SYSTEM AND PERFORMANCE AUDIT OF SURFACE OZONE, CARBON MONOXIDE, METHANE, CARBON DIOXIDE AND NITROUS OXIDE AT THE

GLOBAL GAW STATION LAUDER NEW ZEALAND NOVEMBER 2016

Submitted to the World Meteorological Organization by

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WCC-Empa Report 16/5

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EXECUTIVE SUMMARY AND RECOMMENDATIONS

The second system and performance audit by WCC-Empa[1] at the global GAW station Lauder was conducted from 21 - 25 November 2016 in agreement with the WMO/GAW quality assurance system (WMO, 2017). A previous audit at the Lauder GAW station was made by WCC-Empa in March 2010 (Zellweger et al., 2010).

The following people contributed to the audit:

Dr. Christoph Zellweger	Empa Dübendorf, WCC-Empa
Mr. Dan Smale	NIWA Lauder, Station manager, Lauder in-situ PI
Mr. Gordon Brailsford	NIWA Wellington, Baring Head in-situ PI
Mrs. Audra McClure	NOAA Boulder (PI ozone measurements)

This report summarises the assessment of the Lauder GAW station in general, as well as the surface ozone, methane, carbon dioxide, carbon monoxide and nitrous oxide measurements in particular.

The report is distributed to the Lauder station manager, the New Zealand GAW country contact, the ozone group at NOAA and the World Meteorological Organization in Geneva. The report will be posted on the internet (www.empa.ch/web/s503/wcc-empa).

The recommendations found in this report are graded as minor, important and critical and are complemented with a priority (*** indicating highest priority) and a suggested completion date.

Station Management and Operation

Lauder is operated by the New Zealand National Institute of Water and Atmospheric Research (NIWA). The operation and maintenance of the station is well organized, with clear assignments of responsibilities. Lauder is visited during weekdays by approximately 10 scientists, technical and administrational staff. A few staff members are resident onsite, which enables quick respond to any on-site situation/issue occurring outside normal office hours.

Station Location and Access

Lauder is located in a sparsely populated rural environment on the South Island of New Zealand at 45.0ºS, 169.7ºE, altitude 370 m a.s.l. The surroundings have not significantly changed since the last WCC-Empa audit in 2010. Access to the site is possible by road throughout the year. The station location is adequate for the intended purpose.

Station Facilities

The Lauder (LAU) station comprises extensive laboratory and office space. Kitchen and sanitary facilities as well as on-site housing for visiting researchers are available. Internet access is available with sufficient bandwidth. It is an ideal platform for continuous atmospheric research as well as for extensive measurement campaigns.

Measurement Programme

The LAU station comprises a comprehensive measurement programme that covers all focal areas of the GAW programme. The Atmospheric Research facility was established in 1961 to facilitate research of the upper atmosphere (mainly ionospheric), but changed to research of stratospheric composition and UV radiation in the 1980s. Over the last decade the focus has increasingly moved

[1] WMO/GAW World Calibration Centre for Surface Ozone, Carbon Monoxide, Methane and Carbon Dioxide. WCC-Empa was assigned by WMO and is hosted by the Laboratory for Air Pollution and Environmental Technology of the Swiss Federal Laboratories for Materials Testing and Research (Empa). The mandate is to conduct system and performance audits at Global GAW stations every 2 – 4 years based on mutual agreement.

to include climate change, tropospheric chemistry and full spectrum radiation research. This makes LAU a valuable atmospheric measurement site with a combination of ground-based remote sensing, balloon-sonde, in-situ and flask measurements. NIWA has a long term in situ flask sampling programme at Baring Head and Arrival Heights, Antarctica (Lowe et al., 1994; Moss et al., 2012). The flask samples collected at Lauder follow the same collection methods and laboratory analysis. For the time of the audit at LAU, the surface ozone analysers from the Baring Head station were brought to LAU to be also compared during the audit. The regional GAW station Baring Head (BAR), which is located in pristine environment on the North Island, has also an extensive measurement programme focusing on surface measurements. BAR is regarded as the main GAW site for as the primary GAW surface measurement station in New Zealand. An overview on measured species is available from GAWSIS and the station web sites of LAU (https://www.niwa.co.nz/atmosphere/facilities/lauder-atmospheric-research-station) and BAR (https://www.niwa.co.nz/atmosphere/facilities/baring-head).

Recommendation 1 (*, important, ongoing)**
It is recommended to update GAWSIS yearly or when major changes occur. Part of the reviewed information was not up to date while others (e.g. information on in-situ GHG measurements) were entirely missing.

Data Submission

Data has been submitted to the World Data Centre for Greenhouse Gases (WDCGG) for CH_4 (2007-2016) and surface ozone (2003-2015). For CH_4, only hourly data for the period 3pm - 4pm (local time) has been submitted. This period is considered to represent well mixed background air if the hourly mean wind speed is exceeding 5 ms^{-1} at the same time. Data shown in this report was accessed on 23 November 2017. CO, CO_2 and N_2O data has not yet been submitted.

Recommendation 2 (*, important, ongoing)**
Data submission is an obligation of all GAW stations. It is recommended to submit data to the corresponding data centres at least in yearly intervals. One hourly data must be submitted for all parameters.

Data Review

As part of the system audit, data within the scope of WCC-Empa available at WDCGG were reviewed. Summary plots and a short description of the findings are presented in the Appendix.

Documentation

All information is entered in electronic and hand written log books. The instrument manuals are available at the site, and weekly checklists are available. Standard Operating Procedures (SOPs) have been prepared by NIWA. The reviewed information was comprehensive and up-to-date.

Air Inlet System

The design of the air inlet systems has not been changed since the last audit by WCC-Empa. The FTIR instrument is using its own dedicated inlet line leading to the top of the 10 meter tower which is located approximately 40 meters away of the laboratory building. The ozone inlet is mounted directly on the roof of the laboratory building. Both inlet systems are adequate, and no change is required.

Surface Ozone Measurements

The surface ozone measurements at LAU were established in collaboration with the National Oceanic and Atmospheric Administration (NOAA) in 2003, and continuous time series are available since then. Data evaluation and submission is still made by NOAA.

Instrumentation. LAU is equipped with one ozone analysers (Thermo Scientific 49C). No ozone standard is available, but the instrument is calibrated every few years using a transfer standard which is shipped to LAU from NOAA. At the time of the audit, two ozone analysers from the regional GAW station Baring Head (BHD) were also available at LAU for comparison with the Empa reference instrument.

Data Acquisition. Data (1-min time resolution) is currently manually downloaded using the Thermo Scientific iPort software. All instrument parameters are available with iPort, but download requires manual intervention, and data is not available in near-real time.

Intercomparison (Performance Audit). The LAU and BHD analysers were compared against the WCC-Empa travelling standard (TS) with traceability to a Standard Reference Photometer (SRP). The internal ozone generator of the WCC-Empa transfer standard was used for generation of a randomised sequence of ozone levels ranging from 0 to 90 ppb. The result of the comparisons is summarised below with respect to the WMO GAW Data Quality Objectives (DQOs) (WMO, 2013). The data was acquired by the WCC-Empa data acquisition system, and no further corrections were applied to the BHD instrument. Data of the LAU instrument were corrected by the function provided by NOAA. The raw data was multiplied by 1.029 and an offset of 0.16 ppb was added. This is still the same correction function as in 2010. A NOAA travelling standard (2B tech model 306) was also sent to LAU in May 2016; however, the correction function provided by NOAA for this calibration (LAU Final O_3 = Raw O_3 * 1.147 + 0.78 ppb) seems to be off, and NOAA is currently investigating the cause of the large change. It was therefore decided to use the 2010 function. The following equations characterise the bias of the instruments:

