European inter-comparison campaigns on test methods to determine particle matter emissions (PME) from residential heating appliances and boilers burning solid fuels performed within the EN_PME_TEST Eranet project

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1. Introduction

Domestic heating using biomass combustion is strongly involved in the development of renewable energy. However, it can be associated to particulate matter emissions (PME) characterized by a distribution of very fine particles and large amounts of organic condensables, which strongly contribute to PM ambient air concentrations. Recent discussion within the CEN TC 295 WG 5 looking at methods for the measurement of atmospheric emissions from residential wood combustion (RWC) have come to the selection (prEN 16510-1 draft standard[1]) of two methods based on the DIN+ [2](heated filter) combined to organic gaseous carbon (OGC) measurements using Flame Ionization detector (FID) and the NS3058[3] (dilution tunnel).

The prEN16510-1 describes the collection of PME using a heated filter maintained at 160°C combined to OGC measurements using FID at 160°C or using a filter under ambient conditions after dilution using a dilution tunnel (NS 3058). A correlation obtained thanks to results of trials performed within the CEN TC 295 WG5 actions in 2011, is used to connect the results obtained by both methods. However, although the prEn16510-1 allowed to reach a consensus, there has been no agreement on the choice of a unique European method moreover, there are still some issues to be addressed. For example, it does not propose the inclusion of a cut-off of the aerosol prior to collection, the question of the temperature of the flue gas in the filtration area and organics OGC measurement which is a key parameter is not addressed.

The objective of the EN_PME_TEST as a co-normative research project is to develop and validate a common European test method to determine PME from residential heating appliances and boilers burning solid biofuels which shall be the basis for a European standard for PME-measurement. Special emphasis is given to the scientific basis for the candidate method and its relevance to health and environmental issues. The work is performed by a consortium of 17 partners (research and test establishments) from 10 countries, under the umbrella of the ERA-NET Bioenergy with the financial support of public authorities, national environment agencies as well as self-funding of some of the partners.

A key step of the program consists in performing inter-laboratory-comparisons (ILC) during which several sampling teams (4 to 12) meet and carry out simultaneous PME measurements from solid fuel burning appliances and boilers using selected candidate methods. The review of existing methods achieved earlier by the partners, lead to the selection of several candidate methods:

- for the short to medium term: a method based on prEN 16510-1, solid mass determined on a heated filter/ VOCs determination by FID measurements,
- for the long term method, several approaches are considered: online mass measurement dedicated to RWC (new techniques commercially available since 2013), dilution approaches (portable and simple dilution devices), secondary organic aerosols (SOA) formation potential measurements (use of a micro smog chamber).

The ILCs are being performed:

- at INERIS (France) on a horizontal flue duct testing facility initially designed for gas emissions. This facility is equipped with 12 sampling ports and is connected to a pellet boiler as the flue gas source,
- at VSB (Czech Republic) on a horizontal or vertical flue duct testing facility. This facility will be connected to a log wood stove.

The paper proposed here presents:

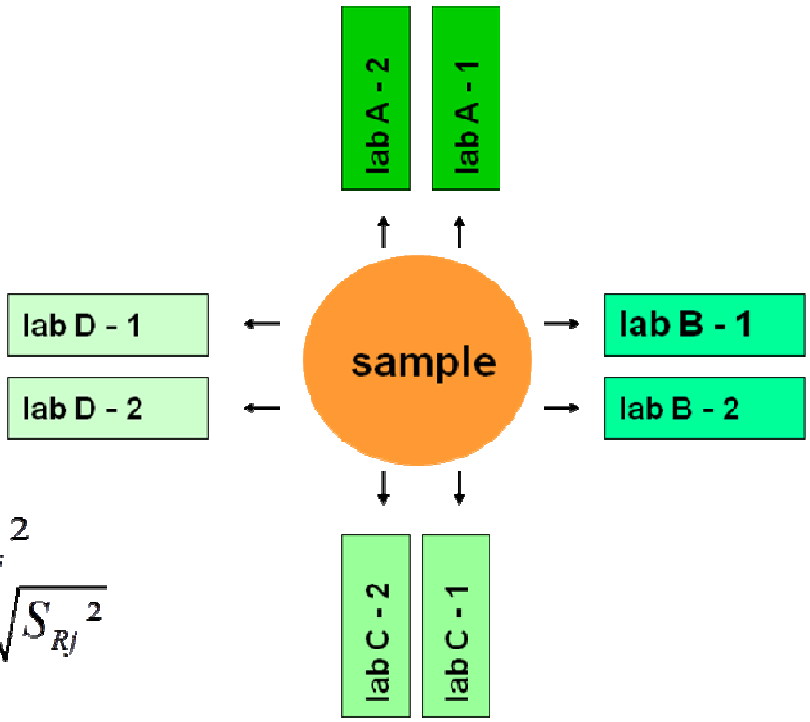
- the principle and objective of such ILCs campaigns,
- the work that was performed on the INERIS horizontal testing facility prior to the ILC in order to provide several levels of PM emissions and evaluate the homogeneity of the emission levels along the 12 sampling ports,
- preliminary results obtained during the ILC performed at INERIS regarding the comparability of the tested candidate methods and their level of uncertainty.

