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Benjamin Cea, Isaline Fraboulet, Océane Feuger, Francesca Hugony, Carmen Morreale, et al.. Development and Evaluation of an Innovative Method Based on Dilution to Sample Solid and Condensable Fractions of Particles Emitted by Residential Wood Combustion. Sustainable Energy & Fuels, 2021, 10.1021/acs.energyfuels.1c02595 . ineris-03436128

HAL Id: ineris-03436128

<https://ineris.hal.science/ineris-03436128>

Submitted on 19 Nov 2021

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Development and evaluation of an innovative method based on dilution to sample solid and condensable fractions of particles emitted by residential wood combustion

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ABSTRACT

An innovative and simple method based on dilution, named as the dilution chamber (DC), allowing the measurement of solid and condensable fractions of particulate matter emitted by residential wood combustion appliances has been developed and its performances evaluated. DC method has then been tested by five European institutes (Ineris, ISSI/ENEA, DTI and RISE) on the advanced residential wood log/pellet stoves, under nominal output and low output combustion conditions and using different fuel types. The aim of the study was to evaluate the capability of DC method to collect the condensable fraction. DC method was compared with another manual method used to collect the solid and condensable fraction at the same time: the dilution tunnel (DT) on 4 sampling platforms. A third method, combining heated filter and impinger filled in with isopropanol collection (SPC-IPA), was also used by Ineris only for comparison with DC method. PM measurements based on DC method globally showed a linear correlation with PM measurements based on DT (R^2 ranged between 0.81 and 0.99, $p < 0.05$) specifically for the residential wood stoves under low output conditions when the condensable fraction contributes the most. An analysis and quantification of PAHs related to the total mass of PM of sampled taken by DC method performed by ENEA/ISSI showed that it produces a condensation effect of semi volatile species comparable or even greater than the DT method. PM emission factors calculated from PM measurements based on DC method were i) about 2 to 20-fold higher for the residential wood stoves (EF ranged between 201 to 2420 g GJ⁻¹) compared to those obtained for the residential pellet stoves (EF ranged between 108 to 556 g GJ⁻¹) and ii) of the same magnitude of PM emission factors from the literature or the EMEP/EEA air pollutant emission inventory guidebook.

1. INTRODUCTION

Biomass combustion is increasingly used for domestic heating in some parts of the world such as Europe since it is considered as a renewable energy ¹⁻⁵ neutral in terms of CO₂ emissions. However, the combustion of biomass in the residential sector is responsible for the emissions of a wide range of pollutants and is an important contributor to ambient air pollution, mainly in terms of fine particles ⁶⁻⁹ and organic compounds. Inhaling fine particles (PM₁₀ and PM_{2.5}: i.e. 10 to 2.5 µm or smaller in size) and organic compounds (1.3-butadiene, benzene, formaldehyde or acetaldehyde) emitted by wood combustion ^{2,9-11} is shown to be harmful to human health. Fine particles cause severe health problems (WHO, 2005) and emission sources need to be addressed to improve air quality. Contribution of wood combustion emissions to ambient air PM is due to primary and secondary particles. The primary particles, emitted from the combustion chamber and the duct, are constituted by solid and condensable fraction. The secondary particles are formed after emission, in the atmosphere by photo-oxidation phenomena, mainly from organic gaseous precursors, in the case of emissions from residential wood combustion (RWC).

The solid fraction of primary particles includes soot particles, salts and heavy organic compounds, it is formed in the fuel rich parts of the combustion chamber and the salts are condensing after evaporation when reaching temperatures of approx. 500 - 600 °C. The combustion of solid biomass, in addition to high quantities of particulate, produces high quantities of organic compounds with different boiling points: very volatile, volatile and semi-volatile (VVOCs, VOCs and SVOCs) with high contents of toxic and carcinogenic substances such as polycyclic aromatic hydrocarbons (PAHs) ¹³. Those molecules are present at the temperature leaving the combustion chamber, as gases and liquid in the flue gas, but with the dilution and the lowering of the temperature, they can form particles due to nucleation and condensation processes ¹⁴. This process leads to the formation of condensed particles. Elsasser et al. ¹⁵ found that the organic matter was the main contributor to the total mass of particles during the measurements performed by means of an Aerodyne aerosol mass spectrometer (AMS) in different burning phase of a 30kW log wood boiler (approximately 93%). Likewise, a more recent study which compared two PM measurement methods (“Hot filter” and AE33 aethalometer coupled with Aerosol Chemical Speciation Monitor) through combustion tests using a “conventional” and an Ecodesign stove using five different solid fuels also found that organic matter dominated PM composition in the range between 50 and 95% ¹⁶. So, reducing the contribution of domestic biomass combustion to ambient air particulate matter concentrations does not only imply reducing solid particulate matter emissions, but also and especially

those of the condensable fraction. Such a reduction will allow a co-benefit on the emission levels of these compounds which are pollutants as such, and which are also subject to reduction targets and monitoring in the ambient air in accordance with directives 2004/107/EC and 2000/69/EC.

There is a need to complete existing methods used to implement Ecodesign Directive (2005/32/EC) in this product area for which the main environmental characteristics of the products covered by the Lot 15 / Lot 20 are the emissions, among them PM emissions, and for which existing standards are not sufficiently adequate to quantify condensables. Moreover, there are evidences in the literature of a gap existing between predicted ambient PM obtained thanks to modelling of emissions and ambient PM observed thanks to measurements ¹⁷. The concentrations of organic particles measured in the atmosphere are higher than expected from modelling of reported emission factors (EF). The main reason for this gap is the heterogeneity that exists between emission factors reported in different countries in Europe which is due to the conditions applied during the tests i.e. appliances operating conditions and measurement methods under which the emissions factors are determined. Denier van der Gon et al. ¹⁷ showed that the emission data needed for modelling study partly originates from different methods that countries use and cannot be directly compared, as no detailed description of measurement methodology is given. Two types of sampling methods of PM emitted by RWC have traditionally been used in Europe, methods based on part flow sampling on heated filter (HF) at a temperature higher than e.g. 70°C, which samples the solid fraction of PM only (e.g. HF method: EPA Method 5H, case of the DIN + method or EN_PME_TEST method), and methods based on the dilution of the full flue gas using ambient air in a dilution tunnel (DT) combined to a filter maintained at 30 °C (e.g. case of the NS 3058 method, and US EPA 5G method), those methods are inventoried in Janhäll et al.¹⁸. Dilution of the flue gas with lower temperature air allows condensation phenomena to occur, and the condensable phases to condensate on the particles prior to sampling on the filter. The sample collected at the outlet of the dilution tunnel includes both the solid and condensable phases of PM. Condensable can also be collected by absorption in a cooled solution contained in washing bottles or impingers placed after the heated filter. The liquid solution can be water (US EPA 5H method) or isopropanol (case of the SPC-IPA, Fraboulet et al. ¹⁹, method developed by Ineris). Portable methods based on dilution of part flow sampling such as CTM-039 (EPA, 2004) can be used to collect both solid and condensable fractions of PM from combustion processes. The influence of PM measurement methods used on reported EF has previously been documented by Nussbaumer et al. ²⁰, Seljeskog et al. ⁴ and most recently