> **Recommendation 3 (***, important, 2018)**
> *It is recommended that the NOAA travelling standard is shipped more frequently (at least every two years) to Lauder. Alternatively, it could be considered to purchase an ozone calibrator which could be used as a travelling reference instrument for calibrations of ozone instruments at all stations operated by NIWA.*
>
> **Recommendation 4 (***, important, 2018)**
> *NOAA currently uses instrument from Thermo Scientific and 2B Technologies as travelling standards. It was recognised that calibrations using the 2B travelling standard are less reliable compared to Thermo Scientific 49C-PS / 49i-PS ozone calibrators. It is therefore recommended that Thermo Scientific travelling standards are used for calibrations.*

Thermo Scientific 49C #0326101959 (LAU) (BKG 0.0 ppb, SPAN 1.000, corrected using above calibration function):

Unbiased O_3 mole fraction (ppb): X_{O3} (ppb) = ([OA] + 0.31 ppb) / 1.0101 (1a)

Standard uncertainty (ppb): u_{O3} (ppb) = sqrt (0.29 ppb^2 + 2.61e-05 * X_{O3}^2) (1b)

Thermo Scientific 49i #0532213220 (BHD) (BKG 0.0 ppb, SPAN 1.000), no corrections applied:

Unbiased O_3 mole fraction (ppb): X_{O3} (ppb) = ([OA] - 0.19 ppb) / 0.9721 (1c)

Standard uncertainty (ppb): u_{O3} (ppb) = sqrt (0.32 ppb^2 + 2.88e-05 * X_{O3}^2) (1d)

Thermo Scientific 49i #01152220033 (BHD) (BKG -0.2 ppb, SPAN 1.027), initial settings:

Unbiased O_3 mole fraction (ppb): X_{O3} (ppb) = ([OA] - 0.59 ppb) / 1.0293 (1e)

Standard uncertainty (ppb): u_{O3} (ppb) = sqrt (0.33 ppb^2 + 2.73e-05 * X_{O3}^2) (1f)

Thermo Scientific 49i #01152220033 (BHD) (BKG -0.1 ppb, SPAN 0.997), final:

Unbiased O_3 mole fraction (ppb): X_{O3} (ppb) = ([OA] + 0.19 ppb) / 0.9996 (1g)

Standard uncertainty (ppb): u_{O3} (ppb) = sqrt (0.33 ppb^2 + 2.71e-05 * X_{O3}^2) (1h)

The results of the comparison are further presented in the following Figures.

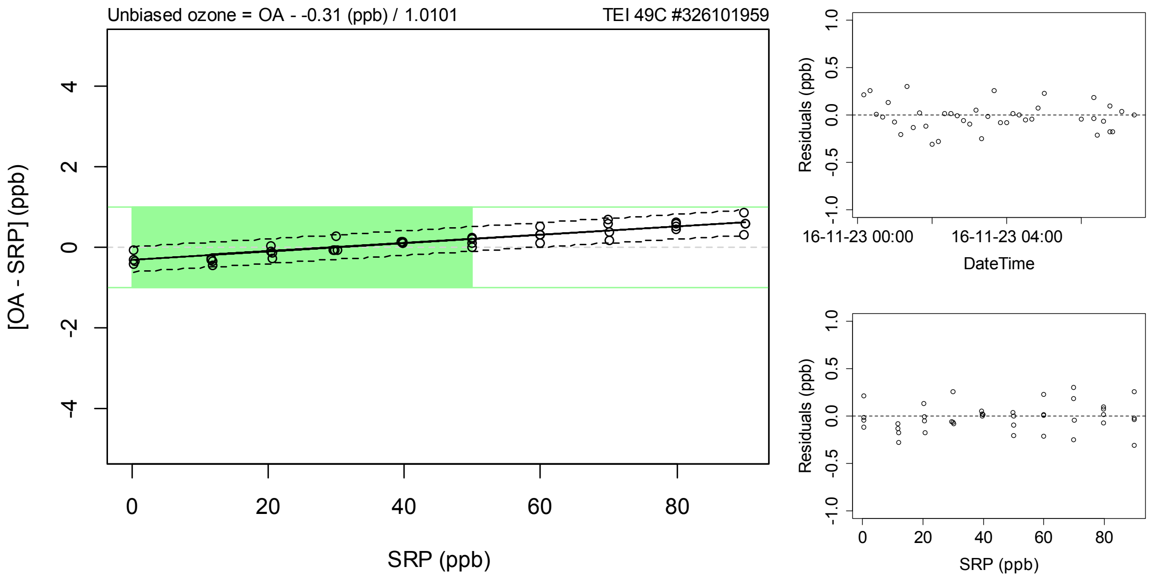

Figure 1. *Left: Bias of the LAU ozone analyser (Thermo Scientific 49C #0326101959) with respect to the SRP as a function of mole fraction. Each point represents the average of the last 5 one-minute values at a given level. The green area corresponds to the relevant mole fraction range, while the DQOs are indicated with green lines. The dashed lines about the regression lines are the Working-Hotelling 95% confidence bands. Right: Regression residuals of the ozone comparisons as a function of time (top) and mole fraction (bottom).*

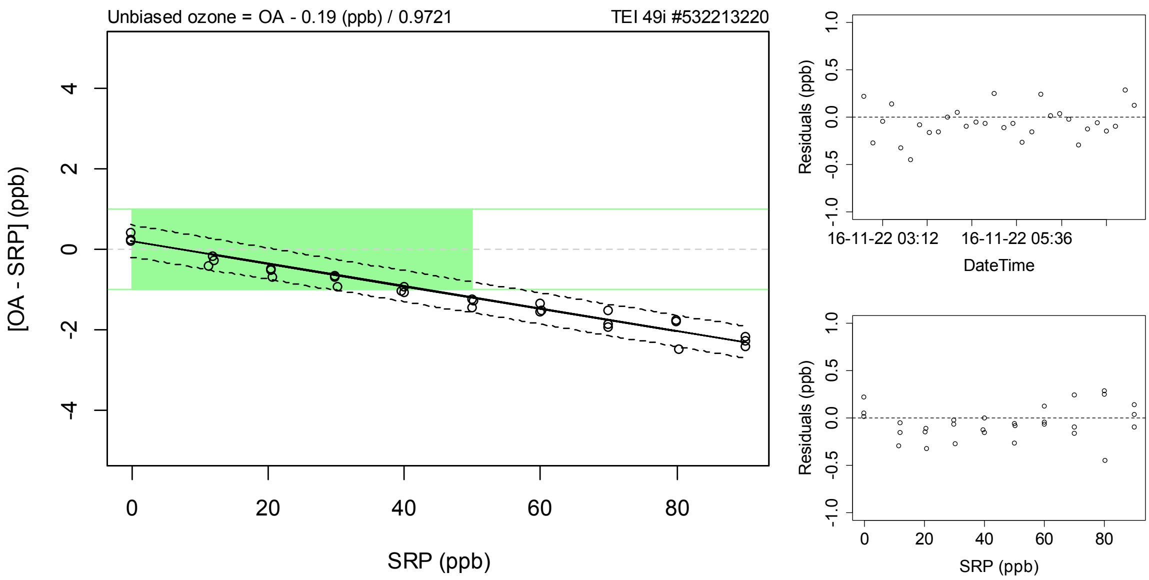

Figure 2. *Same as above for the BHD ozone analyser (Thermo Scientific 49C #0532213220).*

