

**NPL REPORT AS 94**

# **EURAMET comparison 1282 Comparison of condensation particle counters**

# **FINAL REPORT**

**Paul Quincey Dimitris Sarantaridis Thomas Tuch Jaakko Yli-Ojanperä Richard Högström Felix Lüönd Andreas Nowak Anke Jordan-Gerkens Francesco Riccobono Kenjiro Iida Hiromu Sakurai Miles Owen**

# NOT RESTRICTED

NOVEMBER 2014

National Measurement System . . .

# EURAMET 1282: Comparison of condensation particle counters

Paul Quincey<sup>1</sup>, Dimitris Sarantaridis<sup>1</sup>, Thomas Tuch<sup>2</sup>, Jaakko Yli-Ojanperä<sup>3</sup>, Richard Högström<sup>4</sup>, Felix Lüönd<sup>5</sup>, Andreas Nowak<sup>6</sup>, Anke Jordan-Gerkens<sup>6</sup>, Arne Kuntze<sup>6</sup>, Francesco Riccobono<sup>7</sup>, Kenjiro lida<sup>8</sup>, Hiromu Sakurai<sup>8</sup>, and Miles Owen<sup>9</sup>

<sup>1</sup> Analytical Science Division, National Physical Laboratory, Hampton Road, Teddington, TW11 0LW, UK.

<sup>2</sup>Leibniz Institute für Troposphärenforschung (TROPOS), Permoserstraße 15, D-04318 Leipzig, Germany

 ${}^{3}$ Tampere University of Technology (TUT), Department of Physics, Aerosol Physics Laboratory, Korkeakoulunkatu 3, FI-33101 Tampere, Finland

4 MIKES – Centre for Metrology and Accreditation, Tekniikantie 1, FI-02151 Espoo, Finland

5 Federal Institute of Metrology (METAS), Lindenweg 50, CH-3003 Bern-Wabern, **Switzerland** 

6 Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, D-38116 Braunschweig, Germany

7 European Commission, Joint Research Centre (JRC), Via E. Fermi 2749, I-21027 Ispra (VA), Italy

<sup>8</sup>National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Umezono, Tsukuba, Ibaraki, Japan

<sup>9</sup>US Army Primary Standards Laboratory (APSL), Bldg 5435 Fowler Rd, Redstone Arsenal, AL 35898, United States

© Queen's Printer and Controller of HMSO 2014

ISSN 1754-2928

National Physical Laboratory Hampton Road, Teddington, Middlesex, TW11 0LW

Extracts from this report may be reproduced provided the source is acknowledged and the extract is not taken out of context.

> Approved on behalf of NPLML by Dr Michael Adeogun, Head of Analytical Science Division.

# EURAMET 1282: Comparison of condensation particle counters

### **EXECUTIVE SUMMARY**

Aerosol particle number concentration has recently featured in vehicle emission legislation and is becoming increasingly important in other areas such as ambient air monitoring. Number concentration measurements are also often integral to particle size distribution measurements, such as when using a Mobility Particle Size Spectrometer.

The draft ISO standard ISO/DIS 27891 [1] describes a calibration procedure for Condensation Particle Counters (CPCs - the usual type of instrument for measuring particle number concentration in the size range from a few nanometers to a few micrometers) either by reference to an aerosol electrometer, or to a reference CPC. The DIS refers to the role of NMIs in providing certification for both reference aerosol electrometers and reference CPCs.

The aim of this comparison was to compare the results of different laboratories' measurements of particle number concentration using CPCs (in  $cm^{-3}$ ).

The comparison took place at the Leibniz Institute for Tropospheric Research (TROPOS) in October 2013 as part of the EMRP project ENV02 PartEmission (Automotive combustion particle metrics), Deliverable 1.2.2.

Because this is the first multi-NMI comparison of CPCs, EURAMET participants were joined by other participants with strong metrological expertise in this area.

The comparison included aerosol particle concentrations between about 100 and 20,000  $\text{cm}^3$ , and aerosol particle sizes from 13 to 100 nm, using aerosol particles composed of unsintered silver, sintered silver and soot. The results show discrepancies between instruments with a relatively high (23 nm) 50% cut-off size, even at aerosol particle sizes well above the cut-off size. Apart from this, the results showed that for the full concentration range, and sizes between 23 and 100 nm, agreement to ±10% between reference laboratories is currently achieved.