by Trubetskaya et al.¹⁶. Nussbaumer et al.²⁰ proposed a detailed survey and review of the various emission factors used in Europe, also in relation to the type of measurement techniques. They showed how emission factors varied between measurements of solid particles (indicated as SP), solid particles plus condensable found collected thanks to impinger (SPC), and total particles collected in a dilution tunnel. Seljeskog et al.⁴ performed tests using a wood log stoves operating under different conditions combustion (fuelled by birch and spruce, operating at high output /nominal output/and low output) where HF and DT methods were used simultaneously to measure PM emissions. In average, over the tests performed, the mass of particles collected with the DT method was about 6,5 times (range 4 for birch to 8 for spruce) higher than the mass of particles collected with the HF method. Trubetskaya et al.¹⁶ compared “hot filter” method and a method coupling AE33 (aethalometer) with ACSM (Aerosol Chemical Speciation Monitor) to characterise the PM emissions from combustion of five solid fuels placed in a “conventional” and an Ecodesign stove. The authors showed that PM EFs ranged from 0.2 to 108.2 g GJ⁻¹ for solid fuels and the PM EF strongly depended on the measurement method employed and on user behaviour. The use of reliable and comparable methods allowing the determination of both the solid and condensable fraction of PM emitted by domestic wood combustion is necessary to determine comparable emission factors from appliances across Europe in order to provide homogeneous emission inventories from one country to another and contribute to close on the long term the gap between measured and modelled ambient PM. But so far, there is no consensus in Europe on the choice of a method to do so although several established standard methods of PM sampling exist in Europe. These methods are listed in the report of Janhäll et al.¹⁸. In the present paper, an innovative and simple method based on dilution, allowing the measurement of solid and condensable fractions of particulate matter emitted by residential wood combustion appliances has been developed and its performances evaluated. The method, named as the dilution chamber (DC) developed by ISSI/ENEA in collaboration with Dado Lab is based on CTM 039 method from US EPA²¹ and was developed using CFD simulation. The condensation rate of SVOCs (19 PAHs) present in a flue gas emitted by 2 pellet and 3 wood log stoves collected by the DC method and a dilution tunnel (DT method) have been compared.

The performance of the dilution chamber has been evaluated by five European institutes: Ineris (French National Institute for Industrial Environment and Risks), ISSI (Innovhub Stazioni Sperimentali per l’Industria) and ENEA (Italian National Agency for New Technologies, Energy and Sustainable Economic Development), RISE (Research Institutes of Sweden) and DTI

(Danish Technological Institute). The results of characterization of PM (solid and condensable fractions) emissions from residential wood log and pellet stoves using the DC method on the one hand, the DT method (on 4 sampling platforms) and the SPC-IPA (on Ineris platform only) on the other hand, have been compared. A total of 80 combustion tests were performed. Based on the PM measurements performed with the DC method, emission factors have been determined for the different conditions tested and results compared to the literature.

2. EXPERIMENTAL

2.1. Description of the stoves tested. Two types of stoves were used: wood log stoves and pellet stoves. Each partner was free to choose the advanced stove design models. The test conditions of combustion devices are presented [Table 1](#). The residential pellet stoves (RPS) and the residential wood log stoves (RWS) chosen as well as the fuel type used depended on the institute because the aim of the experimental campaign was to compare the results of the three different sampling methods and to test the performance of DC, not to verify its replicability.

2.2. Stove operating procedures. The test durations varied from about 20 to 30 minutes for the RPS to 30 to 45 minutes for the RWS. The RPS were heated up prior to measurements by automatic operation and the sampling started 1 hour later during the steady state operation. The RWS were heated up by firing a batch of wood logs prior to measurements to avoid influence of initial transient phase on emissions. Then, when CO₂ levels reached 4%, a new batch of wood was loaded, and the measurements started just after the door was closed. As the burn-out phase ended, a new batch of wood was loaded. RISE and Ineris performed combustion tests with 2 kg load of wood at start-up (arranged according to the recommendations given by the manufacturer) before introducing two equally sized pieces of wood stacked in a T-shape with one piece on top of the other (about 1.4 kg) for the following batches. ISSI/ENEA carried out combustion tests with about 2.4 kg load of wood (following the recommendations of manufacturer). And for DTI, wood logs were cut from beams with square cross section and assembled prior to firing with three parallel logs in the bottom, and two logs on top for each batch (Table 1). The burning tests were performed under two different combustion conditions: nominal output and low output at Ineris and nominal load only for wood log at ISSI/ENEA, DTI and RISE (Table 2). The nominal output and low output conditions were defined according to the O₂ intake in the combustion chamber (maximum to minimum intake, respectively) to sustain the fire. RISE tested two fuel types

(birch with or without bark and spruce without bark) during the RWS tests. The emissions are expressed as concentration in milligrams of pollutants per m³ under STP conditions, on dry gas and corrected to 13% O₂.

2.3. Sampling set-up and measurement methods. During each combustion test, measurement of combustion gases was performed using online measurement techniques while particulate matter was sampled using the DC, DT and SPC-IPA methods (Ineris only), measurement and sampling methods were set-up simultaneously. The measurement set-up is presented in Figure 1: Residential wood log/pellet stove (1), chimney (2) and dilution tunnel (17). The first sampling line (SPC-IPA method) included: a heated probe (3), a heated filter, a filter holder and quartz filter (4), a cooling bath (5), washing bottles filled in with isopropanol (6), a foam PUF (7), a desiccator (8) and a sampling pump (9). The second sampling line (DC method) was composed of a heated probe (10), a dilution cabin, at a controlled temperature around 35-40°C, containing the dilution chamber (DC), the thermocouple measuring the diluted gases temperature, the dilution air heater (11), a flow regulation unit, to collect the exhaust gases from the chimney with the desired dilution ratio (12), a laptop connected via Bluetooth, with the control software (13) an air compressor (14), a desiccator (15) and a sampling pump (16). The third sampling line (DT method) comprised: an in-stack probe (18), a filter holder and a quartz filter (19), a desiccator (20) and a sampling pump (21). The fourth sampling line (before dilution) was composed of: a heated probe and a line for O₂, CO₂, CO and OGCs analyzers (22) monitored by computer (24). The fifth sampling line (after dilution) included: a heated probe and a line for O₂, CO₂ and CO analyzers (23) monitored by computer (24).

2.4. Combustions gases measurement methods. O₂, CO₂ and CO were measured by Ineris using a HORIBA PG250; RISE used a HORIBA PG 350, a Rosemount Analytical XSTREAM and a M&C PMA10, while ISSI/ENEA used TESTO 330-2 for raw gas sampling and TESTO 350 for diluted gas sampling, and DTI used an ABB IR-analyzer (Type AO2020). The organic gaseous compounds (OGCs) were monitored by Ineris and RISE using a JUM FID, by ISSI/ENEA using a PCF Elettronica Mod 2001/C and by DTI using a M&A thermo FID.

