T-Q configurations respectively indicate that the Q-Q filter configuration overestimates the vapor phase share by approximately 30 %.

Therefore, it was decided that the best estimate of the “real” OC emissions is to multiply the OC analysis result with a correction factor of 0,70. This is applied to the sum of OC contents from front and backup filters, i.e.

$$\text{OC}_{\text{corr}} = (\text{OC}_{\text{front}} + \text{OC}_{\text{backup}}) \times 0,7 \quad (1)$$

All OC results in this report have been corrected according to (1).

7.3 Influence of methods used for determining EC/OC and BC data

Until recently there has been several interpretations of what is Black carbon. The issue has been studied among diverse universities in Europe and in the US, leading to various interpretation of BC and various measurement principles. Below, a very short overview of existing methods is given. In Appendix 5, a more detailed survey is reported.

In 2013, Bond et al for the first time gives a fairly precise definition of BC [8]. In this, BC is characterized by four physical properties:

- It strongly absorbs visible light
- It is highly refractory, can withstand up to 4000 Kelvin
- It is insoluble in water
- It exists as an aggregate of small carbon spherules

Linked to the thermo-optical analysis method, it has been common to break down BC into the two main fractions EC and OC. In this project we have also assessed the light absorbing principle leading to BC (and UVP) figures.

The illustration below from [9], is frequently used to illustrate the main measurement principles.

Fig. 62: Principles of measurement for different carbonaceous compounds

By the thermal-optical method, there is a sharp splitting point between EC and OC, whereas the light-absorption classification recognizes the Brown Carbon fraction, which is an intermediate stage between the solid fraction and the vapor phase fraction. There is an increasing awareness that Brown Carbon as a SLCF is more important than the OC fraction.

So the applied method and the measurement results are closely linked together, and the measurement results are not compatible between different measurement and analysis principles.

The definition of Bond et al has been commonly recognized, and has led to widespread use of dissipating BC specimen on Quartz filters for subsequent analysis with a thermal-optical analysis method. Among several analysis protocols available, the current project has used the NIOSH870 method, applied by SUNSET laboratories. The NIOSH method returns two results, one for the EC fraction and one for the OC fraction.

Upon request the filter specimen was also analyzed with an Aethalometer instrument based on optical analysis. The Aethalometer returns a result for the BC fraction.

Fig. 63 below is intended to illustrate the different ways of characterizing stove emissions, ranging from solid material like soot or ashes, over liquid particles to pure vapor phase components. (the lower, the more solid; the higher, the more vapor phase)

Fig 63: Schematic representation of terms for PM related emissions

In terms of the carbonaceous components like BC, EC and OC, the results step down from different analysis methods, and thus they are not comparable. In order to evaluate a given carbonaceous emission result, one needs to know not only the measurement principle but also the analysis protocol.

Concerning the difference between NS3058 and the EN type test parameters dust and OGC, NS3058 PM emissions are sampled in a full flow dilution tunnel at ambient temperature, thus representing all those components of the flue gas that can condensate at around 30 degrees Celsius. But those still on vapor phase are passing through the filter and are not represented. The NS3058 therefore "upgrades" the measured result with an exponential factor adding approximately 40% to the result which is reported.

In the EN test the solid fraction is measured at a heated filter (70 °C initially) and the remaining fraction including the purely vapor phase components like methane and ethane are measured as VOC using a Flame Ionization Detector (every C-atom sparks a tiny flame, which is summed up).

So, as was the case with BC/EC/OC, NS3058 and dust + VOC emissions are just not comparable.

7.4 Aethalometer as a means of determining BC data

In general the aethalometer can be regarded as a fairly adequate way to characterize BC emissions. It has been shown that there is a fairly good relation between the BC and the EC test results as mentioned above. However, the relevance of absolute values from the OT21 Sootscan instrument has to be further investigated, e.g. in terms of software.

The correlation between the virtual parameter UVPM delivered by the aethalometer and OC values from thermo-optical analysis is on the other hand very weak.

7.5 PM measurements using quartz filters

7.5.1 General

The current project includes reporting of emissions of PM_{2.5} based on gravimetric determination of PM deposition on the quartz filters, mainly intended for thermo-optical analysis for EC and OC. The coarser particles are removed by means of a PM_{2.5} cyclone upstream of the filter.

Normally glass fiber filters are used when performing PM measurements in the full flow dilution tunnel. This is a proven concept which has demonstrated its ability for many years of testing to the Norwegian particle measurement standard, NS3058. However, over recent years, as the stoves have become increasingly cleaner burning, there have occurred cases where the PM emission was reported at or close to the detection limits of the method.