NPL Report AS 94

### **TABLE OF CONTENTS**

# **EXECUTIVE SUMMARY**

### **TABLE OF CONTENTS**



**APPENDIX A2 – PARTICIPANTS RESULTS PROFORMAS 16**

NPL Report AS 94

### **1. INTRODUCTION**

Aerosol particle number concentration has recently featured in vehicle emission legislation and is becoming increasingly important in other areas such as ambient air monitoring. Number concentration measurements are also often integral to particle size distribution measurements, such as when using a Mobility Particle Size Spectrometer.

The draft ISO standard ISO/DIS 27891 [1] describes a calibration procedure for Condensation Particle Counters (CPCs - the usual type of instrument for measuring particle number concentration in the size range from a few nanometers to a few micrometers) either by reference to an aerosol electrometer, or to a reference CPC. The DIS refers to the role of NMIs in providing certification for both reference aerosol electrometers and reference CPCs.

Although not strictly a chemical measurement, the comparison belongs in the Gas subcommittee of TC-MC because of the similarity to gas concentration measurements, following the precedent of earlier projects 893 (workshops to establish "Metrology infrastructure for airborne nanoparticles") and 1027 ("Comparison of combustion particle number concentration and size").

The aim of this comparison was to compare the results of different laboratories' measurements of particle number concentration using CPCs (in cm<sup>-3</sup>).

The comparison took place as part of the EMRP project ENV02 PartEmission (Automotive combustion particle metrics), Deliverable 1.2.2.

Because this is one of the first multi-NMI comparisons of CPCs, EURAMET participants were joined by other participants with strong metrological expertise in this area.

### **2. OPERATION OF THE COMPARISON**

### **2.1. PARTICIPANTS**

The 8 participating laboratories in the EURAMET comparison were:

- NPL United Kingdom (co-ordinating laboratory)
- TROPOS (Leipzig Institute for Tropospheric Research) Germany, the hosts
- Tampere University of Technology (TUT), in collaboration with MIKES Finland
- METAS Switzerland
- PTB Germany
- JRC-IET EU
- AIST Japan
- APSL (US Army Primary Standards Laboratory) USA

### **2.2. PARALLEL EXERCISES**

Three distinct exercises were carried out during the week.

(i) A comparison of the participants' ability to calibrate their CPCs in the "plateau" (size- and particle composition-independent) region of their operation, using a range of particle size, material and concentrations.

- (ii) An investigation of the detection efficiencies of the participants' CPCs at particle sizes below the plateau region, using a range of particle size, material and concentrations. This is known to depend on the particle size and composition in subtle ways that make comparisons between different laboratories and particle sources difficult. In this context, several models of CPC with different size characteristics were used, while one participant (PTB) brought two different models of CPC (see Table 1 below).
- (iii) As part of a separate deliverable within EMRP project ENV02, non-metrological organisations also took measurements, to demonstrate the suitability of the procedures for wider dissemination of traceability for these measurements.

Only the first of these is reported in detail in this report, though some other data are also included, for convenience.

### **2.3. PROCEDURE**

The comparison was held at the Leipzig Institute for Tropospheric Research (TROPOS) in Germany during the week 14-18 October 2013.

Because transportable measurement standards for aerosols are not easily available, participants brought their CPCs and any associated equipment to TROPOS. The CPCs were connected to a common aerosol source using pipework designed to minimise differences between the ports, taking into account different diffusive losses due to the different flow rates of the CPCs by adjusting the length of the conductive tubing to each CPC, according to theoretical calculations. A length of 70 cm was used for the 1 l/min instruments, and aerocalc software used to determine equivalent loss lengths for the other instruments.

Two types of airborne particle generator were used for the comparison, with three distinct types of particle being produced. A ceramic furnace condensation-type aerosol generator was used to produce Ag particles. These were usually sintered to produce more spherical particles, but on some runs unsintered particles were used, to assess the effect of particle morphology. The second generator was a miniCAST (Series 5200) generating soot particles.

All measuring equipment was operated by people from the relevant participant laboratories, with the exception of the APSL equipment, which was operated on their behalf by TROPOS.



Several different commercial designs of butanol CPC were used, as set out in Table 1.

Table 1: Participants' CPCs

Particle sizes were selected within the nominal range 6 to 100 nm, and the concentration range was between around 100 and 20 000 particles  $cm^{-3}$ .

Aerosol particle size was characterised by a Mobility Particle Size Spectrometer (MPSS). Accurate assignment of size to aerosol particle distributions is a complex topic in itself, and was not a central aspect of the comparison. The comparison was primarily concerned with the ability of the participants to measure the particle number concentration of the aerosol, while the different sizes were used to indicate limitations of the CPCs and the experimental design.

Further details of the procedure followed are given in the Protocol (Appendix 1), and of the equipment and methods used by the participants in the Results Proformas (Appendix 2).