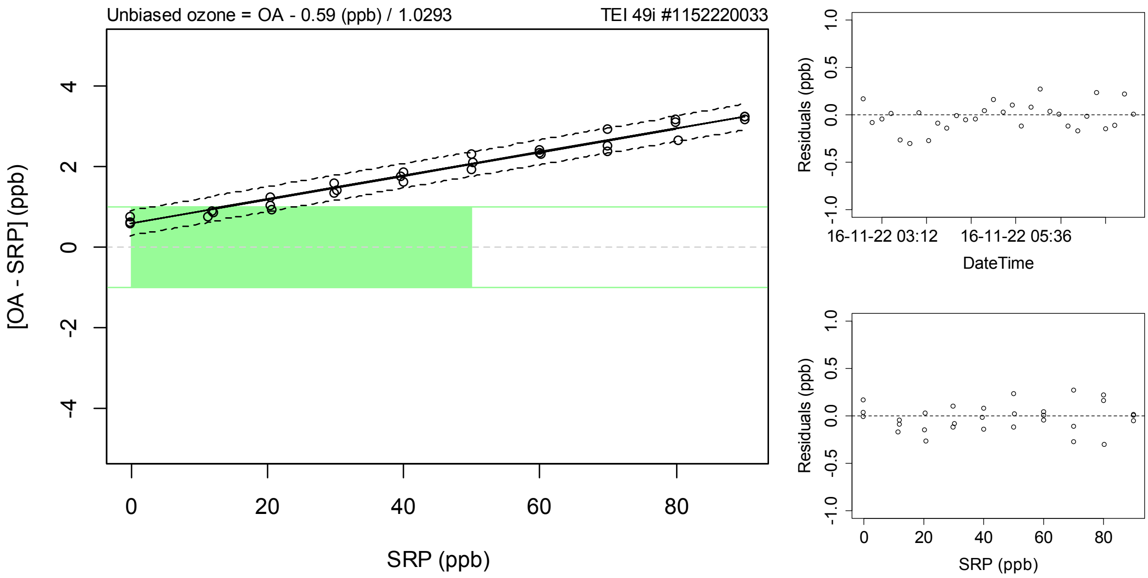

Figure 3. *Same as above for the future BHD ozone analyser with initial (factory) calibration settings (Thermo Scientific 49C #01152220033).*

After adjustments of the calibration settings, the results were as follows:

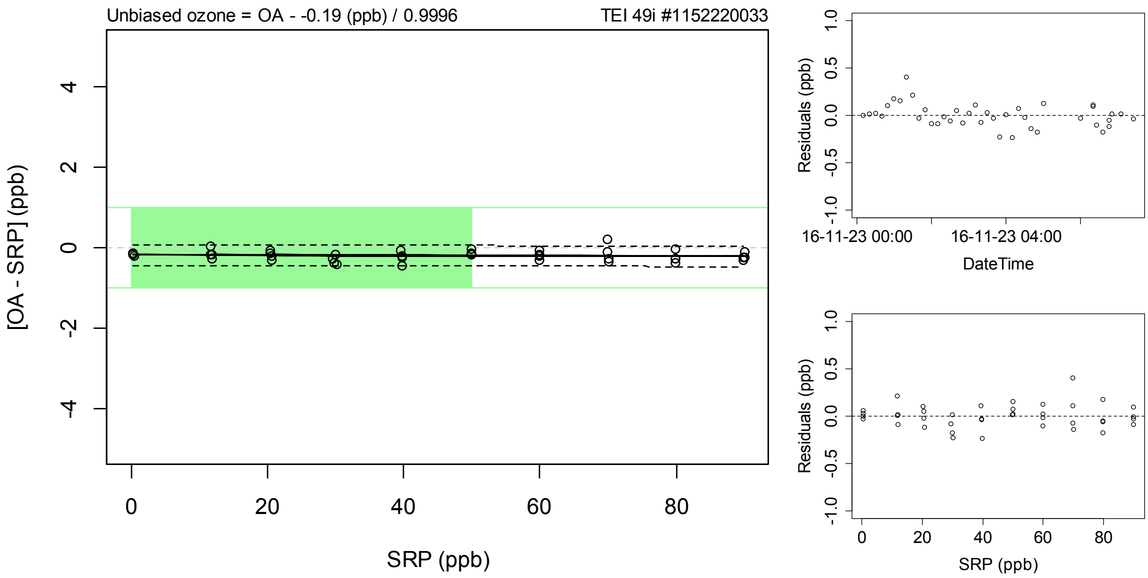

Figure 4. *Same as above for the future BHD ozone analyser with final calibration settings (Thermo Scientific 49C #01152220033).*

The results of the surface ozone audit can be summarised as follows:

Good agreement between the WCC-Empa travelling instrument and the LAU analyser was found when the calibration function provided by NOAA in 2010 was used. The newer correction function from May 2016 however was identified to be invalid.

The main BHD analyser was measuring low compared to the WCC-Empa reference, and a correction should be applied. The instrument that will replace the BHD analyser fully complies with the DQOs with the new calibration settings.

Recommendation 5 (, important, 2018)**
Care must be taken when determinations of the calibration function with the NOAA travelling are made. If large differences are observed, as it happened during the last check in 2016, the reason must be identified.

Recommendation 6 (, important, 2018)**
The main BHD ozone instrument is uncalibrated, and no corrections have been applied so far. It is recommended to correct ozone data using equation (1c).

Carbon Monoxide Measurements

Carbon monoxide measurements at LAU were established in 2007, and continuous time series are available since then.

Instrumentation. LAU is equipped with an in-situ mid-infrared Fourier Transform Spectrometer (IFTS). The IFTS was developed and constructed at the University of Wollongong, Australia. The version installed at LAU in 2007 was a prototype system of that described in Griffith et al. (2012) and Hammer et al. (2013). In April 2013 the LAU prototype analyser had a major software and hardware upgrade to bring it into line with the 2013 commercial version of the instrument manufactured by Ecotech.

Standards. A set of reference standards is used to calibrate the IFTS analyser. The standards have been calibrated by NIWA against NOAA laboratory standards. A list of available standards is given in the Appendix.

Intercomparison (Performance Audit). The comparison involved repeated challenges of the LAU instrument with randomised carbon monoxide levels using WCC-Empa travelling standards. The following equations characterise the instrument bias, and the results are further illustrated in Figure 5 with respect to the WMO GAW DQOs (WMO, 2014):

IFTS analyser:

Unbiased CO mixing ratio: X_{CO} (ppb) = (CO – 1.62) / 0.9554 (2a)

Remaining standard uncertainty: u_{CO} (ppb) = sqrt (0.2 ppb^2 + 1.01e-04 * X_{CO}^2) (2b)

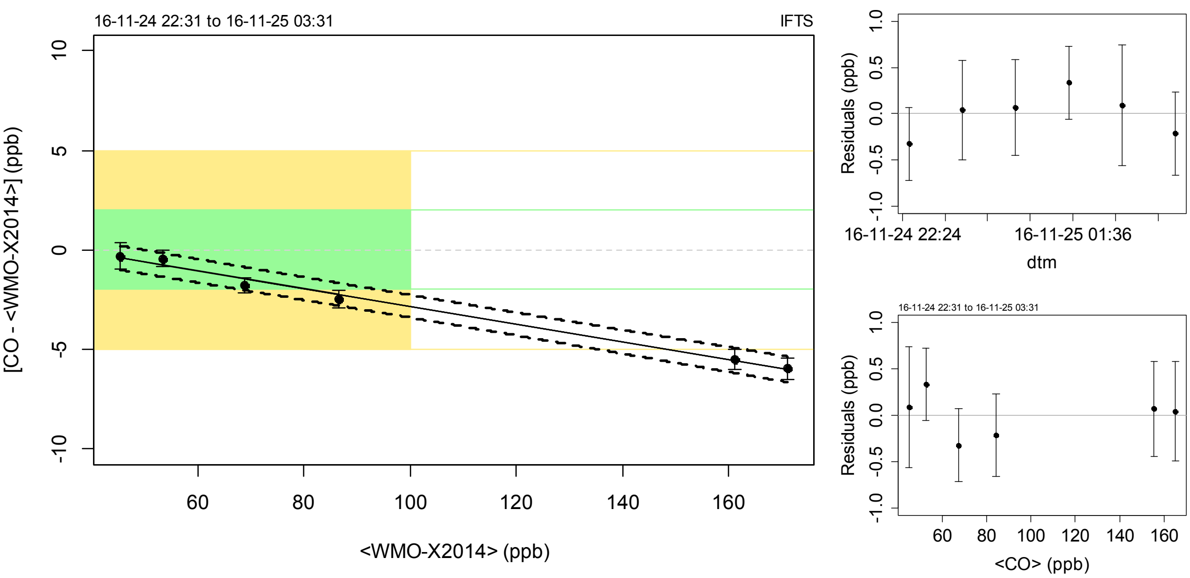

Figure 5. *Left: Bias of the LAU IFTS carbon monoxide instrument with respect to the WMO-X2014A reference scale as a function of mole fraction. Each point represents the average of data at a given level from a specific run. The error bars show the standard deviation of individual measurement points. The green and yellow lines correspond to the WMO compatibility and extended compatibility goals, and the green and yellow areas to the mole fraction range relevant for LAU. The dashed lines around the regression lines are the Working-Hotelling 95% confidence bands. Right: Regression residuals (time dependence and mole fraction dependence).*