2. Principle and objectives

Performing an inter-comparison study is a possible way to evaluate the uncertainty of measurement methods. It takes part to the standardisation process of measurement methods. It is a very valuable way of detecting major sources of uncertainty. It is also useful to disseminate good practices to evaluate the capabilities of different laboratories to apply standard measurement methods.

A sample is provided to several laboratories and the results obtained are compared. A description is provided in Picture 1 . In principle, each laboratory uses two sets of measurement method, which allows the determination of the intra-laboratory dispersion (s_r) which qualifies the scattering of the results within each laboratory, whereas the inter-laboratories dispersion (s_L) which qualifies the scattering of the results between laboratories is evaluated using the results of all participants. This leads to the calculation of the global uncertainty of the method (ISO5725-2[4]).

Picture 1 : Principle of an inter-comparison



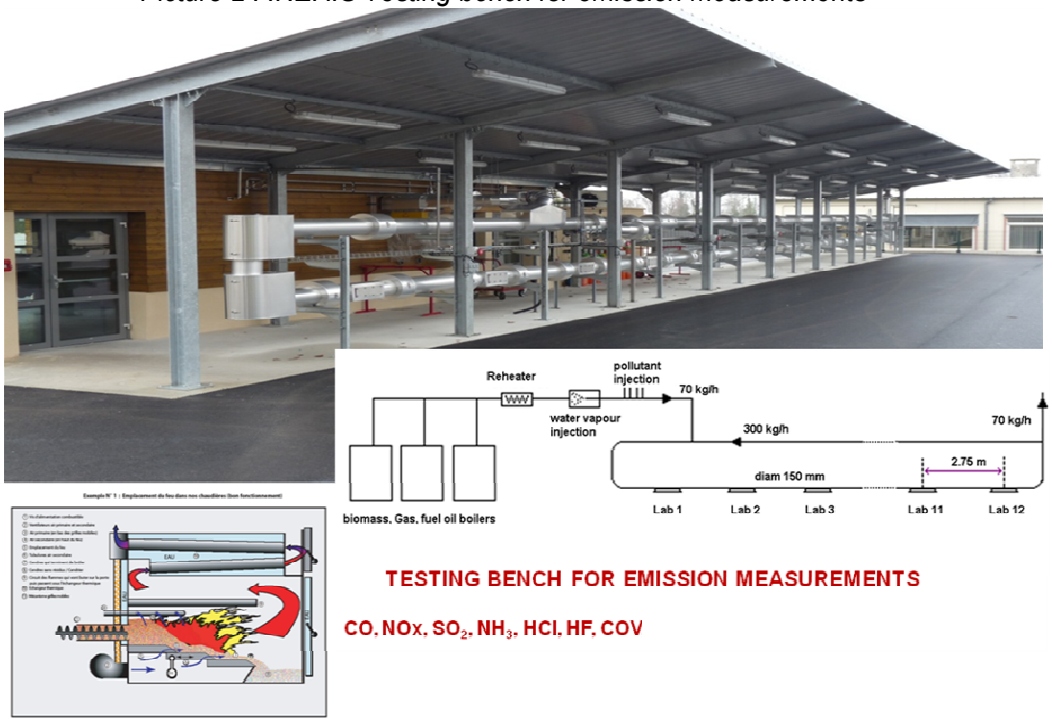
$$S_{Rj}^{2} = S_{rj}^{2} + S_{Lj}^{2}$$

$$\rightarrow \quad U(y) = k.\sqrt{S_{Rj}^{2}}$$

The aim of the work package of EN_PME _TEST dedicated to inter-comparison studies, is to perform two ILCs:

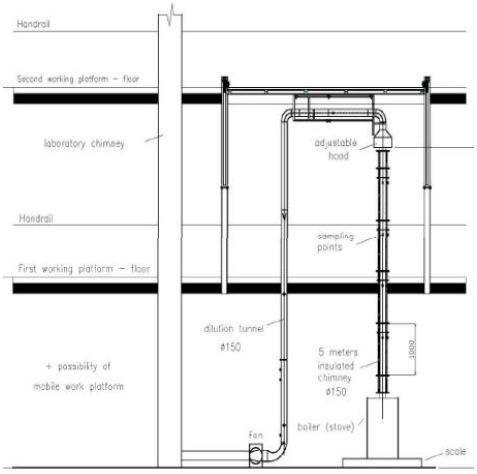
- the first one at INERIS (France) in February 2014, using the testing bench for emission measurements, an horizontal testing facility equipped with two sampling ports and connected to a pellet boiler ;

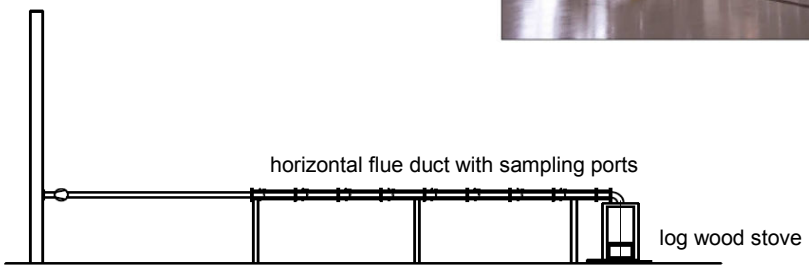
Picture 2 : INERIS Testing bench for emission measurements



- the second one at VSB (Ostrava, Czech republic), in October 2014. For this second inter-comparison campaign, the set-up horizontal or vertical is still to be defined, the emission source will be a log wood stove.

Picture 3 : VSB biomass testing facility





3. Validation of the INERIS horizontal testing facility

Material and method

The homogeneity of the sample delivered to participants of an inter-comparison is the key factor of success of such campaings. As a result, prior to the realisation of an inter-comparison campaign on PM emissions using the INERIS testing facility, it was necessary to evaluate the homogeneity of the emission levels of PME along its twelve sampling ports. This horizontal facility initially designed for gas emissions has been used for several years by INERIS to perform ILCs on gaseous regulated compounds. In France, air pollution control laboratories are obliged to participate at such ILCs every three years in order to obtain an agreement from the French ministry of environment to perform measurement on industrial sources. The results obtained on different gaseous pollutants have been previously presented[5]. The testing facility is equipped with three different types of combustion sources : a gas boiler, an light fuel oil boiler and a pellet boiler. Since the gas and oil boiler emit very low levels of dust, the validation of the homogeneity for PME emissions was performed using the pellet boiler as a source.

Table 1 : Validation trials description

	Site blank at the beginning of the sampling day	Number of sampling port equipped						Site blank at the end of the sampling day	Fuel	Number of repetition
Trial 1		SP1	SP3	SP5	SP7	SP9	SP11		Pellets	6
Trial 2		SP2	SP4	SP6	SP8	SP10	SP12		Pellets	6
Trial 3	In situ system 2	SP2	SP4	SP6	SP8	SP10	SP12	In situ system 2	Pellets	6
Trial 4	In situ system 8	SP7	SP8	SP9	SP10	SP11	SP12	-	Pellets	6
Trial 5	In situ system 2	SP2	SP4	SP6	SP8	SP10	SP12	In situ system 4	Wood chips	6

The validation was performed using two different fuels, pellets and wood chips, aiming at generating 4 repeatable and stable levels of concentration, by applying to each fuel different combustion conditions:

- concentration level around 10 mg/m$^3_{STP}$ pellet combustion,optimal conditions
- concentration level around 25 mg/m$^3_{STP}$: pellet combustion,
- concentration level around 35 mg/m$^3_{STP}$: wood chips combustion, optimal conditions
- concentration level around 55 mg/m$^3_{STP}$: wood chips combustion.