### **2.4. COMPARISON RUNS**

There were 52 designated runs, described in Table 2 below.

In summary:

Runs 1 - 22 were of sintered Ag particles, of sizes from 6 to 60 nm and concentrations between 100 and 20,000  $cm^{-3}$ .

Runs 23 – 32 were of unsintered Ag particles, of sizes 23 or 41 nm and concentrations between 100 and 20,000  $cm^{-3}$ .

Runs 33 – 52 were of soot particles, of sizes from 23 to 100 nm and concentrations between 100 and  $20,000$  cm<sup>-3</sup>.

### **2.5. REPORTING OF RESULTS**

As described in the Protocol (Appendix 1), final results were sent via email using the agreed Proformas (Appendix 2), to allow for recalibration of equipment after its return to the home laboratory.

Participants decided whether particle size was within the plateau region of their CPC, and estimated their own measurement uncertainties independently, with rationales explained on the Proformas.



Table 2: Description of each run.

### **3. RESULTS**

### **3.1. REPORTED RESULTS**

The full set of reported results for the comparison is given in Table 3.



Table 3: Reported results.

The numbers given a yellow background are those that were designated by the participants as being away from the plateau of their CPC. These data are not considered further in this report.

It is notable that while TROPOS and PTB consider that their TSI 3772 instruments are reporting valid results for the 13 nm particles (Runs 16 – 20), AIST and APSL do not.

As explained in the Results Proforma in Appendix 2, the METAS instrument required repair at the start of the comparison, and was only able to participate from the third day (run 33). The AIST data for runs 48 to 52 were inadvertently not recorded.

### **3.2. PRELIMINARY ASSESSMENT**

A preliminary assessment of the results showed that while there was generally good agreement, the two TSI 3790 CPCs (JRC and PTB 2) gave significant lower results than the other instruments, even at particle sizes considered to be in their plateau region. These results are presented in the charts below, but their results are not taken into account when calculating the comparison reference values.

The data from the three laboratories reporting for the 13 nm particles (Runs  $16 - 20$ ) - NPL, TROPOS and PTB 1 – also showed significantly more variation, which can be attributed to the size being close to the edge of the plateau region, especially for the TSI 3772 instruments used by TROPOS and PTB.

### **3.3. COMPARISON REFERENCE VALUE**

Independent measurements of particle number concentrations, using an aerosol electrometer, were supplied by TROPOS. However, high accuracy traceable values of particle number concentration also require detailed knowledge of the presence of multiply-charged particles, which will be increasingly significant at the higher particle sizes used, and experimental measurements of these were not available.

The comparison reference value is taken simply to be the mean of the results reported as being "on plateau", with the exception of the cases mentioned in Section 3.2.

### **3.4. GRAPHICAL PRESENTATION OF RESULTS**

Selected results are presented graphically in 9 Figures:

Figure 1: 60 nm sintered Ag at a range of concentrations (Runs  $1 - 5$ ) Figure 2: 26 nm sintered Ag at a range of concentrations (Runs 11 - 15) Figure 3: 41 nm unsintered Ag at a range of concentrations (Runs 23 – 27) Figure 4: 23 nm unsintered Ag at a range of concentrations (Runs 28 – 32) Figure 5: 100 nm soot at a range of concentrations (Runs 33 – 37) Figure 6: 41 nm soot at a range of concentrations (Runs 43 – 47) Figure 7: 23 nm soot at a range of concentrations (Runs 48 - 52) Figure 8: 20,000 cm<sup>-3</sup> sintered Ag at a range of sizes (Runs 1, 6, 11, 16) Figure 9: 20,000  $\text{cm}^3$  soot at a range of sizes (Runs 33, 38, 43, 48)

In all cases the y-axis shows percentage difference from the comparison reference value.



Figure 1: 60 nm sintered Ag at a range of concentrations (Runs  $1 - 5$ )



Figure 2: 26 nm sintered Ag at a range of concentrations (Runs 11 ‐ 15)







Figure 4: 23 nm unsintered Ag at a range of concentrations (Runs 28 – 32)



Figure 6: 41 nm soot at a range of concentrations (Runs 43 – 47)



Figure 8: 20,000  $\text{cm}^{-3}$  sintered Ag at a range of sizes (Runs 1, 6, 11, 16)

 $-25.0$ 



### **4. DISCUSSION AND CONCLUSIONS**

### **4.1. SUMMARY OF RESULTS**

In general terms, two aspects of this type of measurement are examined by this comparison: firstly, the ability of the participants to calibrate their CPCs in the "plateau" region of CPC operation; and secondly, the range of particle size, concentration and material over which the calibration is valid.