2.5. Particulate matter measurement methods. The CTM-039 US/ EPA method, which the DC method developed and tested here is based on, is a portable sampling train which causes a mix of the sampled gas with dilution air allowing the condensation semi volatile species on solid PM. The DC method is described in a paper from Hugony & al.²¹. The residence chamber designed for CTM039 has a volume of around 3.5 x 10⁻³ m³, the dilution ratio suggested is between 1:10 to 1:40. Different

sampling gas flows are proposed for different temperatures. According to the range of gases temperatures in wood combustion, an appropriate range of flow between 10 to 20 l min⁻¹ leads to a residence time in the dilution chamber between 0.2 to 2 s. A plane filter with a diameter of 142mm collects PM at the very end of the sampling train. To design the dilution system ISSI and ENEA took advantages of several tests performed with a sampling train corresponding to the CTM039 ^{22,23}, which guarantees the sampling of condensable PM. The need to reduce the volume and simplify the sampling regulation of the flows, lead in 2016 to the design of the new method. The key parameters considered were the way of mixing the two flows (sampling gas and dilution air), the contact time between the two gases before filtering and the isokinetic or non-isokinetic sampling. The low and varying gas velocity in fumes emitted by RWC appliances, in general lower than the quantification limit of Pitot tubes techniques, and the high dust content of these flue gases, make it nearly impossible to adjust the sampling rate, as required in the iso-kinetic particulate sampling. For this reason, a constant flow rate has been adopted here, as in most methods used in this field. This approach is not significantly detrimental since most of the particles emitted by RWC are smaller than 2.5 µm (PM_{2.5} > 94% of total PM) ²⁴ and their behavior can be considered as similar to gases. The dilution chamber has been designed for a residence time up to 3s. 2-4s which is quite a typical range for dilution sampling methods based on dilution tunnel ^{25,26}. The new designed method is in principle applicable to all processes (industrial, centralized thermal plants or domestic heat appliances) where the emissions of VOC and SVOC is significant. The prototype used in this work is designed for laboratory tests on domestic application and it is not suitable for field measurements. To collect condensable particles the dilution chamber extracts a portion of flue gases at high temperature by means of a heated nozzle disposed at 90° against flue gas direction. Flue gas are conveyed to the dilution chamber, made from glass, where the sample is perfectly mixed with a pretreated air at a ratio defined by the operator in a range between 1 to 10. At the bottom of the dilution chamber, after the mixture took place, a 47mm plane filter is positioned to capture the particles contained in the gas mixture after dilution. The entire system is composed of five parts: a compressor of ambient air; a control unit, containing the pump and connecting the other 4 devices, a flow regulation unit to collect the exhaust gases from the chimney with the desired dilution ratio, a dilution cabin at a controlled temperature around 35-40°C, containing the dilution chamber, the thermocouple measuring the diluted gases temperature, the dilution air heater and a laptop connected via Bluetooth, with the control software. The control unit coupled with the flow regulation unit allows the regulation of the flows: the exhaust gases sampling rate and the dilution ratio. The dilution cabinet contains the

dilution chamber where the flue gas coming from the chimney is mixed with dried, filtered, pre-heated air, coming from the compressor. The dilution air gets into the dilution chamber through a mixing cone, to increase turbulence and improve the mixing of the two flows. The temperature inside the dilution chamber is monitored by a thermocouple and it is adjustable by means of an air heater. The whole system is maintained at a constant temperature around 35-40°C during the test. A plane filter, with standard dimensions of 47mm, was integrated in the configuration assuring a simple handling during the filter positioning and removing. All the components are controlled by means of a dedicated software and all the parameters are recorded. DT method is described in NS 3058, US EPA 5G standard methods. It is based on the dilution of the full flue gas using ambient air in a dilution tunnel (DT) combined to a filter maintained at 30°C. Dilution of the flue gas with lower temperature air allows condensation phenomena to occur, and the condensable fraction to condensate on the particles prior to sampling on the filter. The sample collected at the outlet of the dilution tunnel includes the solid and condensable phases of PM. SPC-IPA method was implemented by Ineris only. It consists in combining sampling of the solid fraction (SP) on a heated filter (160°C) to the collection of the condensable fraction using two washing bottles placed in series in a cooling unit in order to hold a sampling temperature below 20°C and filled in with 150ml of IPA to collect the condensable particles. The sampling temperatures and the dilution ratios of DT method and DC method are presented in Table 3. The DC method presented a global average mixing temperature of the gases and a global average dilution ratio more homogeneous compared to the DT method with 31°C to 38°C and 7.9 to 8.5 for the temperature and the dilution ratio for RPS and RWS cases, respectively.

2.6. Filters weighing and conditioning procedures. Before and after sampling, filters from DC, DT and SPC-IPA methods were conditioned prior and after sampling and weighed using laboratory weighing scales. The filters are pre-dried in an oven at 180°C for at least 1 hour; then cooled down in a desiccator for at least 4 - 8 hours and weighted at a 0.01 mg scale. After the sampling, the filters are stored directly in a desiccator for 24 hours at least. Finally, they are cooled down in a desiccator for 4 to 24 hours according to the partner. For samples taken by Ineris using the SPC-IPA method, isopropanol solution contained in impingers was evaporated to dryness at ambient temperature. after sampling. The matter remaining after evaporation was weighed using a weighing scale, with a resolution of 0.01 mg. The results obtained was added to filter weighing result. The analytical LoQ used for the characterization of solid and condensable fractions of PM based on the DC and DT methods corresponds to the LoQ of the weighing scale of Ineris for the 47mm filters i.e. 1.0 mg. The analytical LoQ used for the

characterization of solid and condensable fractions of PM based on SPC-IPA methods (3.0 mg) correspond to the sum of analytical LoQ of the weighing scale of Ineris and the analytical LoQ of dry extract from the SPC-IPA washing bottles (2.0 mg). The emission factors are expressed in milligrams by kilogram of dry wood burnt.

2.7. Tests of DC performance through CFD simulations. The dilution chamber has been set up through CFD simulation and experimental tests. In particular, the ANSYS ® Fluent 15.0 commercial code has been used to implement a thermo-fluid-dynamic model of the dilution chamber in order to estimate its characteristics and its performances. It consists of a steady state calculation of the flow field inside the chamber using, as boundary conditions, the inlet flow rates and temperatures of the exhaust gases and the dilution air. An unstructured tetrahedral grid and a conventional k-epsilon model of turbulence have been adopted within a Eulerian-Lagrangian approach, where Lagrangian particles have been injected in the chamber from the flue gas inlet to simulate the presence of particulate matter in this gas stream. A thin layer of porous zone just before the outlet of the dilution chamber has been set in order to simulate the head loss due to the filtering membrane used for the PM sampling.

2.8. SVOCs condensation compared to DT method. To verify the effective condensation of SVOCs in a series of tests, PM were sampled simultaneously on 15 hot filters (corresponding to 2 pellet stove tests and 3 wood stove tests), in the dilution tunnel and with the dilution chamber. These samples have been analyzed according to the norms ISO11338-1:2003 M and B and ISO11338-2/2003 to quantify the percentage of 19 PAHs and related to the total mass of PM.

2.9. Comparison of the DC method with DT and SPC-IPA methods. The correlation coefficients (r) were calculated with the StatView software between the three PM measurements methods then with the OGCs measurements for RPS and RWS to examine the response of these methods to condensable fraction. In order to explain the significant relationship between the DC method and the DT method, a linear regression model was used in Microsoft Excel®. For each linear correlation coefficient R^2 , the p . value was associated (Fisher test, $\alpha = 0.05$). To improve the linear model fit, 95% confidence intervals were plotted in order to exclude the outliers. The equations of linear regressions and the R^2 are given without outliers (outside of the 95% confidence intervals). The raw data of PM measurements based on DC, DT and SPC-IPA are given in the supplements (Table S1).