This project has been our first scale experience, using quartz filters for gravimetric PM determination. Dictated by considerations on not overloading the filters to avoid saturation under the analysis process for EC/OC, the amount of PM dissipated has been further reduced, compared to the normal NS3058 sampling procedure. This precaution has aggravated borderline phenomena with filters carrying only limited amounts of PM. The quartz filters have been pre- and post-treated conditionally with as well internal quality procedures as with the provision of Sunset Laboratories (incl. pre-heating to 850 °C and drying in a desiccator cabinet prior to and after exposure) during the course of the measurements. In spite of this, it soon turned out that the differential weight of the exposed filters occurred quite low compared to the shading visible, and in some cases even negative mass increment was recorded. This was quite a puzzle, but to a start the phenomenon was explained by lack of moisture control, despite due precautions taken.

In relation to the SUNSET NIOSH 870 analysis protocol, the ideal PM load is 1-15 µg/cm². Recalculated to Ø = 90 mm disc filter used by DTI, this amount equals to an absolute PM mass of 0,063 – 0,945 mg. In comparison the uncertainty of the precision scale is 0,210 mg, so given due consideration up front, it might have been realized that safe PM determination was going to be difficult, at best.

Once it had been realized that any occurrences of negative filter mass increment had nothing to do with moisture control, we searched literature for possible causes and for other similar observations. It soon turned out that quartz filters are known to be prone to losing mass during handling and use. Two papers held documentation of loss of filter material among popular used filter brands, [10] and [11].

The Executive Summary of the NPL report, [10], reads:

“In order to investigate these potential losses NPL undertook a study to measure losses from filters. A number of instrument suppliers provided example filter holders and filter papers, and these were tested using air drawn from the NPL stack simulator containing humidified stack gases with no particulates. The aim was to study losses from the filters, in the absence of particulate matter. **Significant losses were observed in all filter holders tested and in all filters, in the range of 0.3 to 1.4 mg.** Flow rate did not appear to affect the mass loss, within the statistical variability of the measurements. **Significant losses appeared to be a result of the handling of the filters, particularly due to placing them into and out of the filter holders.** Filters produced using a new laser-

cutting technique appear to lose material due to mechanical damage caused during installation in the filter holder.”

As quoted above, the uncertainty of the scale was 0,21 mg and the ideal PM load was 0,062-0,945 mg, so it goes without further argument, that loss of filter mass in the range of 0,3 to 1,4 mg is prohibitive to secure PM determination, or, at best, leaves the PM figure with an accuracy margin of up to or more than 100% uncertainty.

The US EPA paper, [], has a wider scope of investigation and shows more diversified filter mass deviation among glass, teflon and quartz filters. Some lose mass, other gain mass. Table 1 in this shows mass increments or losses over eight repeated weightings. As for the Pallflex Quartz filter brand, which is among a number of filter brands used by DTI, a mass variation in the range of - 0,23 mg to + 0,26 mg is reported. Again, this is of same magnitude as the accuracy of the scale and well above the lower limit of ideal PM load range of 0,062 mg to 0,945 mg.

The report does not suggest any reason for positive mass increment, neither is the filter arrangement shown, but in case the traditional setup, with a front filter and an back filter, it is reasonable to assume that any filter material lost from the front filter, is going to be dissipated on the back filter, thus causing potential mass increment of the back filter. Summing up on the two papers, it is quite obvious that quartz filters because of their delicate nature is prone to losing mass, and thus they are not suitable for the finer PM measurement applications.

7.5.2 Quantification of measurement uncertainty

The quartz filters are handled a number of times during the entire process, as illustrated in Fig. 64 below. Pre and after weighing is indicated symbols w_1 (prior) and w_2 (after) in the flow chart. As it has been established above, determination of the particle mass ($\Delta w = w_2 - w_1$) is affected by several factors introducing uncertainty, most important the accuracy of the scale and the unknown factor caused by loss of filter material.

Fig. 64: Scheme of handling steps in the gravimetric determination of PM

The accumulated uncertainty, $U(\Delta W)$, is determined from available data from literature. Because it is the most recent paper and the one dedicated on study of material loss solely, we base the calculation of $U(\Delta W)$ on data from the NPL report [10]. The report documents, including use of blinds, the influence of mounting the filter, dismounting of the filter, passing through of clean air. However it does not take into consideration the influence of filter handling with tweezers in relation to weighing of the filter prior to, and after exposure, which would have added further to the resulting mass determination uncertainty $U(\Delta W)$.