A first step consisted in validating those levels of concentrations by measuring the dust emissions in sampling ports 1 and 12 simultaneaously.

The homogeneity of the dust matrix in the 12 sampling ports was then tested by performing 5 trials in which 6 sampling ports were simultaneously equiped using in situ dust sampling systems, each trial was repeated 6 times, and on site blank was taken during each trial. As a result a total of 180 samples and 5 site blanks were taken and weighed afterwards following the EN 13284-1[6] weighing procedure. The following table presents a description of each trial.

The data was evaluated following two criteria:

- the absolute gap obtained between sampling ports was compared to the limit of quantification (LOQ) and to the reproducibility confidence intervals of the measurement method used ;
- the location of the sampling ports associated to the extreme values has been studied in order to evaluate if a decrease of the concentration along the sampling ports could be observed;

A statistical analysis based on the ISO 13528[7] standard was also performed in order to evaluate if the inter-sample dispersion S_t has to be taken to evaluate the capability $\hat{\sigma}_j$ of participants of future intercomparison exercise.

Results

- Preliminary trials aiming at validating the levels of concentrations to be generated indicated that it was very difficult to generate stable and repeatable levels of concentration for a given fuel and combustion conditions set-up. Moreover, the levels of concentration generated was not connected to what could have been expected according to the fuel and set up applied. This is probably due to the fact that the draft in the boiler is quite sensitive to the operations of opening and closing the sampling ports that were necessary to take the samples. Due to these operations, the draft i.e the quality of the combustion could not be maintained during each trial and between two trials performed using the same fuel and combustion set-up. The levels of concentrations generated using the two fuels are presented in the following table, they show that the two types of fuels lead to similar levels of concentrations.

Table 2 : Average concentration generated in the testing facility (mg/m$^3_{STP}$) according to the fuel used and the trial number

Fuel	Average concentration generated in the testing facility (mg/m$^3_{STP}$)		Trial
pellets	20,9 (8,0 – 54,4)	14,9 (8,2 – 21,3)	1
		13,2 (8,0 – 21,4)	2
		29,1 (14,6 – 47,8)	3
		31,9 (14,2 – 54,4)	4
wood chips	23,8 (10,7 – 34,7)		5

These results lead to the conclusion that the trials aiming at validating the homogeneity of the matrix along the sampling ports cannot be performed using stable matrix composition and it is not possible to target nor to repeate a given level of concentrations.

Blank levels observed are presented in the following table, all data are below the limit of quantification.

Table 3 : Field blank description and values

	Date	Field blanks		Concentration measured (mg/m$^3_{STP}$)	Fuel
Trial 1	20/11/2013	In situ system 2	beginning of the sampling day	< 0,6 (< LOQ/3)	pellets
Trial 2	20/11/2013	In situ system 2	end of the sampling day	< 0,6 (< LOQ/3)	pellets
Trial 3	21/11/2013	In situ system 8	end of the sampling day	< 0,7 (<LOQ/3)	pellets
Trial 4	22/11/2013	In situ system 2	beginning of the sampling day	< 0,7 (<LOQ/3)	pellets
Trial 5	22/11/2013	In situ system 4	end of the sampling day	< 0,7 (<LOQ/3)	Wood chips

The results of the 5 tests of homogeneity are presented below. They indicate that the maximum gaps between concentrations measured at the sampling ports are comprised between 2,8 and 6,4 mg/m$^3_{STP}$ which represent from 11,7 to 23,6 % of the average generated concentration.