#### 4.1.1 Plateau region

With 100 nm soot, the largest particle size used (which should be most comfortably in the plateau region of the CPCs), there is agreement, with the exception of the JRC results, at the level of about ±7% (Figure 5). This level of agreement holds across the full range of concentrations, between 100 and 20,000 cm<sup>-3</sup>. The exception was from one of the TSI 3790 instruments with a relatively high 50% cut-off size of 23 nm, whose results are in contrast to those of the similar instrument PTB(2).

With 60 nm sintered silver, the largest silver particle size used, the two TSI 3790 instruments underread by similar amounts, while agreement between the other instruments was similar to the 100 nm soot case, at about ±10% (Figure 1).

4.1.2 Effect of particle size, concentration and material

With the understandable exception of the TSI 3790 instruments at sizes below 80 nm, the level of agreement was consistent at about ±10% over the full range of concentrations used (100 to 20,000 cm<sup>-3</sup>), and for each of the three particle materials (unsintered silver, sintered silver, and soot), for sizes down to 23 nm (Figures 2, 3, 4, 6, 7 and 9).

Agreement at 13 nm particle size, for the three participants who reported it, is less good, as expected because of the proximity to the 50% cut-off size (Figure 8).

### **4.2. SUPPORTED CMC CLAIMS**

It is proposed that this comparison can be used to support CMC claims for condensation particle counter calibrations in the range 100 to 20,000  $cm^{-3}$ .

### **5. REFERENCES**

[1] ISO/DIS 27891: Aerosol particle number concentration — Calibration of condensation particle counters

### **APPENDICES**

### **APPENDIX A1 – EURAMET 1282 PROTOCOL**

### EURAMET 1282

### **Comparison of Condensation Particle Counters**

Coordinating Laboratory: NPL, UK Host: TROPOS, Leipzig, Germany

### **Protocol (final version)**

### **Background**

Aerosol particle number concentration has recently featured in vehicle emission legislation and is becoming increasingly important in other areas such as ambient air monitoring. Number concentration measurements are also often integral to particle size distribution measurements, such as when using a Differential Mobility Analyzer System.

Condensation Particle Counters (CPCs) are the usual type of instrument for measuring particle number concentration in the size range from a few nanometers to a few micrometers. These instruments have a large size range over which they have constant detection efficiency for nanoparticles of all compositions (the "plateau" region), and an instrument and particlematerial dependent drop in detection efficiency at low sizes. The drop in detection efficiency at large sizes is of much lower importance, as the number of larger particles is negligible.

Calibration of CPCs can be done via comparison with a reference CPC or a reference aerosol electrometer. Procedures for doing this have been set out in ISO/DIS 27891. The DIS refers to the role of NMIs in providing certification for reference aerosol electrometers and reference CPCs.

Although not strictly a chemical measurement, the comparison belongs in the Gas subcommittee of TC-MC because of the similarity to gas concentration measurements, following the precedent of earlier projects 893 (workshops to establish "Metrology infrastructure for airborne nanoparticles"), 1027 ("Comparison of combustion particle number concentration and size"), and 1244 ("Comparison of aerosol electrometers"), which took place in March 2013 as part of the same EMRP project, ENV02 PartEmission.

The aim of this comparison is twofold:

- (1) to compare the accuracy of different laboratories' measurements of particle number concentration in the plateau region of their CPC, as in a traditional metrological comparison, and
- (2) to measure the detection efficiencies of the CPCs at sizes below the plateau region using a selection of common particle sources. In this case there are no "correct"

answers, and the aim is to provide information to the participants.

As in the EURAMET 1244 comparison of aerosol electrometers, EURAMET participants are being joined by other participants with expertise in this area.

### **Comparison protocol**

The comparison will be held at TROPOS in Leipzig, Germany, during the week 14-18 October 2013.

Participants will be responsible for the transport of their instruments to and from Leipzig, and for their setting up and operation. This includes the independent calibration of the CPCs and any flow meters used and the collection of data. Butanol can be provided by TROPOS if necessary.

The electricity supply at Leipzig is 230V 50Hz with CEE 7/4 socket (plug type F). Participants must provide their own electrical adaptors if necessary.

Participants will sample the test aerosol (particles+nitrogen) at flow rates that have been arranged individually (in the range  $0.3$  to 1.5 litre/min (at  $25^{\circ}$ C and 101.3 kPa)), with diffusion losses compensated by differing lengths of sample tubing. Participants are expected to take readings every second. Participants' CPCs must connect to ¼-inch TSI conductive tubing. The outlet connection of each CPC (i.e. connection to the vacuum line, if needed) should be either a  $\frac{1}{4}$ " Swagelok tube connector or a  $\frac{1}{4}$ " tube. Participants must provide their own adaptors if needed.