The accuracy of the lab scale is taken direct form the calibration certificate.

Both papers from EPA and NPL shows changes in filter weight ($\Delta W < 0$) during blind tests. It is also clearly shows considerable spread on recorded ΔW . In Fig. 65 below, plots including error bars based on data from EPA Table 2 and NPL Table 1 are shown, both illustrating change of filter weight of a filter processed through a blind test.

Fig. 65: Weight changes including error bars for blind tests in [10] and [11].

In Table 24, the uncertainty data from [10] and [11] are shown as well as the chosen data for calculation of uncertainty in this project is shown.

Table 24: Uncertainties in PM determination from [10] and [11]

Source of uncertainty	contrib, k=1 [mg]	Selected	comment
Lab scale precession	0,210	0,210	from calibration cerificate
EPA (filter handling)	0,540		from CEN paper, table 1
EPA (blind test)	1,780		from CEN paper, table 2
NPL (bias)	0,342	0,342	from NPL paper, table 1
NPL (spread)	0,322	0,322	from NPL paper, table 1
<i>combined uncertainty (k=1)</i>		0,515	
Uncertainty given 95 % confidence level		1,029 mg	

NPL's data is used because they are the most recent and because they are derived from a process identical to the weighing and loading procedure of the current project. This leads to that the uncertainty $U(\Delta w) = 1,03 \text{ mg}$. U is calculated based on confidence interval 95 %, which is the normal in relation to reporting of measurement results.

As an illustration of the significance of the uncertainly, two measurements ($\Delta w = 2,3$ og $1,8 \text{ mg}$) including error bars is shown in Fig. 65 below. Further, Table 25 shows the relative uncertainty of U_{rel} as a function of PM mass figures in the range of 1-10 mg.

Fig. 66: Uncertainty in PM determination for two weighing examples

Table 25: Relative uncertainty of ***U rel*** for PM mass figures in the range of 1-10 mg.

dw (mg)	1	2	4	6	8	10
U rel. (%)	103	51	26	17	13	10

Figs 67 and 68 show details of DTI filter holders. It's quite obvious that the one having filter support of expanded metal is more likely to cause loss of filter material than the one having filter support of wire mesh.

The filters from measurements on boilers were treated in a slightly different way. Both the primary filter and the back-up filter was weighed together at the same time. This means that the uncertainty must be calculated accordingly. This is made below.

Figs 67 and 68: Details of DTI filter holders

7.6 Filter handling and moisture

7.6.1 Moisture in the filter holder

During some of the measurements on wood boilers there was a problem with some moisture in the filter holder and cyclone. This happened mainly in the first measurements on each batch. This is probably caused by the early rapid water vaporizing from the wood. In order to minimize the moisture the probe was kept at 50°C and the cyclone and filter holder was insulated.

To ensure that all the moisture in the filter was gone before the weighing they were kept in an desiccator for several days.

To speed up the drying in future projects it may be a good idea to dry the filters in an oven at low temperature, for example overnight. According to Sunset Labs, there is little or no risk that anything will happen to the EC/OC content below 80°C.

7.6.2 Filter treatment

The filters are 90mm quartz fiber filters that are preheated to 800°C from the factory. Before the measurement the filters were stored in a desiccator for approx. 12h. After the measurements the filter were also stored in a desiccator long enough to ensure that the filters were dry.

After the weighing a 47 mm circle of the filters were cut out to analyze in the OT21. After the analysis in the OT21 had been made, the filters were stored in petri dishes pending transport to Sunset Labs for analysis.

8 Conclusions

The following conclusions can be made from the project:

1. For domestic central heating boilers, the EC emissions as average values (including ignition) were between 0,5 and 35 mg/MJ; the mean value being 11 mg/MJ. For residential heaters, the EC emissions varied between 3 and 122 mg/MJ. The mean value was 32 mg/MJ.
2. The OC emissions for boilers varied between 6 and 776 mg/MJ, and the mean value was 96 mg/MJ. For residential heaters, the OC emissions varied between 6 and 441 mg/MJ, and the mean value was 76 mg/MJ.
3. For PM 2,5 emissions, the average values for boilers varied between 14 and 1975 mg/MJ for boilers. The mean value was 216 mg/MJ for all boilers. For residential heaters, the emissions varied between 35 and 821 mg/MJ, and the mean value was 160 mg/MJ.
4. EC, OC and PM 2.5 emissions are much lower for modern log wood boilers than for old-type boilers. The mean values for EC emissions were 5 and 24 mg/MJ, and for OC emissions they were 15 and 290 mg/MJ respectively. For PM 2.5 emissions, the mean values were 42 and 777 mg/MJ.
5. Comparing integrated pellet boilers with modern wood boilers, the emissions of EC, OC and PM 2,5 were almost the same. The mean values for EC were 3,5 and 5 mg/MJ respectively, and for OC 18 and 15 mg/MJ. For PM 2,5, the mean values were 29 and 42 mg/MJ.
6. The emissions of EC, OC and PM 2,5 were significantly higher from the pellet burner in a combination boiler than from the integrated pellet boilers.
7. For old-type boilers, the emissions of PM 2.5, OC, unburnt hydrocarbons and CO are much higher at part load than at nominal load. This is also generally the case for residential heaters. The difference is larger for the older stoves than for the modern stoves. However, the EC emission does not differ much between part load and nominal load for neither boilers nor residential heaters.
8. When firing an old-type boiler at part load with small fuel batches, all the emissions are in the same level as those at nominal heat load.
9. As for boilers, most of the residential heaters show higher OC emissions than EC emissions. However, those who have very basic combustion air systems (i.e. mainly primary air) show higher EC emissions than OC emissions. The EC emissions for these stoves are highest among all stoves.
10. The state-of-the-art stove and the continuously burning pellet stove showed the lowest emissions of both EC and OC.
11. EC, OC and PM_{2,5} emissions are all significantly higher during the ignition phase than during following fuel batches. This is valid both for boilers and residential heaters.

12. The correlation between EC and PM 2,5 emissions is very weak both for boilers and residential heaters. This indicates that EC is only a small part of PM emissions; this is also shown by the numerical values of EC and PM 2,5 emissions respectively. In contrast, the correlation between OC and PM 2,5 emissions is very strong both for boilers and residential heaters.

13. When comparing BC values from the aethalometer with the corresponding EC values from thermo-optical analysis, there is a clear correlation. The absolute values do however not coincide very well, the EC values being about 2,5 times higher than BC values. The reason for this could not be explained within this project, but it is recommended to further investigate the aethalometer's possibilities for application in emission measurements. The correlation between "UVPM" values from the aethalometer and OC values are very weak, but it is recommended to study also this issue further.

14. In general, the measured values are well in line with what was expected from earlier experience concerning both levels and relations between different product types

9 Future work

Since science is moving continually forward on the BC issue, how to define it and how to measure it, it is recommended to sort of 'calibrate' the Nordic understanding of Black carbon and perhaps other equal important fractions against the most recent scientific achievements, and equally important against the most recent understanding of the climate and health impact related to those fractions.

This could lead to a clear perception of the carbonaceous fractions we want to focus on, preferable not just in relation to domestic wood burning, but ideally a common perception for all the different combustion processes, emitting carbonaceous particles to the atmosphere.

Then we need to define how to measure the fractions and to decide what we want to do with the results.

The Nordic countries are all due to implement the Ecodesign regulation on stoves and boilers; for solid fuel boilers from 2020 and for the stoves from 2022. Having got a PM limit on board, the Ecodesign regulations already, it is unlikely that an auxiliary measure like BC could be introduced nationally in the Nordic countries before that. After the revisions of the Ecodesign regulations, coming for boilers 2022 and for stoves in 2024, BC and other limits might be installed as European initiative, provided there is an standardized measurement method and limit values commonly agreed upon.

On the other hand, this gives us good time to decide how we want to represent and to measure BC and other relevant substances.

Bearing in mind BC is just another way of representing soot and particles mixed up of condensed volatile organic composition, we need to draw clear lines between the classic PM/dust and OGC approach and the new BC/BrC/EC/OC approach, as to prevent overlapping representation of the same substance in several parameters.

Opposed to the current PM emission factors which are divided into classes by stove age, there is no strong indication, that emissions of BC/EC displays the same stove age relation. Other parameters seems to predict the formation of BC/EC and this could be worth a closer study.

A more specific issue to work further on, which may contribute to an easier way of quantifying BC related emissions, is to further investigate the possibilities for using the aethalometer in BC and OC emission measurements. Implementing a method which do not require lab analysis is a very attractive possibility.

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SP Technical Research Institute of Sweden

Box 857, SE-501 15 BORÅS, SWEDEN

Telephone: +46 10 516 50 00, Telefax: +46 33 13 55 02

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SP Report 2016:06

ISBN 978-91-88349-13-2

ISSN 0284-5172

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