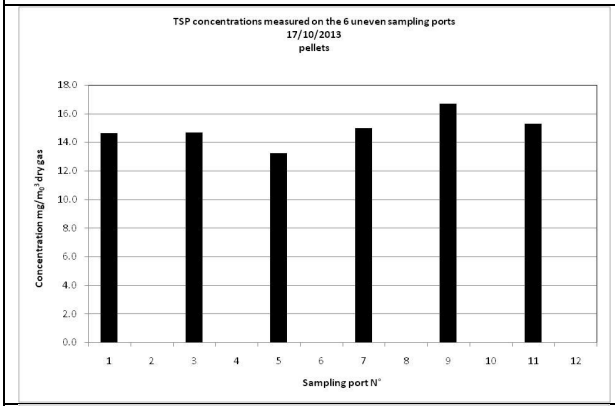
These data have to be compared for consistency with:

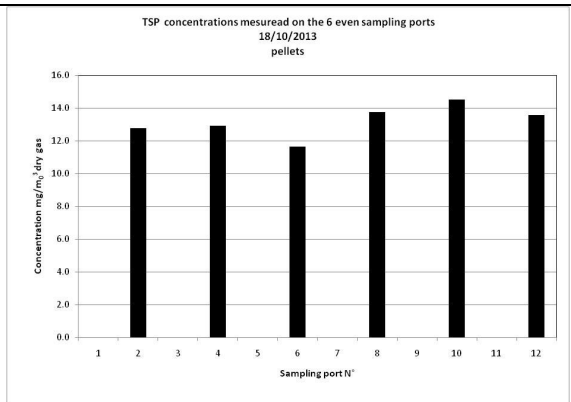
- the LOQ of the method obtained according to the measurement conditions was comprised between 1,7 and 2,3 mg/m$^3_{STP}$
- the reproducibility confidence intervals of the measurement method used to determine concentrations below 50 mg/m$^3_{STP}$ are:
  - 22,6 % for an average concentration of 27 mg/m$^3_{STP}$ according to NF X 44-052[8] standard),
  - 90 % for an average concentration of 4,7 mg/m$^3_{STP}$ ([2-17 mg/ m$^3_{STP}$] (according to NF EN 13284-1 standard).

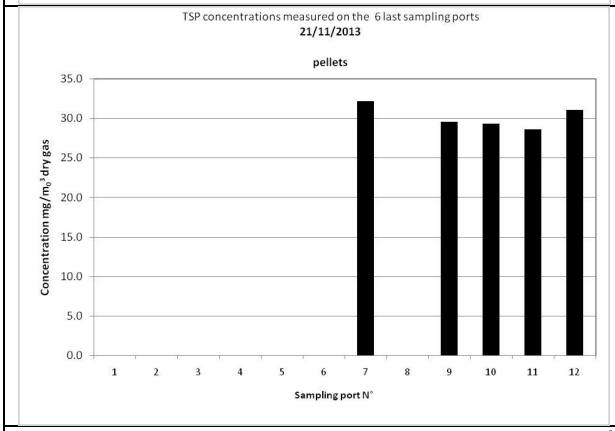
This comparison shows that the gaps of concentration determined between sampling ports are strongly influenced by the performance of the measurement method used.

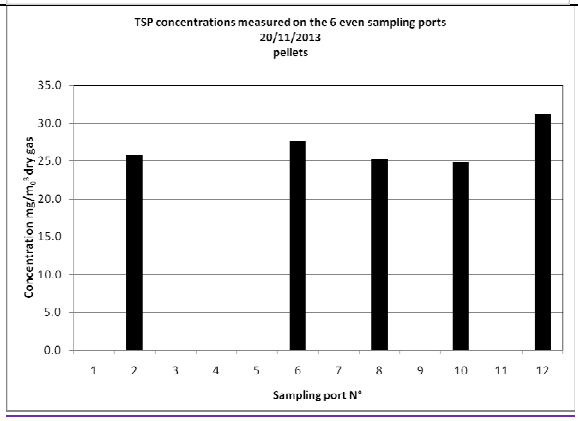
No losses along the sampling ports were observed, no systematic decrease of the measured concentration is observed from sampling port 1 to sampling port 12.