The results of the comparisons can be summarised as follows:

The bias is small and within the compatibility goals at low mole fractions up to 80 ppb, which are most relevant for LAU. However, the observed bias at higher levels was exceeding the extended compatibility goal, which cannot be explained by scale differences, since both LAU and WCC-Empa are referring to the WMO-X2014 scale. Most likely, the issue is related to the FTIR measurements, since a variable bias was also observed during the ambient air comparison, which is shown further below.

Recommendation 7 (, important, 2018)**
It is recommended to identify the reason for the observed bias in the CO measurements. Experiments resolving this issue are highly encouraged.

Methane Measurements

CH_4 is also measured with the IFTS system, and continuous time series are available since 2007.

Instrumentation and Standards. Same system as for CO.

Intercomparison (Performance Audit). The comparison involved repeated challenges of the LAU instrument with randomised CH_4 levels from travelling standards. The results of the comparison measurements for the individual measurement parameters are summarised and illustrated below.

The following equation characterises the instrument bias. The result is further illustrated in Figure 6 with respect to the relevant mole fraction range and the WMO/GAW compatibility goals and extended compatibility goals (WMO, 2014).

IFTS analyser:

Unbiased CH_4 mixing ratio: X_{CH4} (ppb) = (CH_4 – 4.1 ppb) / 0.9989 (3a)

Remaining standard uncertainty: u_{CH4} (ppb) = sqrt (1.0 ppb^2 + 1.30e-07 * $X_{CH4}{}^2$) (3b)

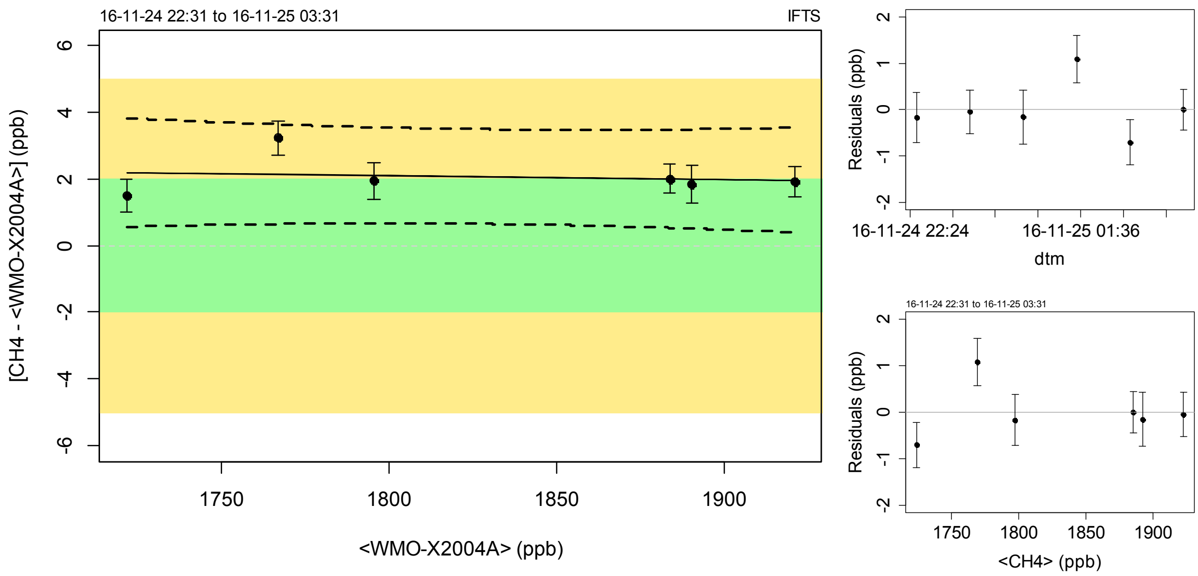

Figure 6. *Left: Bias of the IFTS methane instrument with respect to the WMO-X2004A CH_4 reference scale as a function of mole fraction. Each point represents the average of data at a given level from a specific run. The error bars show the standard deviation of individual measurement points. The green and yellow lines correspond to the WMO compatibility and extended compatibility goals, and the green and yellow areas to the mole fraction range relevant for LAU. The dashed lines around the regression lines are the Working-Hotelling 95% confidence bands. Right: Regression residuals (time dependence and mole fraction dependence).*

The result of the comparison can be summarised as follows:

The bias of the LAU methane instrument showed on average an offset of about 2 ppb, which is still within the extended WMO/GAW compatibility goal over the entire relevant mole fraction range. This confirms that the instrumentation is adequate for CH_4 measurements; however, the reason of the bias, which is most likely due to mole fraction assignments of the methane standards, needs to be identified and corrected. A similar bias was also observed during the ambient air comparison, which is shown further below.

Carbon Dioxide Measurements

CO_2 is also measured with the IFTS system. In addition, a NDIR CO_2 analyser is available since 2008.

Instrumentation. See CO, and in addition a LI-COR 7000 NDIR CO_2 analyser.

Standards. A set of reference standards is used to calibrate the IFTS and also the NDIR analyser. The standards have been calibrated by NIWA against NOAA laboratory standards. In addition, a target tank is available for the NDIR instrument. A list of available standards is given in the Appendix.

Intercomparison (Performance Audit). The comparison involved repeated challenges of the LAU instrument with randomised CO_2 levels from travelling standards. The results of the comparison measurements for the individual measurement parameters are summarised and illustrated below.

The following equation characterises the instrument bias. The result is further illustrated in Figures 7 and 8 with respect to the relevant mole fraction range and the WMO/GAW compatibility goals and extended compatibility goals (WMO, 2014).

IFTS:

Unbiased CO_2 mixing ratio: X_{CO2} (ppm) = (CO_2 + 1.53 ppm) / 1.00416 (4a)

Remaining standard uncertainty: u_{CO2} (ppm) = sqrt (0.009 ppm^2 + 3.28e-08 * X_{CO2}^2) (4b)

NDIR:

Unbiased CO_2 mixing ratio: X_{CO2} (ppm) = (CO_2 – 1.86 ppm) / 0.99435 (4c)

Remaining standard uncertainty: u_{CO2} (ppm) = sqrt (0.037 ppm^2 + 3.28e-08 * X_{CO2}^2) (4d)