2.10. Determination of the relative abundance of PAH species. For each PAH specie, the relative abundance has been estimated according to equation 1, this normalization provides the actual concentration of each PAH in the PM independently from the total amount of PM collected

$$\%PAH = \frac{PAHs}{PM_{tot}} * 100 \quad \text{Eq. 1}$$

A comparison between the relative concentration of each PAH specie in the heated filter samples and the corresponding one in the dilution chamber and dilution tunnel samples have been carried out. The percentage increase of the PAH concentration compared to the heated filter reference has been calculated according to equation 2.

$$CE\% = \frac{(\%PAHx - \%PAH_{hf})}{\%PAH_{hf}} * 100 \quad \text{Eq. 2}$$

Where CE% is the condensation extent in percentage, %PAHx is the percentage of PAH species collected in the dilution tunnel or in the dilution chamber, %PAH_{hf} is the percentage of PAH species collected in the dilution tunnel or in the dilution chamber, %PAH_{hf} is the percentage of the same species collected on the heated filter. This condensation extent indicates the amount of each species condensed after dilution. All the species investigated have been categorized by their molecular weight and reported in Figure 3.

2.11. Determination of PM emission factors using the DC method. Emission factors are expressed as mass of pollutant divided by a unit energy, volume or weight of the activity emitting the pollutant (e. g.: grams of particulate emitted per kilogram of wood burnt). In order to compare the mass of the particles measured between the DC and other methods in the literature, the EF_{PM} (of solid and condensable particles) were calculated. In this present work, the EF_{PM} were calculated using the following equation 3 ²⁷:

$$EF_{PM} = \frac{[PM]_{(DC)} \times DF \times WSEFI \times CD}{Mass\ of\ wood\ burned} \quad \text{Eq. 3}$$

With:

EF_{PM} in mg kg⁻¹ (dry mass basis).

[PM]_(DC) in mg m⁻³ STP dry corrected per 13% O₂.

DF, the dilution factor at the corresponding sampling point for the DC and the DT methods. For the SPC-IPA method, the DF = 1.

WSEFL, the wood stove emission flows in $\text{m}^3 \text{h}^{-1}$ STP (for dry gas) have been approximated from the equation p. 85 of the EN16510-1:2016(F) norm described in the supplements (Equation S1).

CD, the combustion duration (in hours).

Mass of wood burnt in kg corrected for moisture (Table 1).

The EF_{PM} in mg kg^{-1} dry wood have been recalculated to g GJ^{-1} following the conversion factors assumed: 18 MJ kg^{-1} for wood logs and 19 MJ kg^{-1} for pellet logs.

3. RESULTS AND DISCUSSION

3.1. Development of the dilution chamber through CFD simulations. The simulated effect of dilution and mixing of the exhaust gases, represented by the presence CO_2 , is shown in Figure 2 for the final geometry of the dilution chamber. The results obtained show that the final geometry of the dilution chamber allows a good mixing of the two gas streams and allows the particles to stay within it for a time long enough (2-3 s) to produce the condensation of a large part of the SVOCs present.

3.2. Percentage PAH related to the total mass of PM. The species with molecular weight greater than 250 Da (Daltons) are already in solid phase at high temperature and no further condensation occurs after dilution. Species with molecular weight below 250 Da exhibit a strong increase of mass correlated to an effective condensation; this effect is greater with species having a lower molecular weight. This data show that the dilution chamber produces a condensation effect of semi volatile species comparable or even greater than the dilution tunnel. In the RWS tests, this effect may be attributed to the lower temperature reached in the DC ($\sim 30^\circ\text{C}$) compared to the DT ($\sim 60^\circ\text{C}$) at the same dilution ratio and even with high temperature of the flue gases ($>400^\circ\text{C}$).

3.3. General trends and study of correlations. Figure 4 reveals the results based on the three sampling methods DT, DC and SPC-IPA tested by the five European institutes (ISSI/ENEA, DTI, RISE and Ineris) obtained from the tests performed under nominal output or low output conditions with RPS (Figure 4, A) and RWS (Figure 4, B). The PM measurement results within the same sampling platform show some significative differences between the three methods used (Figure 4). In the literature, from about the same experimental design, Boman et al.²⁵ showed that the dilution ratio had an impact on the concentration of organics observed after dilution. In our study, the dilution ratio of DT method was higher than the dilution ratio of DC method (Table 3), this could explain the differences observed since the organics contribute to the concentration of PM. The OGCs

measurements (secondary y axis) are given in order to highlight the contribution of the condensable fraction according to the test configuration (low output or nominal output conditions and RPS or RWS). In general, the PM and OGCs measurement results obtained by the five partners are higher for tests performed on the RWS (between 39 and 567 mg m⁻³ for PM and between 129 and 1180 mg m⁻³ for OGCs) compared to tests performed on RPS (between 24 and 257 mg m⁻³ for PM and between 2 and 187 mg m⁻³ for OGCs) whatever the sampling method or the stove model (Table 2). Indeed, the advanced pellet stoves with a modern technology (automatic load, secondary air supply, combustion control, etc.) generate a range of particulate emissions lower than manual wood combustion devices²⁰ It is therefore necessary to develop sampling methods with LoQ allowing the measurement of both the solid and condensable fraction of particles at lower concentrations. In our study, according to the sampling method used to characterise PM, specific ranges have been obtained for the advanced wood log/pellet stoves of this present study (SPC-IPA: 31 - 516 mg m⁻³; DT :37 - 567 mg m⁻³; DC: 24 - 501 mg m⁻³). The PM emissions of RPS exhibit a lower range than RWS (24 to 220 mg m⁻³ versus 40 to 567 mg m⁻³). Excessive PM emissions are found during low output conditions of combustion. This observation agrees with results from the literature¹⁴. For the RPS tests, the ranges of PM measurements differ according to the sampling method (SPC-IPA: 31 mg m⁻³; DT: 37 to 220 mg m⁻³; DC: 24 to 129 mg m⁻³). In the same way, for the RWS tests, DT, DC and SPC-IPA ranged between 75 to 567 mg m⁻³, 39 to 501 mg m⁻³ and 126 to 516 mg m⁻³, respectively. In particular, the values are higher under low output conditions when the concentrations of condensable particles are more important. Moreover, Figure 4 (B) shows OGCs emissions higher for the RWS.

These emission levels suggest a contribution of condensable particles more important in the emissions from RWS compared to RPS emissions as described in the literature^{28–30}. Regarding the tests performed with the RPS at Ineris, DTI and RISE (Table 2 and Figure 4, A), the PM measurements based on SPC-IPA, DT (except Ineris due to the dilution ratio high, Table 3) and DC under nominal output conditions are of the same order of magnitude (between 24 and 38 mg m⁻³) and the OGCs mean concentrations remain lower than 35 mg m⁻³. For the PM measurements based on DC method performed by RISE on the RWS (Table 2), the results remain approximately the same (about 40 mg m⁻³). Moreover, the PM measurement based on DT method (about 90 mg m⁻³ on average), on DC method (about 44 mg m⁻³ on average) and the OGCs measurement results (129 – 255 mg m⁻³) obtained following RISE tests are lower than those obtained by the other partners.

Regarding RWS (Figure 4, B and Table 4), when the emissions of OGC are high, DC shows good agreement with DT and SPC-IPA. In general, for the particulate emissions from combustion tests performed with RWS, the PM measurements based on DC are close to the PM measurements based on DT method ($r = 0.92$ for $n = 42$). This result seems to confirm that a significant part of condensable particles is collected by the DC method. Emissions of OGCs from RPS are lower than RWS, at those levels of OGCs concentrations, DC shows good agreement with DT and SPC-IPA, apart from results from DTI and Ineris which presented average filter weights close to the analytical LoQ 0.98mg (Table 4 and Table S2). In the case of Ineris, it is due to an unexpected high dilution ratio during the trials (Table 3). With DC method the dilution ratio is defined and set by computer in a range between 1 to 10, which is not the case with the DT. Results of dry extract in the SPC-IPA washing bottles representing the condensable fraction are below analytical LoQ (3.0mg) suggesting the absence of condensable in the emissions from RPS.