pellets

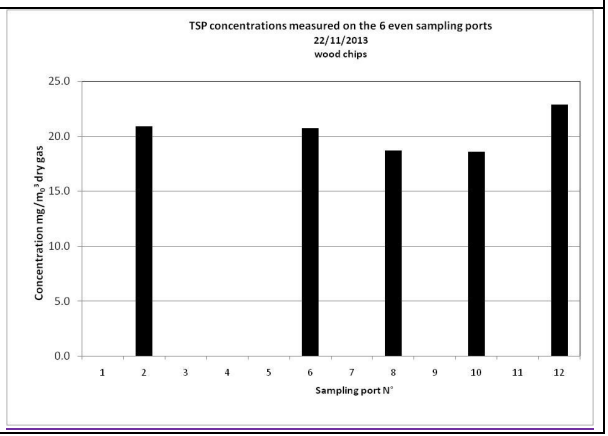








wood chips



4. Inter-comparison campaign performed at INERIS in February 2014-04-11

Experimental set-up

- 13 laboratories (see Table 4) gathered the first week of February 2014 at INERIS in order to test several candidate methods selected earlier in the project:
  - for the short to medium term: a candidate method based on prEN 16510-1.The aim of the candidate method proposed here is to tackle the issues of the temperature stability and the cut-off or the aerosol :
    - the design of the offline candidate method proposes to maintain the flue gas temperature in the filtration area and for OGC measurements at 180°C in order to avoid gap or overlapping of temperature which would lead to some species missed out or accounted twice;
    - the probe design has proven to maintain the flue gas temperature in the area of filtration to 180°C for a wide range of duct flue gas temperature comprised between 40°C and 400°C,
    - the question of a simple way of cut-of for particles larger than 2.5µm based on modeling performed during the project and empiric calculation proposed by Tsuji et al by[9], is also raised by this candidate method. Therefore backwards particle sampling was conducted.

    Moreover, the issue of the contribution of the deposition in the probe, and the importance of rinsing between trials is also of interest.

  - For the long term method, several approaches are considered:
    - online PME measurement dedicated to residential wood combustion that were recently developed and made commercially available (online method A, B and C),
    - dilution approaches (portable and simple dilution devices) combined to online mass (TEOM) or size distribution measurements (OPC),
    - SOA formation potential measurements (use of a micro smog chamber),

Table 4 : List of participants to the INERIS inter-comparison

Country	Laboratory
Austria	BE2020
Czech republic	VSB
Denmark	DTI
France	CSTB
	CTIF
	INERIS
	LERMAB
Germany	TFZ
	DBFZ
Italy	ISSI/ENEA
Sweden	SP
Switzerland	CATSE
	FHNW

The sampling duration was set to 30 minutes. The first two days of measurements were dedicated to the short term method approaches, whereas long term method approaches were tested during the last day of measurements. A test of repeatability of measurements using two offline candidate methods on top of each other at the same sampling port was also performed on the last day of measurements.

Table 5 : Combustion fuel and number of samples taken per day of measurement

Day	Fuel	Number of samples taken
04/02/2014	pellets	5
05/02/2014	wood chips	6
06/02/2014	wood chips	5

Table 6 : List of methods applied to the different sampling ports, days one and two

Sampling port N°	PME method	*OGC method*
1	prEN16510-1	*FID*
2	DIN+	FID
3	Offline candidate method	FID
4	DIN+	FID
5	Offline candidate method	FID
6	Offline candidate method	FID
7	Offline candidate method	FID
8	Offline candidate method Online method B	FID
9	-	FID
10	Offline candidate method	FID
11	Offline candidate method	FID
12	Offline candidate method	FID

Table 7 : List of methods applied to the different sampling ports, day three

Sampling port N°	PME method	OGC method
1	prEN16510-1	FID
2	Porous tube dilution+TEOM	FID
3	Online method B	FID
4	Offline candidate method *2 Online method A	FID
5	Disc dilution +OPC+µsmog chamber	FID
6	EN13284	FID
7	Online method A	FID
8	Online method B EN13284	FID
9	-	-
10	Online method A	FID
11	-	-
12	Online method C	FID

Preliminary results

The following graphs show:

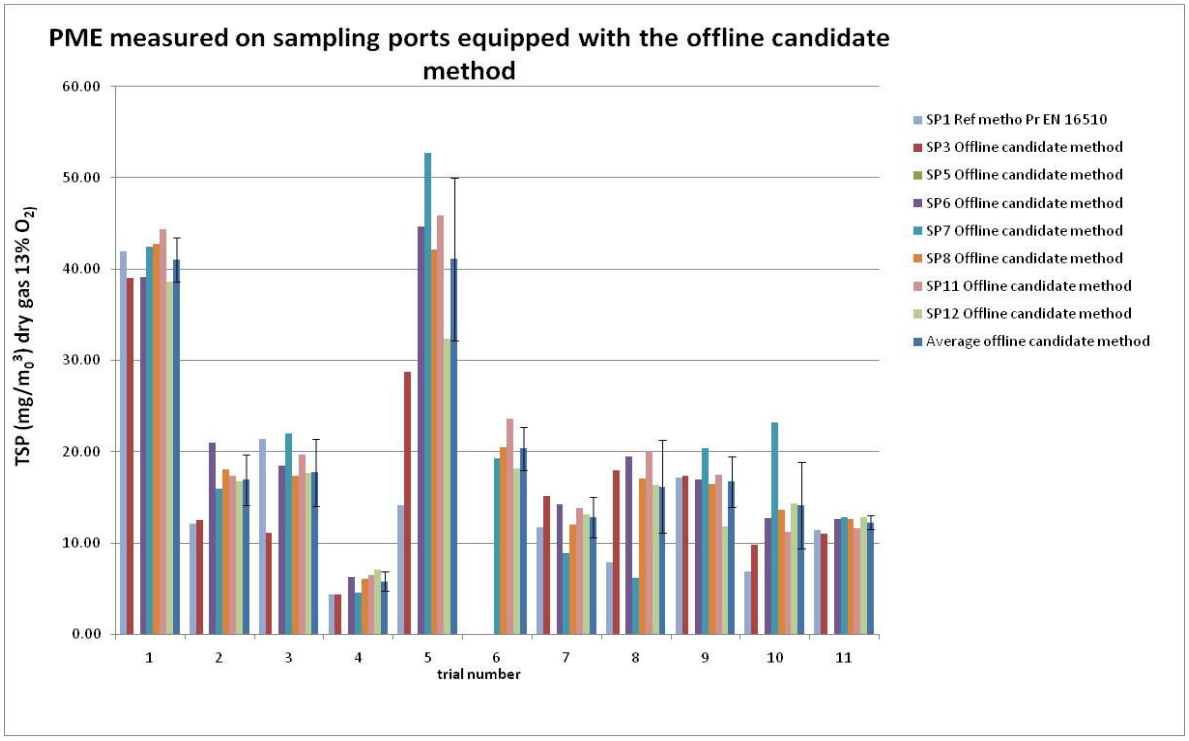
- the results obtained for the PME offline candidate method in comparison to the prEN16510-10 reference method set up on days one and two;
- the results obtained by two PME offline candidate methods set up in the same sampling port (one above the other) on day three;
- results obtained for OGC measurement using FID for days one, two and three.

For the offline candidate method:

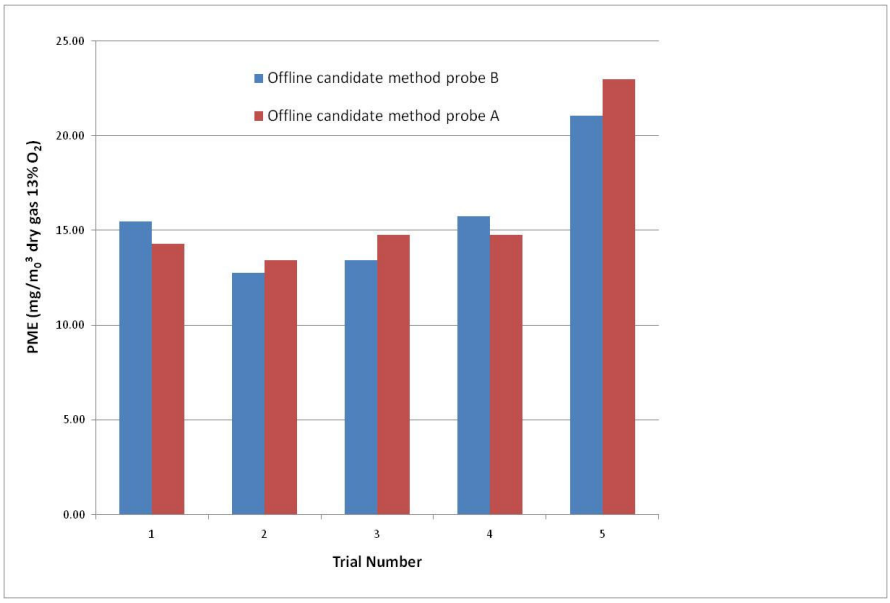
- the results of the first two days (n=11 trials) of measurements lead to gaps between average results of each sampling port and the global average comprised between -15% and +6% of the global average;
- tests of repeatability performed the last day lead to an intra laboratory variance (s_{rj}^2) comprised between 2% and 8% of the average concentration.