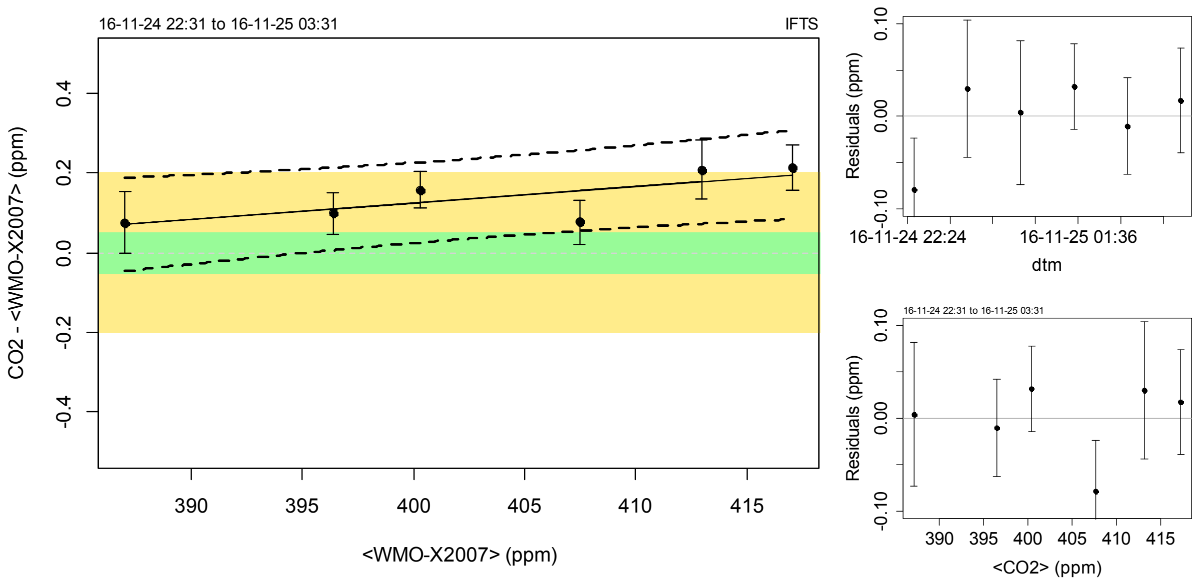

Figure 7. *Left: Bias of the IFTS CO_2 instrument with respect to the WMO-X2007 reference scale as a function of mole fraction. Each point represents the average of data at a given level from a specific run. The error bars show the standard deviation of individual measurement points. The green and yellow lines correspond to the WMO compatibility and extended compatibility goals, and the green and yellow areas to the mole fraction range relevant for LAU. The dashed lines around the regression lines are the Working-Hotelling 95% confidence bands. Right: Regression residuals (time dependence and mole fraction dependence).*

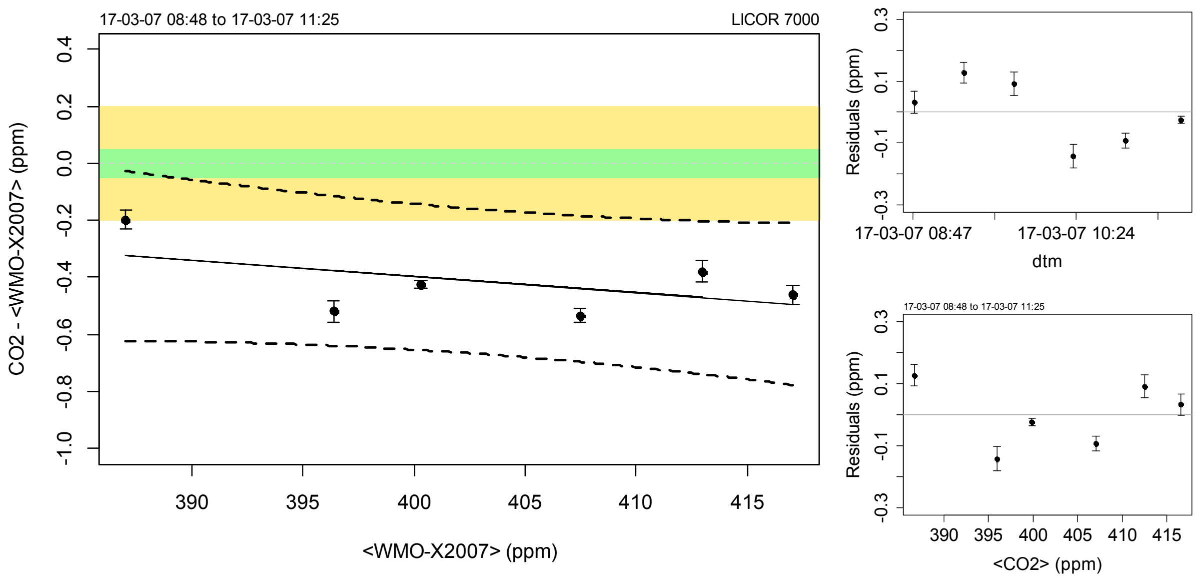

Figure 8. *Same as above for the LAU NDIR CO_2 analyser.*

The result of the comparison can be summarised as follows:

The IFTS showed an offset of about 0.1 ppm CO_2 in the relevant mole fraction range, which is larger than the WMO compatibility goal of 0.05 ppm for the Southern Hemisphere, while the bias of the NDIR instrument was about -0.4 ppm. Similar offsets were also observed during the ambient air comparison, which is shown further below. The results show that both instruments are adequate but need to be further optimised with regard to calibration.

Nitrous Oxide Measurements

N_2O is also measured with the IFTS system, and continuous time series are available since 2007.

Instrumentation and Standards. Same system as for CO.

Intercomparison (Performance Audit). The comparison involved repeated challenges of the LAU instrument with randomised nitrous oxide levels using WCC-Empa travelling standards. The following equations characterise the instrument bias, and the results are further illustrated in Figure 9 with respect to the WMO GAW DQOs (WMO, 2014):

IFTS analyser:

Unbiased N_2O mixing ratio: X_{N2O} (ppb) = (N_2O + 0.07) / 1.0036 (5a)

Remaining standard uncertainty: u_{N2O} (ppb) = sqrt (0.04 ppb^2 + 1.01e-07 * $X_{N2O}{}^2$) (5b)

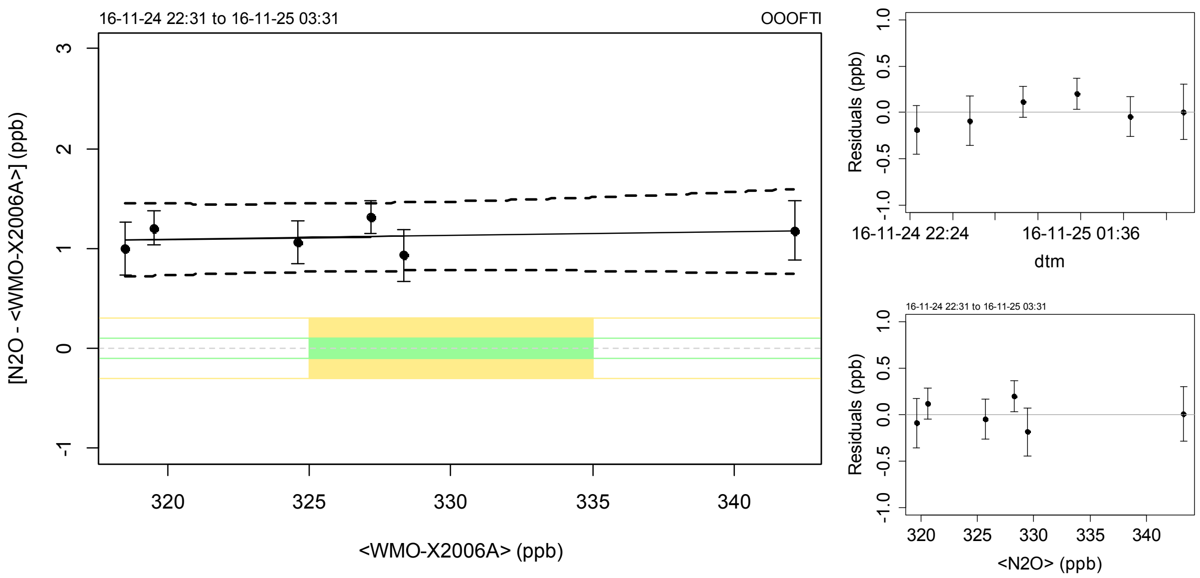

Figure 9. *Left: Bias of the LAU IFTS analyser nitrous oxide instrument with respect to the WMO-X2006A reference scale as a function of mole fraction. Each point represents the average of data at a given level from a specific run. The error bars show the standard deviation of individual measurement points. The green and yellow lines correspond to the WMO compatibility and extended compatibility goals, and the green and yellow areas to the mole fraction range relevant for LAU. The dashed lines around the regression lines are the Working-Hotelling 95% confidence bands. Right: Regression residuals (time dependence and mole fraction dependence).*

The result of the comparison can be summarised as follows:

The LAU instrument showed a bias of approximately 1 ppb compared to the WCC-Empa reference standards. A potential reason could be the fact that the LAU IFTS has been calibrated using a set of standards in synthetic air. A similar difference was also observed when the LAU IFTS instrument was compared with the latest set of four standard tanks available from NIWA, which were calibrated by NOAA. Comparisons made during the last WMO/GAW round robin experiment (https://www.esrl.noaa.gov/gmd/ccgg/wmorr/wmorr_results.php) showed also an offset of about 0.7 ppb for the NIWA analysis.