Particles will be mainly evaporated/condensed Ag nanoparticles between 6 nm and 60 nm in size, both sintered and not sintered, and CAST generated soot particles between 23 nm and 100 nm in size. Where possible, there will be 5 target concentrations between 100 and 20,000 particles cm<sup>-3</sup>.

The measurement period for each run will last for 10 minutes, with a "clean air" interval between runs lasting 5 minutes.

Particle number concentrations are to be reported at standard conditions (25°C and 101.3 kPa, as in the Tampere comparison). Data on the sample temperature and pressure will be supplied.

The schedule for the week is expected to be:



20:00 Joint Dinner

The brown colored runs are to be reported as "plateau" comparison runs, even though some sizes will be well below the plateau region. Results that are clearly below the plateau region will be evaluated separately in the EURAMET report.

The blue colored runs are for the information of the participants, and will not be formally reported on the proforma (below).

On each day, some time will be reserved for data processing.

### **Reporting of the results**

The final results are to be reported, with volume corrected to standard conditions, on the proforma sheets attached. It is expected that these will be submitted by participants after they have returned to their laboratories to allow subsequent checks on the equipment.

Participating laboratories should specify the method and calibration procedure used for the comparison in detail. They should also state the route through which the calibration procedure provides traceability to the SI.

The expanded uncertainty for each measurement in the plateau region should also be calculated. Information should be provided about how the uncertainty budget was calculated.

NPL and TROPOS together will be responsible for collecting and reporting measurement results.

#### **Points of contact:**

General contacts and reporting of the results for the comparison paul.quincey@npl.co.uk tuch@tropos.de

### **APPENDIX A2 – PARTICIPANTS RESULTS PROFORMAS**

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

# Results Proforma

Participant laboratory and people involved:

NPL Paul Quincey Dimitris Sarantaridis

Model / origin of CPC:

TSI CPC 3775

Method of flow control:

Volumetric – critical orifice.

Calibration methods and traceability:

CPC: Calibrated against reference FCE (GRIMM FCE model: 5.705). Reference FCE calibrated using a voltage source (Keithley 213), a 1  $G\Omega$  standard resistor (Welwyn) traceable to NPL primary standards of resistance, and a voltmeter (HP 3458A) traceable to NPL primary standards of voltage.

Flow meter (model MKS 1179A): Calibration performed by determining mass loss from a cylinder of synthetic air during a measured time interval. Traceability to NPL mass standards.

### Components included in the uncertainty calculation:

- 1. CPC random uncertainty: standard deviation of the mean concentration measured for every 5 min run.
- 2. CPC flow rate random uncertainty.
- 3. CPC calibration uncertainty.
- 4. Temperature correction uncertainty.
- 5. Pressure correction uncertainty.



Date results submitted: 27 November 2013

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

### Results Proforma

Participant laboratory and people involved: **TROPOS** Thomas Tuch

Model / origin of CPC: 3772 CPC, TSI Inc. Condenser temperature set to 18 deg. C

Method of flow control: 1 L/min critical orifice

Calibration methods and traceability:

 $CPC$  counting efficiency calibrated at concentrations  $> 1000$  cm<sup>-3</sup> with aerosol electrometer and flow meter. Traceability to SI units is through the ampere and mass flow rates.

CPC linearity calibrated at high and low concentrations with dilution proportionality test, and validated at high concentration against aerosol electrometer linearity test.

Components included in the uncertainty calculation:

Three components are included in the uncertainty calculation of concentration measurements with the CPC. The type B uncertainties in CPC counting efficiency and CPC inlet flow rate are taken from the CPC and flow meter calibration certificates, respectively. The type A uncertainty is calculated as the standard deviation of the data set of concentrations for each measurement.

Date results submitted: 27 November 2013

Filled proformas are to be sent to: [paul.quincey@npl.co.uk](mailto:paul.quincey@npl.co.uk) and [tuch@tropos.de](mailto:tuch@tropos.de) by 29 November 2013.





Date results submitted:  $27<sup>th</sup>$  November 2013.

# EURAMET 1282 – Comparison of condensation particle counters TROPOS 14-18 October 2013

# Results Proforma (4th December 2013)

### Participant laboratory, and people involved:

Tampere University of Technology (TUT), Aerosol physics laboratory, Jaakko Yli-Ojanperä Mikes, Thermal and mass group, Richard Högström

Model / origin of CPC:

Airmodus A20 single flow butanol CPC

Method of flow control:

critical orifice just before the outlet of the CPC

Calibration methods and traceability:

The CPC has been calibrated using the Single Charged Aerosol Reference (SCAR) at Tampere University of Technology. The SCAR is a Faraday cup aerosol electrometer based number concentration standard.