For OGC measurement using FIDs, the gaps between average results ((n=16 trials) of each sampling port and the global average were comprised between -34% and +25% of the global average.
The results of both short terms and long term methods are still being analysed. They will be made available later on.

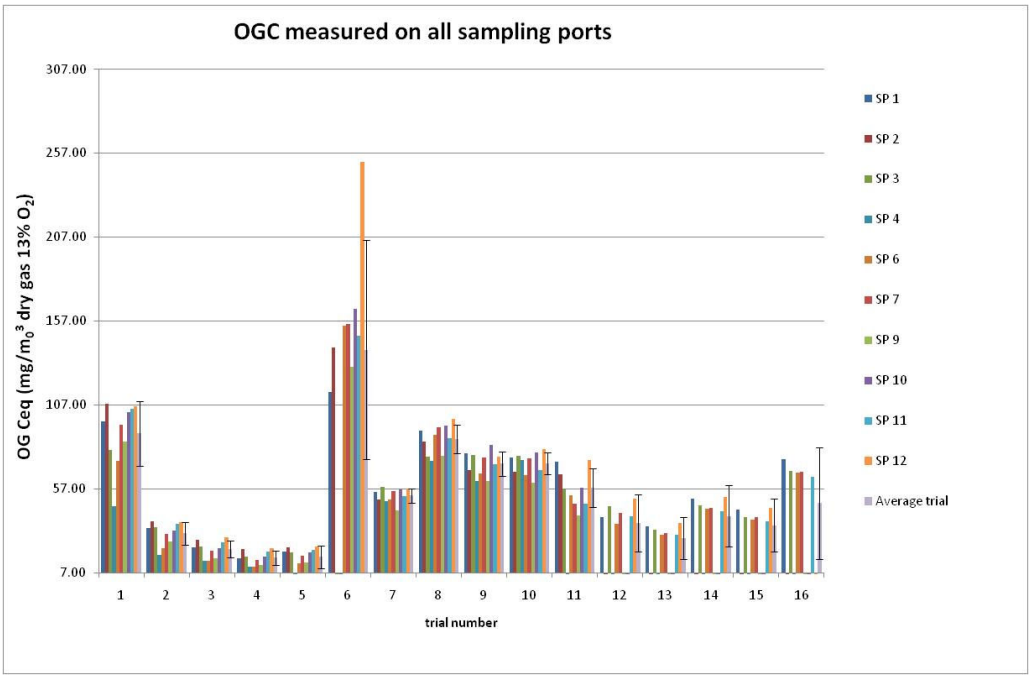
Graph 1 : Results obtained for the PME offline candidate method in comparison to the prEN16510-1 reference method set up on days one and two;



Graph 2 : Results obtained by two PME offline candidate methods set up in the same sampling port (one above the other) on day three



Graph 3 : Results obtained for OGC measurement using FID for days one, two and three



5. Conclusions and perspectives

The work presented here indicates that provided validation of the homogeneity of the matrix prior to measurements, it is possible to perform inter-comparison studies on particulate compounds using the INERIS emission measurement test bench. A first inter-comparison campaign took place in February 2014 within the frame of the ERA-NET program EN_PME_TEST aiming at validating a common measurement method for PME from residential wood combustion. It gathered 13 laboratories for a week during which several candidate methods selected earlier in the project were implemented and tested.

More data processing and statistical analysis are necessary to fully evaluate the results obtained during this inter-comparison campaign.

A second inter-comparison campaign using a log wood stove as the source will be performed in October 2014. All results will be forwarded and presented to stake holders such as CEN TC295 WG5, authorities, industrial, notified bodies

6. Acknowledgements

The authors are thankful to the ERA-NET, to project partners using self funding and national environment agencies for their financial support (ADEME, TEKES, Energinyndigheten, FNR, Danish environment agency, FFG, BAFU).

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