Recommendation 8 (, important, 2018)**
Standards used for the calibration of the IFTS instrument should be in whole air and have traceability to the latest calibration scale of the CCL for N_2O. The reason of the bias needs to be identified.

LAU PERFORMANCE AUDIT RESULTS COMPARED TO OTHER STATIONS

This section compares the results of the LAU performance audit to other station audits made by WCC-Empa. The method used to relate the results to other audits was developed and described by Zellweger et al. (2016) for CO_2 and CH_4, but is also applicable to other compounds. Basically, the bias at the centre of the relevant mole fraction range is plotted against the slope of the linear regression analysis of the performance audit. The relevant mole fraction ranges are taken from the recommendation of the GGMT-2015 meeting (WMO, 2016) for the greenhouse gases and CO and refer to conditions usually found in unpolluted air masses. For surface ozone the mole fraction range of 0 -100 ppb was chosen, since this covers most of the natural ozone abundance in the troposphere. This results in well-defined bias/slope combinations which are acceptable for meeting the WMO/GAW compatibility goals in a certain mole fraction range. Figure 10 shows the bias vs. the slope of the performance audits made by WCC-Empa for CO, CH_4, CO_2 and N_2O, while the results for O_3 are shown in Figure 11. The grey dots show all comparison results made during WCC-Empa audits for the main station analysers but excludes cases with known instrumental problems. If an adjustment was made during an audit, only the final comparison is shown. The results of the current LAU audit are shown as coloured dots in Figure 10 and 11, and are also summarised in Table 1. The percentages of all WCC-Empa audits fulfilling the DQOs or extended DQOs (eDQOs) are also shown in Table 1.

It can be see that the results were only within the DQOs/eDQOs for O_3 and CH_4, while CO, CO_2 and N_2O were exceeding these criteria. However, it should be noted that this covers the entire mole fraction range in the unpolluted troposphere, and actually encountered values at LAU might be in a much narrower range.

Table 1. LAU performance audit results compared to other stations. The 4th column indicates whether the results of the current audit were within the DQO (green tick mark), extended DQO (orange tick mark) or exceeding the DQOs (red cross), while the 5-7th columns show the percentage of all WCC-Empa audits within these criteria since the year 2005.

Compound	Range	Unit	LAU within DQO/eDQO	% of audits within DQOs	% of audits within eDQOs	% of audits outside eDQOs
CO	30 - 300	ppb	✗	22	22	56
CH_4	1750 - 2100	ppb	✓	61	30	9
CO_2	380 - 450	ppm	✗	33	24	43
N_2O	325 - 335	ppb	✗	0	38	62
O_3	0 -100	ppb	✓	62	NA	38

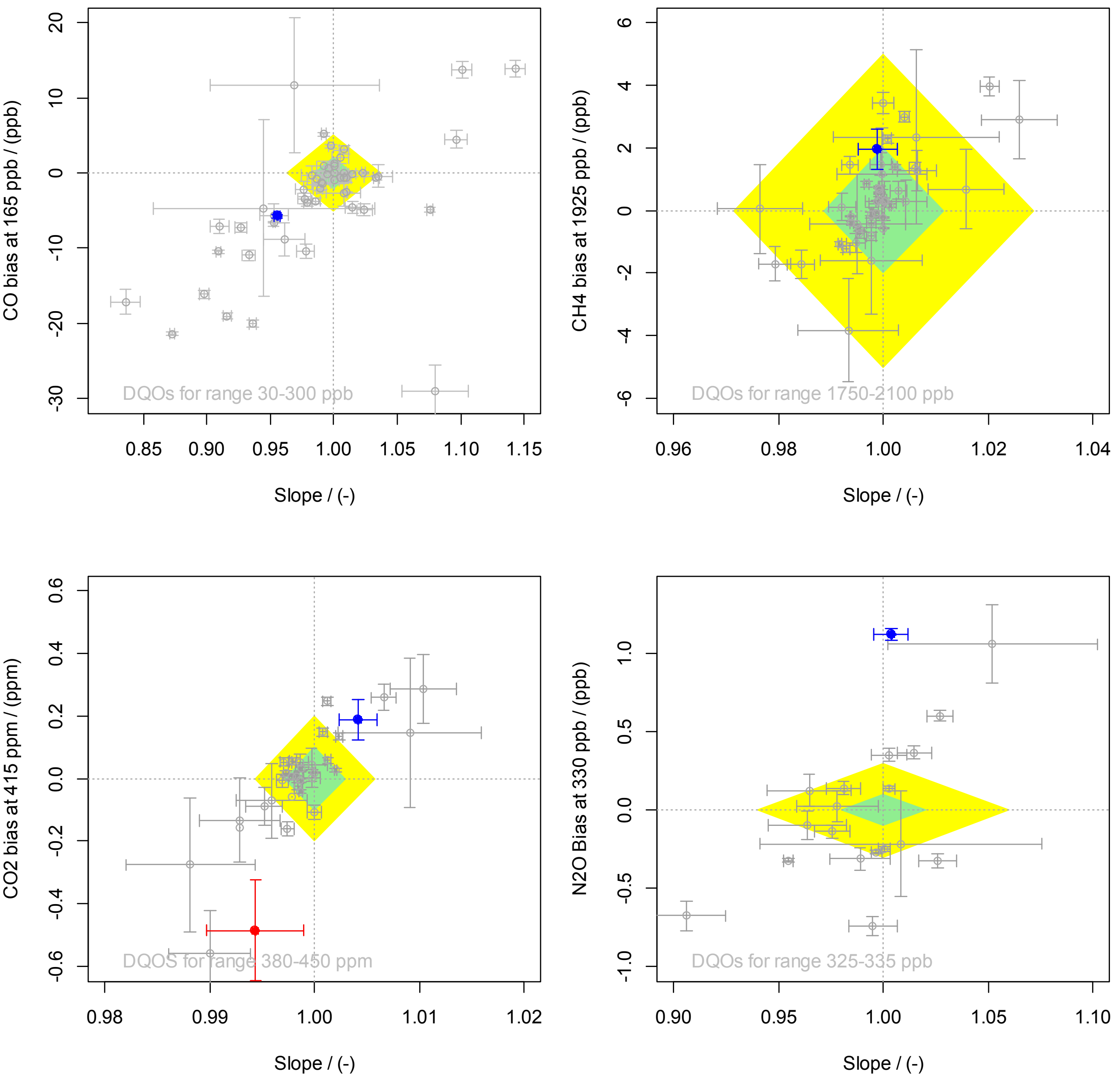

Figure 10. *CO (top left), CH_4 (top right), CO_2 (bottom left) and N_2O (bottom right) bias in the centre of the relevant mole fraction range vs. the slope of the performance audits made by WCC-Empa. The grey dots correspond to past performance audits by WCC-Empa at various stations, while the coloured dots show LAU results (blue: IFTS, red: LICOR 7000). The coloured areas correspond to the WMO/GAW compatibility goals (green) and extended compatibility goals (yellow).*