Faraday cup aerosol electrometer calibration and traceability:

The current measurement function of the electrometer was calibrated with a current source based on a high value reference resistor and a direct voltage source. Traceability of the reference resistor is based on a calibration chain starting from MIKES Quantum-Hall resistance standard. Traceability of the Fluke 5440B direct voltage source is based on a calibration chain starting from MIKES Josephson direct voltage standard

Mass flow meter calibration and traceability:

The flow meter was calibrated against the LFE calibration system. The operation of the LFE is based on laminar flow elements (molbloc, DH Instruments) and it is calibrated against the dynamic weighing system (DWS1). The operation of the DWS1 is based on dynamic gravimetric weighing of a gas vessel. Therefore, mass flow measurements are traceable to the definitions of mass and time.

The electrometer and the mass flow meter were calibrated before the campaign and the CPC was calibrated after the campaign.

Components included in the uncertainty calculation: Electrometer calibration correction, type B. Flow meter calibration correction, type B. Standard deviation of the measured concentration (CPC), type A CPC calibration correction, type B



### NPL Report AS 94



Date results submitted:

4 December 2013

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

### Results Proforma

Participant laboratory and people involved:

METAS, Felix Lüönd

Model / origin of CPC:

METAS Grimm 5412, S/N 54121103

Method of flow control:

Internal pump and flow controller, flow was continuously monitored by an external mass flow meter (Vögtlin Red-y smart series, S/N 150874) at the exhaust of the CPC. A cold trap was used downstream of the CPC exhaust to prevent butanol vapour from influencing the flow measurement.

Calibration methods and traceability:

The flow meter was calibrated against the corresponding METAS primary standard in 2012. The corresponding calibration data were used to correct the measured flow. As the calibration was done with air, a correction factor of 1.002 was used to correct the flow readings obtained with nitrogen during the campaign. As the flow meter measures mass flow, no information about aerosol temperature and pressure during the measurements is required.

The CPC broke during transport to the campaign. Therefore, it was calibrated again after the campaign against the METAS primary standard for particle number concentration (TSI 3068B electrometer, S/N 70701106). This calibration also involved two mass flow meters calibrated against the METAS primary standard for flow in 2011. The electrical part of the electrometer was calibrated in 2013 against the METAS primary standard for small DC current (as low as 10 fA).

The calibration of the CPC was done according to the ISO 27891 draft with miniCAST particles at the sizes 10 nm, 23 nm, 41 nm, 80 nm, and 100 nm for concentrations  $\leq 10'000$ cm-3. For each particle size, the counting efficiency of the CPC was measured in 6 repetitions. Each repetition included subtraction of the electrometer offset and a correction for multiply charged, larger particles. The uncertainty in the counting efficiency averaged over the 6 repetitions contains contributions from both the variability of the instrument readings recorded at 1 Hz frequency and from the variability of the counting efficiency between the individual repetitions (this results in a conservative estimate of the uncertainty because the two mentioned variabilities can partly have the same origin). Deviations from the ISO 27891 protocol in terms of the number of voltage levels taken into account in order to correct the measured concentrations for multiply charged particles were accepted when correction of a specific multiple charge level influenced the resulting counting efficiency by less than 1%. Small particle size or size selection in the far downslope of the initial size distribution reduced the number of required voltage levels usually to two or even one (i.e. no multiple charge correction at all, as in the case of 10 nm particles).

#### NPL Report AS 94

More detailed information about the calibration procedure and the uncertainty calculation can be given on request.

### Components included in the uncertainty calculation:

- Variability (type A uncertainty) of the CPC reading during a 5 min measurement, i.e. standard deviation of the measured values divided by the square root of the number of 1s readings. However, the timing of the individual 5 min measurements was not always precise during the campaign. This leads to a higher uncertainty than just the one based on the standard deviation within the 5 min measurement in cases where the number concentration drifts with time. As in some runs with CAST aerosol the number concentration drifted, we estimated the uncertainty due to the timing of the measurement by estimating the variability of the mean concentration from a data window shifted by  $+$ -1 min compared to the original window. We used the maximum of this variability due to timing uncertainty and the variability based on the 1s readings during the used 5 min measurement as an estimate for the uncertainty of the average CPC concentration.
- Uncertainty in flow measurement: This includes the variability (type A) of the flow measured during the used 5 min period of a measurement as well as a type B contribution from the calibration of the flow meter.
- Uncertainty in the counting efficiency of the CPC as determined during the calibration of the CPC against the reference electrometer.