3.4. Model of analytical adjustment. The PM measurements based on DC method were compared to PM measurements based on the DT method with a model of linear regression. Results obtained by applying the model of linear regression (including the IC 95% and the standard error) from the DC compared to DT are plotted on Figure 4. The equations and R^2 of the linear regressions are given without the outliers (data out of the IC 95% and symbolised by blank signs on the Figure 5), according to the combustion conditions and the fuel types. The model of linear regression confirms a significant positive relationship between the PM measurements based on the DC method compared to the DT method (R^2 ranged between 0.81 and 0.99 with $p < 0.05$) except for the RPS tests at Ineris ($n = 9$, $R^2 = 0.02$ with $p = 0.75$, $slope = 0.10$) and DTI ($n = 10$, $R^2 = 0.68$ with $p = 0.10$, $slope = -0.42$). The linear regressions between DC and SPC-IPA methods for the RPS and RWS tests were also studied (linear regressions no plotted) and also showed a significantly positive relationship ($n = 9$, $R^2 = 0.95$, $slope = 0.92$ and $n = 6$, $R^2 = 0.97$, $slope = 1.03$ with $p < 0.05$ for RPS and RWS, respectively). Indeed, in the case of low PM emissions ($< 50 \text{ mg m}^{-3}$) with the pellet stoves at DTI and Ineris, the DT and DC methods are weakly correlated (Figure 5) whereas DC and SPC-IPA are strongly correlated. This result is partly due to the fact that at those levels, in the conditions of tests applied at Ineris and samples collected by the DT methods exhibited weights close to the weighing method LoQ which affects the quality of results.

3.5. EFs using DC method. The results of EF_{PM} from the DC and the literature are illustrated in Table 5 and Figure 6. The RWS tested by RISE seems to emit less condensable particles which may explaining low OGCs levels, which might be anticipated as the stove tested is characterized by the lowest emissions of the Swedish market. Disregarding the initial assumptions and the initial conditions (the measurement uncertainties, the calculation uncertainties with the equation for the WSEFl, the coefficient conversion to pass from $g\ kg^{-1}$ to $g\ GJ^{-1}$, the different model of advanced stoves or the type of fuel), the results obtained show i) whatever the advanced stove model and the combustion conditions, the EF_{PM} from RPS were 2 – 20 orders of magnitude lower than RWS EF_{PM} (108 against $2420\ g\ GJ^{-1}$), ii) the EF_{PM} of RPS and the EF_{PM} from RWS are higher under low output conditions ($282\ g\ GJ^{-1}$ and $556\ g\ GJ^{-1}$ for the RPS tested by Ineris and RISE, respectively and $2420\ g\ GJ^{-1}$ for the RWS tested by Ineris) as shown in the literature 5, iii) the particulate emission from the combustion of wood with bark are higher (427 , 816 and $488\ g\ GJ^{-1}$ for the beech and birch combustion tested by Ineris, ISSI/ENEA and RISE, respectively), iv) except for the EF_{PM} obtained with the advanced wood log stove tested under low output conditions by Ineris, ($2420\ g\ GJ^{-1}$), the EF_{PM} calculated in this present study for wood log stoves ($108 - 816\ g\ GJ^{-1}$) are in good agreement with EF_{PM} that can be found in the literature for the same type of stoves ($31 - 820\ g\ GJ^{-1}$)^{13,17,31,32} and v) the EF_{PM} obtained by Ineris with the advanced wood log stove under low output condition ($2420\ g\ GJ^{-1}$) was of the same order of magnitude as the EF_{PM} from conventional/traditional wood stove based on $PM_{2,5}$ measurements in the study of Klimont et al.³³ ($3000\ g\ GJ^{-1}$). Using the averages of EF_{PM} results from DC method for all European partners and by type of stove, we find that the EF_{PM} from advanced pellet/wood log stoves determined with the DC method are of the same order of magnitude as those presented in the EMEP Guidebook and used in the emission inventories or the literature (Figure 6). However, the EF_{PM} measured with the DC method during the tests with low output conditions are higher (Table 5). This difference can be explained by the combustion conditions used during our tests (nominal or reduced), the stove technology, the type of fuel and/or the dilution methods used to sample PM. The last European guidebook on the air pollutant emission inventory³⁴ which includes the results of the EF_{PM} from the literature shows a strong heterogeneity. This heterogeneity directly depends on the measurement methods used to characterise the PM emissions (which included or not the condensable fraction), the type of combustion equipment considered (conventional, Advanced, Ecodesign, ...) and the operating conditions used. Indeed, the emission factor is largely impacted by the model of the appliance, the type of fuel or the combustion conditions. Depending on the combustion conditions, the PM

emission factors exhibit values from less than 20 g GJ⁻¹ under controlled nominal ideal combustion conditions up to more than 5000 g GJ⁻¹ under poor combustion conditions. Under poor combustion conditions, the mass of condensable can exceed the mass of solid particles³⁵.

4. CONCLUSION

The analysis and quantification of PAHs related to the total mass of PM of samples taken by ENEA/ISSI using the dilution chamber showed that it produces a condensation effect of semi volatile species comparable or even greater than what is obtained using the dilution tunnel. Comparison of the DC method performed by five European institutes (Ineris, ISSI/ENEA, DTI and RISE) from four EU countries (France, Italy, Denmark and Sweden, respectively), with DT and SPC-IPA methods exhibited a correlation (r) ranged between 0.67 and 0.99 (with DT) and between 0.74 and 0.98 (with SPC-IPA) respectively, Except for the pellet stove tested at Ineris and DTI, the PM measurements based on DC method show a linear correlation with PM measurements based on DT (R^2 ranged between 0.81 and 0.99, $p < 0.05$) specifically for the residential wood stoves used under low output conditions when the condensable fraction contributes the most. PM EFs were about 2 to 20-fold higher for the residential wood stoves (EF ranged between 201 and 2420 g GJ⁻¹) compared to those obtained for the residential pellet stoves (EF ranged between 108 and 556 g GJ⁻¹) which confirms trends described in the literature. The use of such a method associated with the application of testing procedure simulating real life combustion cycles across Europe in the future will contribute: i) to improve emission inventories allowing to close the gap between observed and modelled PM in ambient air, hence improving air quality; ii) to complete existing methods used to implement Ecodesign Directive (2005/32/EC) in this product area for which the main environmental characteristics of the products covered by the Lot 15 / Lot 20 are the emissions, among them PM emissions, and for which existing standards are not sufficiently adequate to quantify condensable. Finally, with new generations of more efficient stoves with lower particulate emissions, measurement methods must be adapted to achieve lower limits of quantification.

SUPPORTING INFORMATION

The supplements contain the calculation of $WSEFI$ parameter of the Equation 3, the raw data set of PM measurements based on DC, DT and SPC-IPA methods with OGCs measurements and the average filter weights based on DC, DT and SPC-IPA methods.