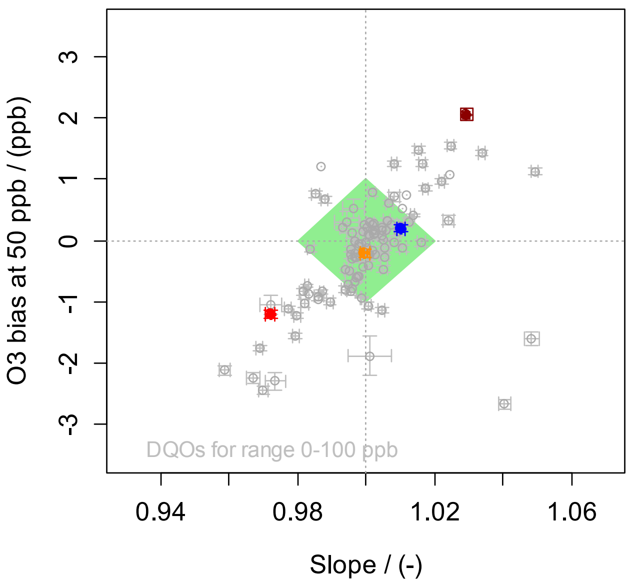

Figure 11. *O_3 bias in the centre of the relevant mole fraction range vs. the slope of the performance audits made by WCC-Empa. The grey dots correspond to past performance audits by WCC-Empa at various stations, while the coloured dots shows the LAU (blue) and BHD (red) results. In addition, the results of the instrument that will replace the BHD analyser are shown (dark-red: initial calibration settings; orange: final settings).The green area corresponds to the WMO/GAW DQO for surface ozone.*

PARALLEL MEASUREMENTS OF AMBIENT AIR

The audit included parallel measurements of CO_2, CH_4 and CO with a WCC-Empa travelling instrument (TI) (Picarro G2401). The TI was running from 22 November 2016 through 17 February 2017; however, data after mid-January could not be used due to a failure of the water measurements of the TI. The TI was also not running in optimal conditions before that period due to drifts in the adjustment of one laser, which resulted in spikes in the CH_4 data. These spikes could be removed by filtering, but the uncertainty is higher than usual. The TI was connected to a spare port of the LAU air inlet system. The TI was sampling air using the following sequence: 1805 min ambient air followed by 30 min measurement of three standard gases (10 min each). To account for the effect of water vapour a correction function (Rella et al., 2013; Zellweger et al., 2012) was applied to the TI data. Details of the calibration of the TI are given in the Appendix. The results of the ambient air comparison are presented below.

Figure 12 shows the comparison of hourly CO, CH_4 and CO_2 data of the LAU IFTS instrument with the TI. In addition, flask measurements of LAU, which were analysed by NIWA, are shown as green dots. In 2018, the working standard on the flask CO_2 GC was revised, and the flask CO_2 mole fractions were updated, which is shown as dark green dots. Hourly averages are shown. The corresponding deviation histograms are shown in Figure 13. The same plots for the LAU NDIR instrument are shown in Figure 14 and 15.

Discussion of the ambient air comparison results

Carbon Monoxide:

The median CO bias of the LAU IFTS instrument was -2.3 ppb compared to WCC-Empa TI, which is slightly larger compared to the performance audit results. However, it should be noted that bias was not constant over time. Several distinctly different periods were observed, with a small, a positive or a negative bias. From these two independent time series, it is not clear which is representing the true value. Agreement with flask measurements was at the beginning of the period better compared with the IFTS instrument, but it changed to better agreement with the TI towards the end of the period. This indicates that either the TI or the IFTS changed sensitivity over time. There is no indication of a drift in the working standard measurements of the TS, but a small dependency on ambient water concentration was found. However, this does not sufficiently explain the step changes that were observed.

Methane:

A median bias of +3.6 ppb was observed for the IFTS in comparison to the TI. This compares relatively well with the results of the performance audit with an observed bias slightly larger than 2 ppb. The results of the LAU flask measurement are also in good agreement with the in-situ IFTS data, which confirms internal consistency of the LAU system. The remaining difference might be attributable to calibration standards.

Carbon dioxide:

A median bias of +0.22 ppm was observed for the IFTS in comparison to the TI, while a negative bias of -0.52 ppm was found for the NDIR instrument. Both results compares well with the results of the performance audit, which indicates consistency of the comparison experiment. The remaining difference might be attributable to calibration standards or other factors influencing the calibration such as linearity etc. In contrast to CH_4, the CO_2 flask measurements made at LAU initially showed a larger negative bias compared to all available in-situ instruments. A revision of the working standard on the flask CO_2 GC in 2018 shifted values up, which resulted in better agreement with the LAU measurements.

In general, the temporal variations of ambient air mole fractions were well captured with all instruments. The scatter in the bias can be explained by different temporal coverage and response times of the instruments. The ambient air comparison showed that both the IFTS and the NDIR instruments are suitable, but the IFTS gave slightly better results for CO_2. An issue with CO (step changes the IFTS or the TI) could not be solved and needs further attention.

Recommendation 9 (, important, 2018)**
Open issues from the ambient air comparison should be followed up by further experiments.