The resulting particle number concentration  $C_{CPC}$  is given by

$$
C_{CPC} = \frac{C_{meas}}{\eta_{CPC}} f_q
$$

where

$$
f_q = \frac{q_{nom}}{q_{meas}} \cdot A
$$

with  $q_{nom}$  being the nominal volumetric flow rate of the CPC (0.6 lpm),  $q_{meas}$  being the flow measured by the mass flow meter (after correction according to the calibration of the flow meter), and *A* being a factor referring the flow measurements to standard conditions (*A* is a constant as no temperature or pressure measurements are involved).  $C_{meas}$  denotes the average measured particle concentration, and  $\eta_{CPC}$  is the counting efficiency of the CPC as determined in the calibration.

The relative uncertainties of the above mentioned influence quantities are added quadratically to obtain the relative uncertainty  $u(C_{CPC})$ .

### Remarks:

- For the METAS Grimm 5412 CPC, data are only available from run 33 due to the CPC being under repair on October 14 and 15.
- For a particle diameter of 23 nm (runs 48 52), the METAS CPC has a detection efficiency of 96.9% which is only compatible with the 100% counting efficiency at sizes above 41 nm if uncertainties in the detection efficiency are taken at 95% confidence interval  $(k = 2)$ . Nevertheless, given the high counting efficiency, 23 nm has been considered as part of the plateau region.
- After the campaign, the METAS 5412 CPC returned to Grimm for further tests which

had been postponed after repair due to the tight schedule of the campaign. According to the Grimm engineers, these tests did not alter the calibration of the CPC. Therefore, the later calibration performed at METAS is considered as valid and representative for the calibration during the campaign.



Date results submitted:  $17<sup>th</sup>$  December 2013; with revised uncertainties  $1<sup>st</sup>$  April 2014

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

# Results Proforma

### Participant laboratory and people involved:

PTB, Arne Kuntze (calibration at TROPOS), Anke Jordan-Gerkens (calibration at TROPOS), Andreas Nowak (data analysis)

Model / origin of CPC: TSI 3772, CPC, temperature difference at 17°C

### Method of flow control:

The flow is adjusted at 1 L/min about a critical orifice. The flow was checked frequently during the workshop against PTB blow flow meter. Currently, we don't have a traceable method, which monitored directly the flow of CPC.

After the workshop, the flow of CPC was calibrated against the primary standard for mass flow measurements at PTB. The calibration of the critical orifice was performed at standard conditions. For that reason, a constant factor was used to correct the particle concentration like:

> $N_{total} = \frac{f_{nomin}}{f_{optinat}}$ *Jcalibr* \* *I*V  $f_{nominal} = 1.0$  l/min  $f_{\text{calibration}} = 1.007$  l/min

We also used the serial output of CPC for detection of the particle number concentration.

### Calibration methods and traceability:

The CPC counting efficiency is calibrated at concentrations > 1000 cm-3 with aerosol electrometer (AE) based on a soot aerosol generated from MINI-CAST. For the AE the charging of capacity was measured against primary standard at PTB. The method is traceable for SI units F, V and s.

### Components included in the uncertainty calculation:

Several parts were included in the calculation of the uncertainty budget:

- 1.) type A: based on the empirical uncertainty:  $u(\bar{q}) = \frac{^{3}p(q)}{\sqrt{n}}$
- 2.) type B: based on several assumptions:
	- a. for the uncertainty of flow calibration:  $u(flow) = 0.2 \%$
	- b. for the bias of flow splitter at TROPOS: u (bias) =  $3\%$
	- c. for the uncertainty of the multiple charge correction:  $u$ (charge) = 2% for particle  $> 40$  nm

Both types were combined to calculate the uncertainty budget following the Guide to the expression of uncertainty in measurement (GUM 5.1.1., JCGM 104:2009). The formula for non correlated input quantities was used:

$$
u_c(y) = \sqrt{\sum_{i=1}^N (c_i * u(x_i))^2}
$$

sensitivity coefficient  $c_i=1$ 



Date results submitted:  $10^{th}$  January 2014; modified version with revised uncertainties  $12^{th}$ May 2014

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

### Results Proforma

### Participant laboratory and people involved:

PTB, Arne Kuntze (calibration at TROPOS), Anke Jordan-Gerkens (calibration at TROPOS), Andreas Nowak (data analysis)

Model / origin of CPC: TSI 3790, EECPC

### Method of flow control:

The flow is adjusted at 1 L/min about a critical orifice. The flow was checked frequently during the workshop against PTB blow flow meter. Currently, we don't have a traceable method, which monitored directly the flow of CPC.