AKNOWLEDGMENT

This work has been supported by the European Metrology Program for Innovation and Research (EMPIR) in the project context IMPRESS 2 coordinated by NPL UK. The EMPIR initiative is co-funded by the European Union'S Horizon 2020 research and innovation program and the EMPIR Participating States. Financial support from H2020 program is gratefully acknowledged. We thank all participants of the IMPRESS 2 project: the coordinator (NPL) and the all the others National Metrology Institutes (CEM, CMI, PTB, RISE, VSL, VTT, CNR, DTI, DTU, ISSI, ENEA, Ineris, TU Delft and UC3M). Ineris thanks Nicolas Karoski, Adrien Dermigny, Mehdi Dionigi and Lars Schwarzer for their contribution to the combustion tests.

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Table 1: Start-up conditions of the combustion devices. Gas combustion temperature values are averaged (\pm se).

Residential pellet stoves (≤ 10 kW)						
European Institutes	Advanced residential stoves	Fuel type	Mass of wood burned (kg h ⁻¹)	Moisture (%)	Output conditions	Temperature of gas combustion (°C)
Ineris	Elance II 8	Pellets (A1)	1.4	6	Nominal & Low	122 (\pm 24)
ISSI/ENEA	Nordica	Pellets (A2)	3.6	6.3	Nominal	109 (\pm 4)
DTI	Heta Greenline	Pellets (A1)	3.0	6	Nominal	86 (\pm 1)
RISE	Divina plus	Pellets (A1)	3.0	6	Nominal & Low	127 (\pm 32)
Residential wood log stoves (≤ 10 kW)						
Ineris	Iwaki	Beech (with bark)	2.4	20 - 25	Nominal & Low	241 (\pm 45)
ISSI/ENEA	Piazzetta 920	Beech (with bark)	4.8	15	Nominal	454 (\pm 31)
DTI	MorsØ 1440	Birch (no bark)	4.0	14.5	Nominal	307 (\pm 24)
RISE	Contura 610	Birch (with/no bark)	2.8	19	Nominal	236 (\pm 15)
		Spruce (no bark)				

Table 2: Averages of emissions of O₂, CO₂ in %, CO, OGCs and PM in mg m⁻³ STP dry to 13% O₂ (where no other units) from residential stoves. Sampling measurement methods used to characterise PM emissions: SPC-IPA for heated filter and impinger filled in with IPA collection (Ineris only), DT for dilution tunnel and DC for dilution chamber.

Residential pellet stoves (≤ 10 kW)										
European Institutes	Fuel type	Output conditions	n	O ₂	CO ₂	CO	OGCs	PM (mg m ⁻³)		
				(%)	(%)	(mg m ⁻³)	(mg m ⁻³)	SPC-IPA	DT	DC

Ineris	Pellets (A1)	Nominal	5	13	8	116	2	31	105	29
		Low	4	15	6	159	4	31	257	31
ISSI/ENEA	Pellets (A2)	Nominal	10	11	9	1364	21	-	167	129
RISE	Pellets (A1)	Nominal	6	17	4	601	34	-	37	33
		Low	7	19	2	2587	187	-	220	118
DTI	Pellets (A1)	Nominal	6	13	8	227	9	-	34	24
Residential wood log stoves (≤ 10 kW)										
Ineris	Beech (with bark)	Nominal	3	12	8	3364	480	126	145	121
		Low	3	14	7	4983	1180	516	567	501
ISSI/ENEA	Beech (with bark)	Nominal	12	10	11	8423	873	-	355	235
RISE	Birch (with bark)	Nominal	7	13	8	2008	224	-	102	46
	Birch (no bark)		7	11	10	1396	129	-	75	39
	Spruce (no bark)		4	13	8	2478	255	-	96	46
DTI	Birch (no bark)	Nominal	6	8	13	3628	672	-	214	229

Table 3: Sampling temperature of PM and dilution ratio (DR) of SPC-IPA, DC and DT methods used during the RPS and RWS tests.

Residential pellet stoves (≤ 10 kW)							
European Institutes	SPC-IPA		DT		DC		
	Sampling temperature		Sampling temperature		DR	Sampling temperature	DR
Ineris	Heated filter: 160°C		27.6 °C		27.4	31.0 °C	8.4
	Washing bottles: 20°C						
ISSI/ENEA	-		31.9 °C		8.7	31.1 °C	8.5
DTI	-		28.5 °C		14.3	30.9 °C	8.0

RISE	-	28.0 °C	10.1	32.1 °C	7.9
Residential wood log stoves (≤ 10 kW)					
Ineris	Heated filter: 160°C Washing bottles: 20°C	30.9 °C	33.5	33.0 °C	8.3
ISSI/ENEA	-	62.9 °C	7.9	38.2 °C	8.9
DTI	-	30.0 °C	12.3	33.1 °C	8.0
RISE	-	34.5 °C	22.4	32.8 °C	8.5

Table 4: Correlation (r) calculated from the comparison of the PM measurements based on heated filter and impinger filled in with IPA collection (SPC-IPA), dilution chamber (DC) and dilution tunnel (DT) and with OGCs measurements for the residential wood log stoves (RWS) and residential pellet stoves (RPS).

European Institutes		DC:SPC-IPA	DC:DT	DC:OGCs	DT:OGCs
	n	r			
Ineris					
RPS	9	0.74	0.13	0.37	0.50
RWS	6	0.98	0.96	0.98	0.98
ISSI/ENEA					
RPS	10	-	0.86	0.44	0.32
RWS	12	-	0.67	0.76	0.50
DTI					
RPS	6	-	- 0.73	0.45	-0.26
RWS	6	-	0.99	0.92	0.94
RISE					
RPS	13	-	0.92	0.98	0.93
RWS	18	-	0.82	0.84	0.79

All						
	RPS	38	-	0.61	0.56	0.52
	RWS	42	-	0.92	0.84	0.87

Table 5: Average of PM emission factors (g GJ⁻¹) obtained from the tests of wood combustion carried out by the Ineris, ISSI/ENEA, DTI and RISE institutes and based on PM measurement with the dilution chamber method (DC). The results of PM emission factors are compared with the literature and emission inventories.

Combustion appliance	Fuel	Fraction sampled	EF _{PM} (g GJ ⁻¹) ^b	References
Advanced pellet stoves (RPS)	Pellet		121 - 282 ^a	Ineris
			332	ISSI/ENEA
			108	DTI
		SPC	206 - 556 ^a	RISE
Advanced woodlog stoves (RWS)	Beech (with bark)		427 - 2420 ^a	Ineris
			816	ISSI/ENEA
	Birch (no bark)		201 - 587	RISE - DTI

	Birch (with bark)		488	RISE
	Spruce (no bark)		357	RISE
Advanced wood stove	Wood logs		120 - 176	
		PM _{tot}		31
Pellet stove	Pellet		75 - 139	
Wood stove	Birch, pine and spruce logs	PM _{tot}	38 - 350 ^a	13
Advanced stoves	Pellet	TSP	31 - 124	17
Modern wood stove	Wood logs	PM _{2.5}	53 - 820 ^a	32

^a represents the maximum value of EF_{PM} measured during poor combustion conditions.

Figures captions

Figure 1: The measurement set-up

Figure 2: Dilution chamber (DC) CFD simulation (CO_2 mixing). The simulated effect of dilution and mixing of the exhaust gases, represented by the presence CO_2 for the final geometry of the DC.

Figure 3: Condensation Extent in a pellet (RPS) and wood stoves (RWS). Blank circles represent the condensation extent in the dilution chamber and the blank triangles represent the condensation extent in the dilution tunnel under nominal conditions.