After the workshop, the flow of CPC was calibrated against the primary standard for mass flow measurements at PTB. The calibration of the critical orifice was performed at standard conditions. For that reason, a constant factor was used to correct the particle concentration like:

$$
N_{total} = \frac{f_{nominal}}{f_{calibration}} * N_{serial}
$$

 $f_{nominal} = 1.0$  l/min  $f_{\text{calibration}} = 0.959$  l/min

We also used the serial output of CPC for detection of the particle number concentration.

### Calibration methods and traceability:

The CPC counting efficiency is calibrated at concentrations > 1000 cm-3 with aerosol electrometer (AE) based on a soot aerosol generated from MINI-CAST. For the AE the charging of capacity was measured against primary standard at PTB. The method is traceable for SI units F, V and s.

### Components included in the uncertainty calculation:

Several parts were included in the calculation of the uncertainty budget:

- 1.) type A: based on the empirical uncertainty:  $u(\bar{q}) = \frac{3p(q)}{\sqrt{n}}$
- 2.) type B: based on several assumptions:
	- a. for the uncertainty of flow calibration:  $u(flow) = 0.2 \%$
	- b. for the bias of flow splitter at TROPOS: u (bias) =  $3\%$
	- c. for the uncertainty of the multiple charge correction:  $u$ (charge) = 2% for particle > 40 nm

Both types were combined to calculate the uncertainty budget following the Guide to the

expression of uncertainty in measurement (GUM 5.1.1., JCGM 104:2009). The formula for non correlated input quantities was used:

$$
u_c(y) = \sqrt{\sum_{i=1}^N (c_i * u(x_i))^2}
$$

sensitivity coefficient  $c_i=1$ 



Date results submitted:  $10^{th}$  January 2014; modified version with revised uncertainties  $12^{th}$ May 2014

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

# Results Proforma

Participant laboratory and people involved: JRC, Francesco Riccobono

Model / origin of CPC: TSI 3790

Method of flow control: Critical orifice

Calibration methods and traceability: CPC inlet flow measured twice a day with a primary bubble flow meter. CPC counts based on calibration performed by the manufacturer's calibration service.

Components included in the uncertainty calculation:

Standard deviation of CPC concentration measured during one run

Uncertainty on the CPC inlet flow rate measurement

Uncertainty on temperature measurement

Uncertainty on pressure measurement

Uncertainty of the calibration of the CPC by the manufacturer





Date results submitted:  $4^{\text{th}}$  December 2013

Ŀ

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

### Results Proforma

Participant laboratory and people involved: U.S. Army Primary Standards Laboratory Miles Owen

Model / origin of CPC: 3772 CPC, TSI Inc.

Method of flow control: 1 L/min critical orifice

Calibration methods and traceability:

CPC counting efficiency calibrated at high concentration with aerosol electrometer and flow meter. Traceability to SI units is through the ampere and mass flow rates.

PAO oil (emery oil) was used as the calibration material.

CPC linearity calibrated at high and low concentrations with dilution proportionality test, and validated at high concentration against aerosol electrometer linearity test.

Components included in the uncertainty calculation:

Three components are included in the uncertainty calculation of concentration measurements with the CPC. The type B uncertainties in CPC counting efficiency and CPC inlet flow rate are taken from the CPC and flow meter calibration certificates, respectively. The type A uncertainty is calculated as the standard deviation of the mean for each measurement.





Date results submitted: 9 December 2013

# EURAMET 1282 – Comparison of Condensation Particle Counters TROPOS 14-18 October 2013

# Results Proforma

Participant laboratory and people involved: NMIJ/AIST

Dr. Kenjiro Iida and Dr. Hiromu Sakurai

Model / origin of CPC: TSI 3772

Method of flow control:

Critical orifice with external vacuum source

Calibration methods and traceability:

The CPC was calibrated against the primary, FCAE-based number concentration standard at AIST as reference, with sucrose (up to 30 nm), poly-alpha-olefin (30 nm to 50 nm) and polystyrene latex particles (100 nm). The charge concentration measurement by the primary FCAE standard has its SI traceability established for the current and volumetric flow rate measurements.

Components included in the uncertainty calculation:

- Uncertainty in the calibration by the primary standard of AIST, which was size and concentration dependent
- Uncertainty due to the variation of the detection efficiency between the measurement at TROPOS and the calibration at AIST
- Uncertainty due to the variation of the flow rate between the measurement at TROPOS and the calibration at AIST
- Uncertainty due to the difference in the particle type between the measurement at TROPOS and the calibration at AIST





Date results submitted: 2 January 2014