Figure 4: Results of the PM characterisation, solid and condensable particles (SPC) in the emission from residential pellet stove (RPS: plot A) and residential wood log stove (RWS: plot B) based on three measurement methods: dilution chamber (DC, speckled bar), dilution tunnel (DT, full bar) heated filter + washing bottles filled in with isopropanol (SPC-IPA, striped bar) during the tests fulfilled by four European institutes: Ineris, ISSI/ENEA, DTI and RISE as part of Impress 2 project. The OGCs are represented on the secondary axis (full line). The results are expressed in mg m^{-3} STP, dry gas and 13% O_2 . Only Ineris has performed the SPC-IPA method.

Figure 5: Regression lines (full line) between the PM solid and condensable particles (SPC) measured with the dilution chamber (DC) and the dilution tunnel (DT), expressed in mg m^{-3} STP, dry gas and 13% O_2 . The PM sampling was carried out at the exit of emission from the residential pellet stoves (RPS) and the residential wood log stoves (RWS) during the tests fulfilled by five European institutes: Ineris (A, B), ISSI/ENEA (C, D), DTI (E,F) and RISE (G, H) as part of Impress 2 project. Standard error (se, dotted line) and 95% confidence interval (non-continuous line) of linear regression are plotted and the equation of linear regression and R^2 are given (without the points out of the IC 95%, blank signs) for the six plots. The results of SPC measured are plotted like nominal conditions (full circle), low output conditions (full triangle), wood with bark (full square), wood without bark (blank diamond) and spruce (cross).

Figure 6: Comparisons between PM emission factors from residential pellet and wood log stoves calculated with dilution chamber method (DC) and emission factors of PM used in emission inventories or literature. Blue histograms are the average of EF_{PM} values. The black lines are the min and max EF_{PM} values obtained during the nominal and low combustion conditions, respectively.

Figure 2.

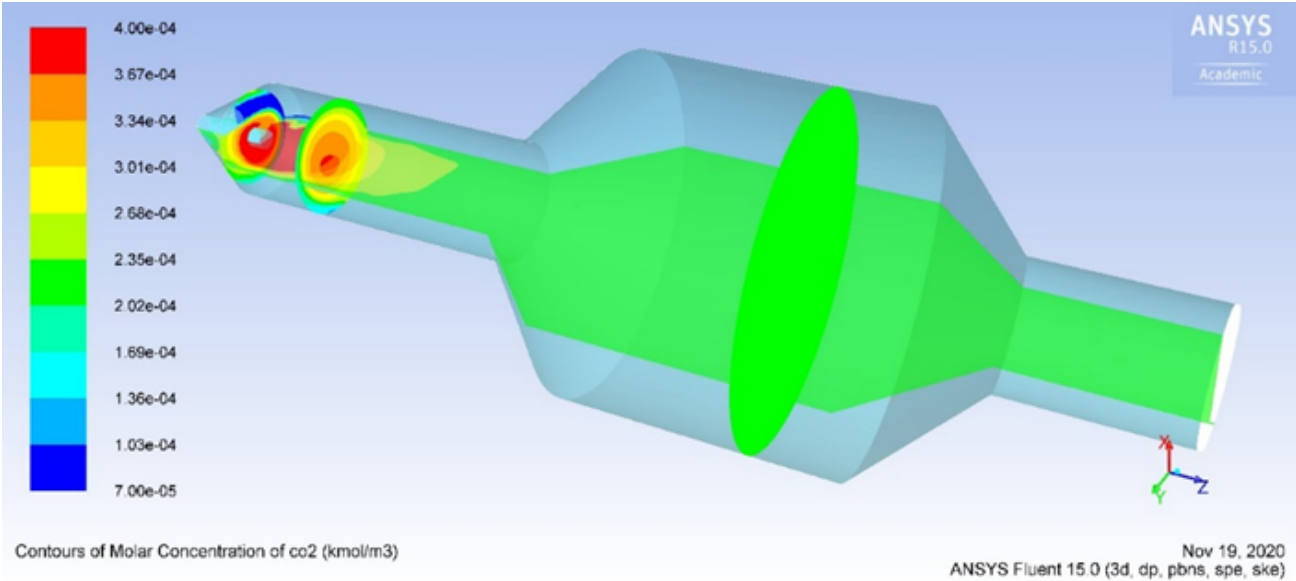


Figure 3.

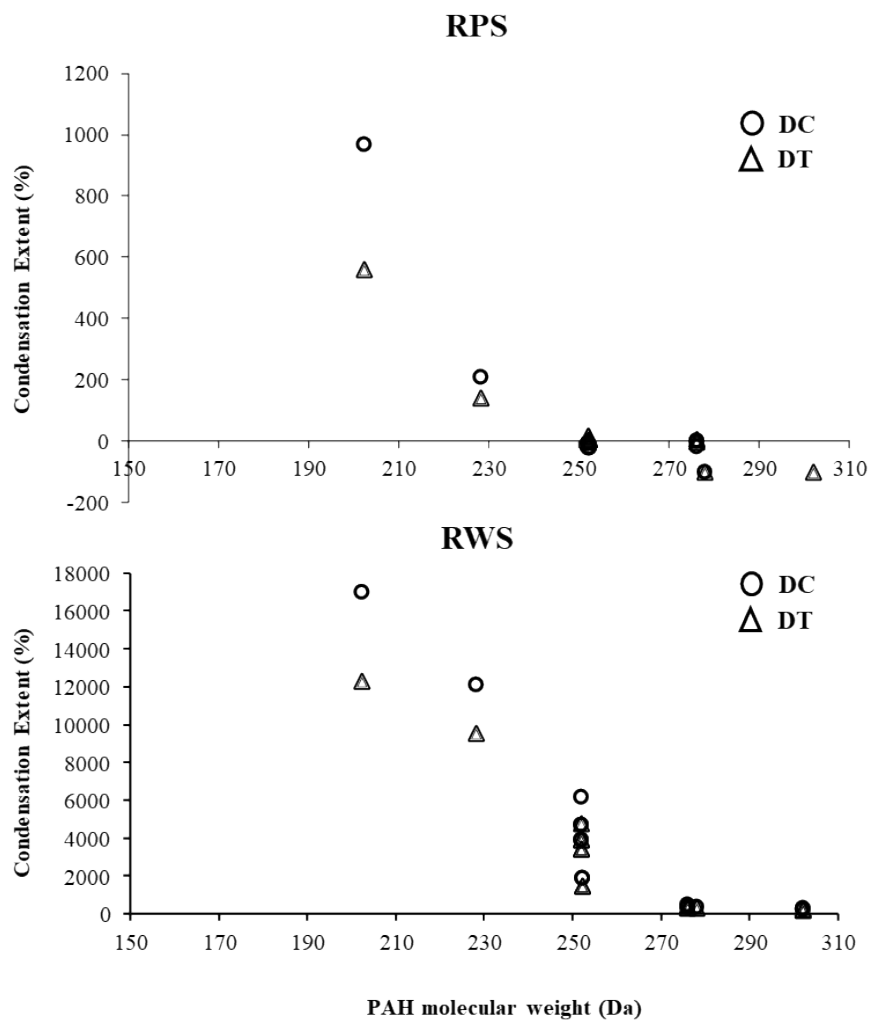


Figure 4.

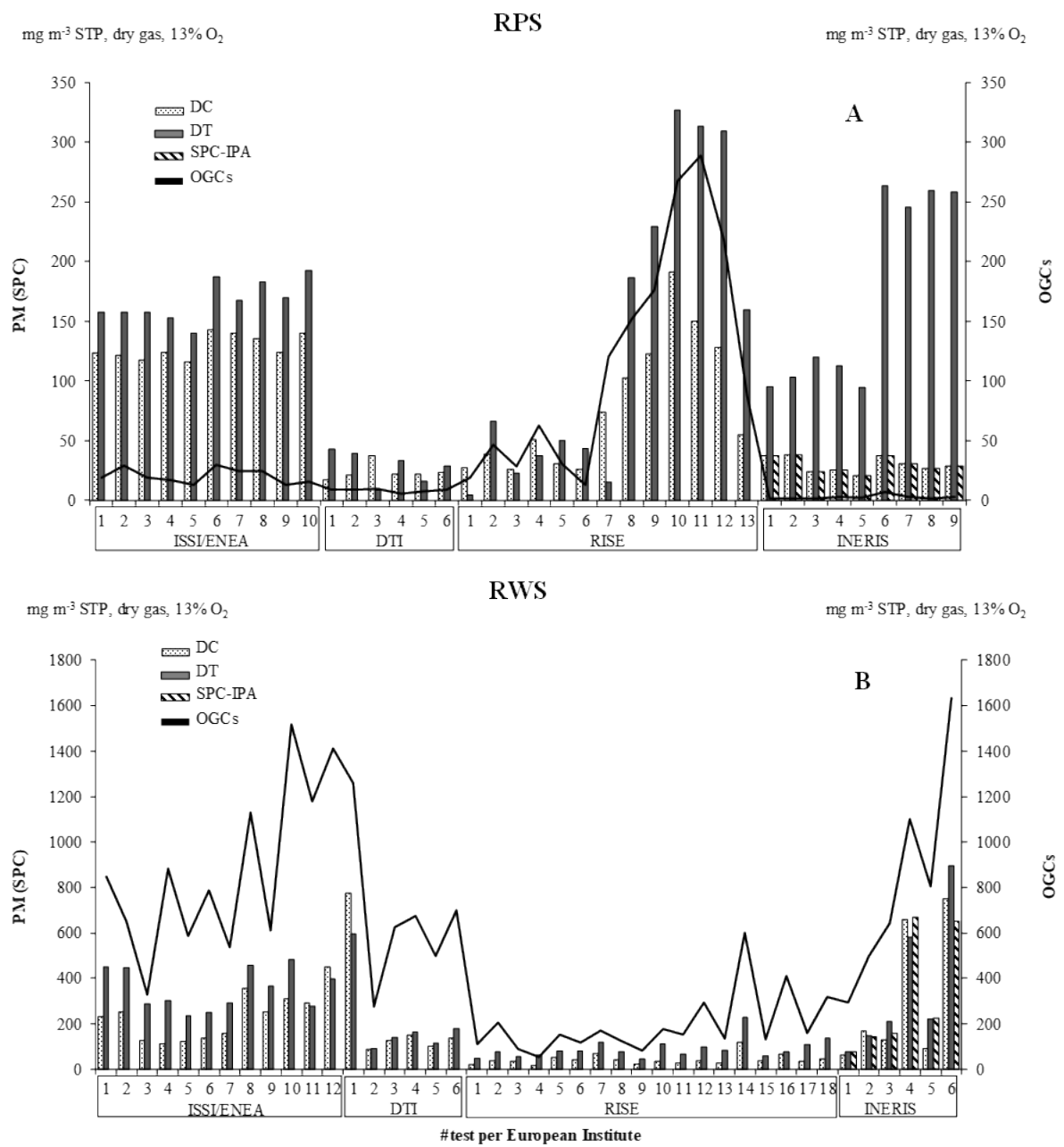


Figure 5.

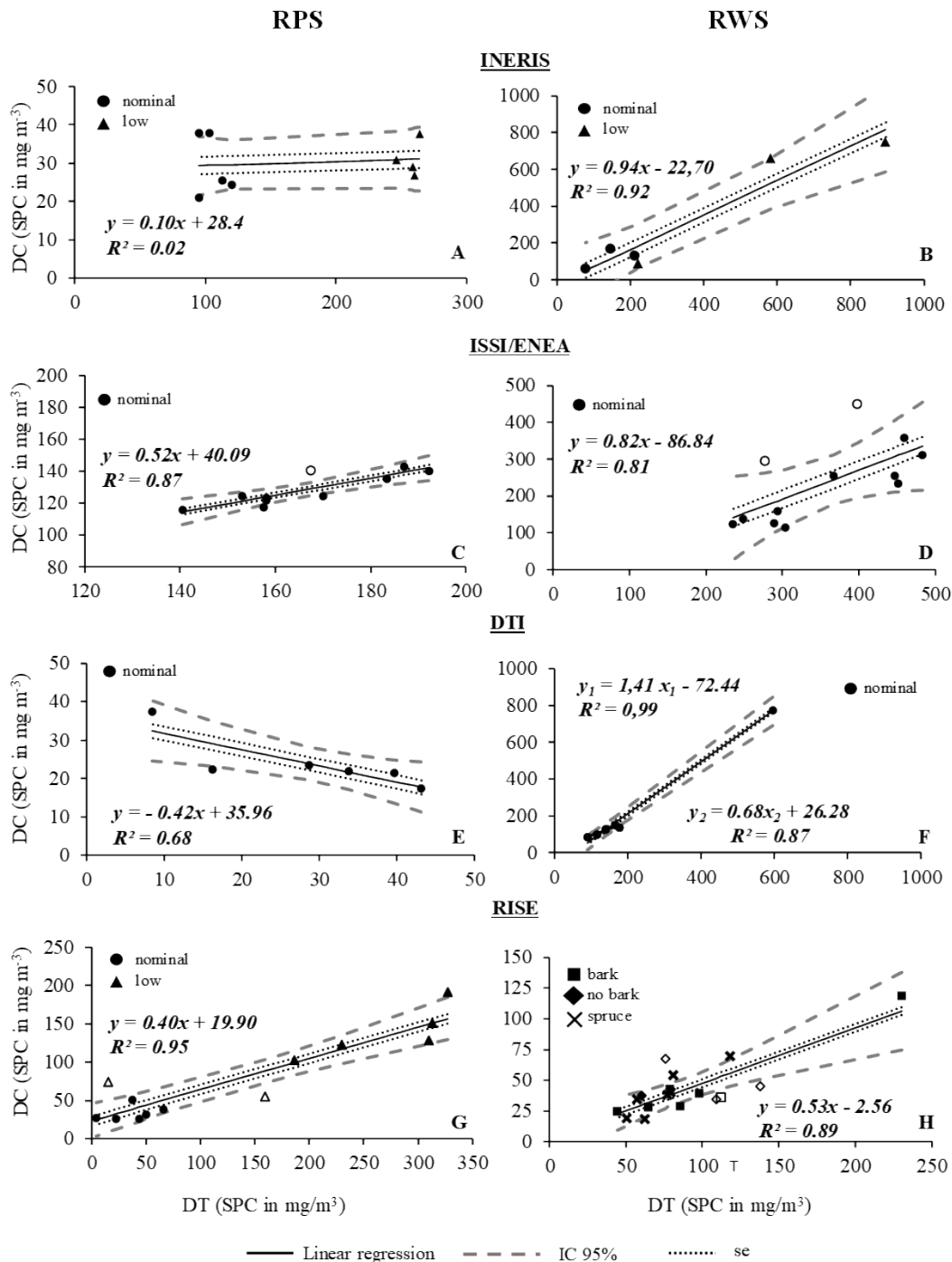


Figure 6.