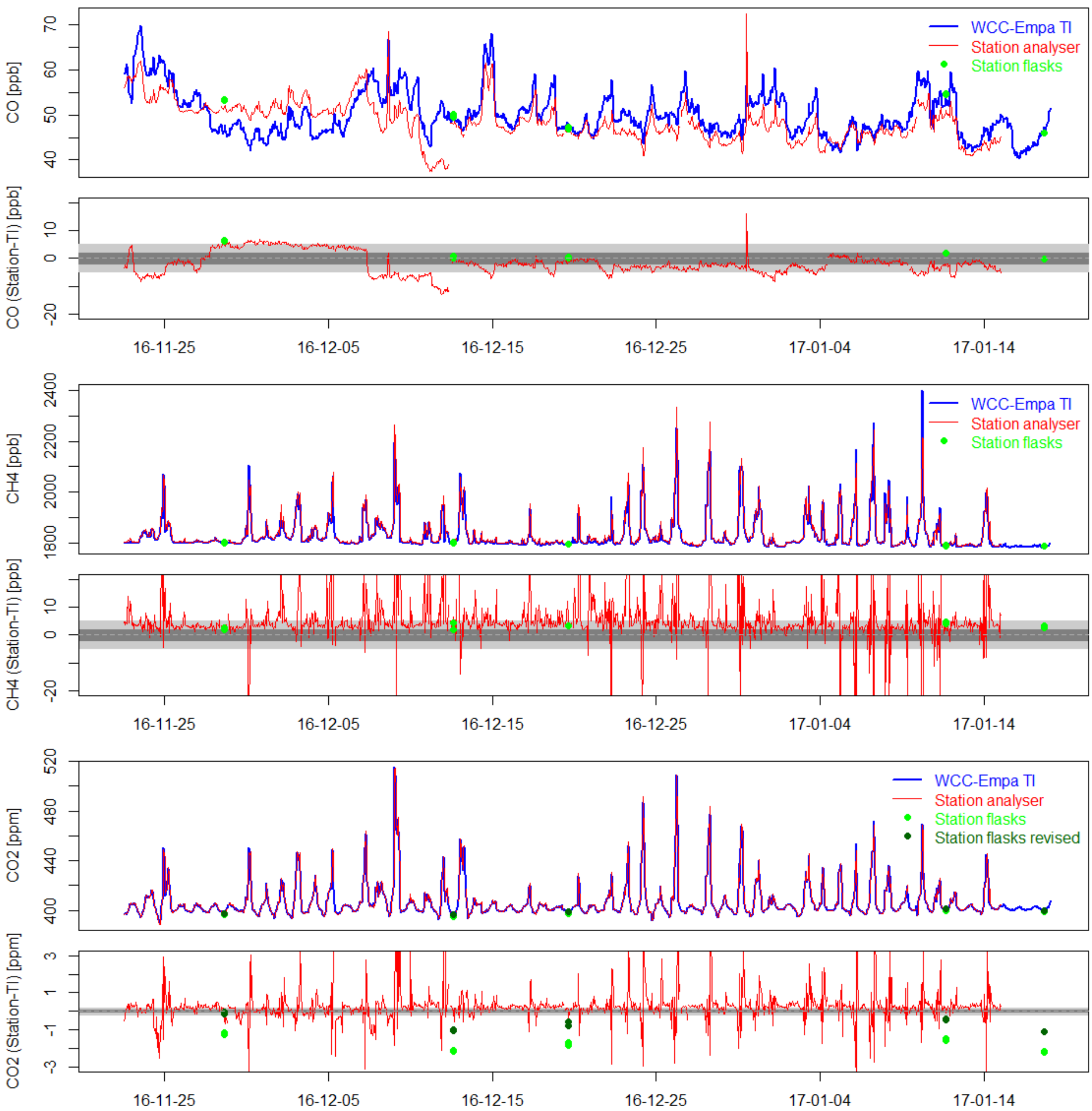

Figure 12. *Comparison of the LAU IFTS analyser with the WCC-Empa travelling instrument for CO (top), CH_4 (middle) and CO_2 (bottom). LAU flask measurements are shown as green dots. Time series based on hourly data as well as the difference between the IFTS and the TI is shown. The horizontal grey areas correspond to the WMO/GAW compatibility (dark grey) and extended compatibility (light grey) goals.*

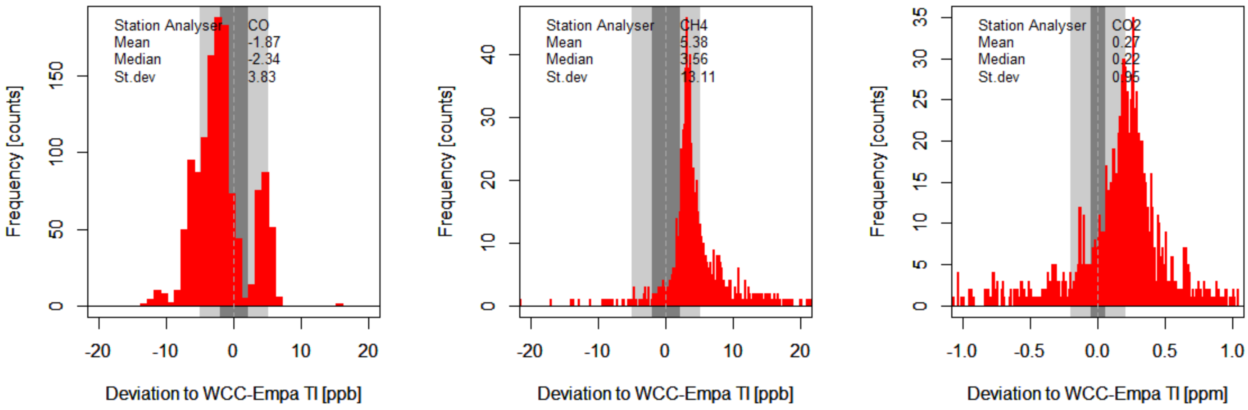

Figure 13. Deviation histograms for the LAU IFTS analyser for CO (left), CH_4 (middle) and CO_2 (right). 1 h data is shown for the station analyser – TI.

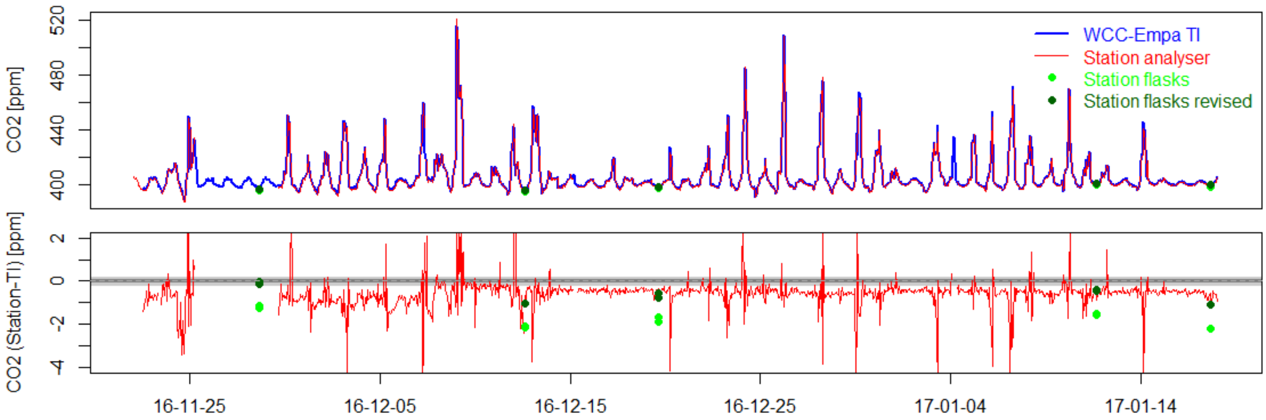

Figure 14. *Comparison of the LAU NDIR CO_2 analyser with the WCC-Empa travelling instrument. Time series based on hourly data as well as the difference between the IFTS and the TI is shown. The horizontal grey areas correspond to the WMO/GAW compatibility (dark grey) and extended compatibility (light grey) goals.*

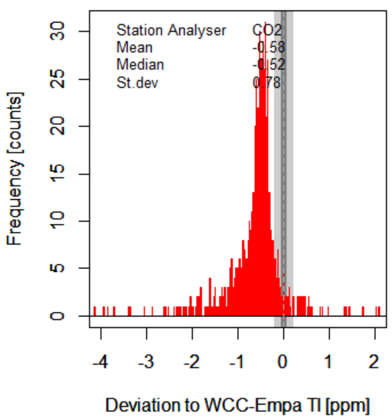

Figure 15. *Deviation histograms for the LAU CO_2 NDIR analyser. 1 h data is shown for the station analyser – TI.*

CONCLUSIONS

The global GAW station Lauder provides extensive research facilities and hosts a large number of long-term continuous observations in all WMO/GAW focal areas as well as research projects, which makes it a very significant contribution to the GAW programme.

Most assessed measurements were of high data quality and met the WMO/GAW extended compatibility goals in the relevant mole fraction range. Table 2 summarises the results of the performance audit and the ambient air comparison with respect to the WMO/GAW compatibility goals. Please note that Table 2 refers only to the mole fractions relevant to LAU, whereas Table 1 further above covers a wider mole fraction range.

Table 2. Synthesis of the performance audit and ambient air comparison results. A tick mark indicates that the compatibility goal (green) or extended compatibility goal (orange) was met on average. Tick marks in parenthesis mean that the goal was only partly reached in the relevant mole fraction range (performance audit only), and ✗ indicates results outside the compatibility goals.

Comparison type	O_3 LAU	O_3 BHD	CO IFTS	CH_4 IFTS	CO_2 IFTS	CO_2 NDIR	N_2O IFTS
Performance audit with TS	✓	(✓)	(✓)	✓	✓	✓	✓
Ambient air comparison	NA	NA	✓	✓	✗	✗	NA

NA no ambient air comparison was made for ozone and nitrous oxide

The continuation of the Lauder measurement series is highly important for GAW. The large number of measured atmospheric constituents in combination with the high data quality enables state of the art research projects.

SUMMARY RANKING OF THE LAUDER GAW STATION

System Audit Aspect	Adequacy#	Comment
Measurement programme	(5)	Comprehensive programme.
Access	(5)	Year round access by road.
Facilities		
Laboratory and office space	(5)	Adequate, with space for additional research campaigns.
Internet access	(5)	Sufficient bandwidth
Air Conditioning	(5)	Fully adequate system
Power supply	(5)	Reliable with very few power cuts
General Management and Operation		
Organisation	(5)	Well-coordinated
Competence of staff	(5)	Skilled staff
Air Inlet System	(5)	Adequate systems
Instrumentation		
Ozone	(5)	Adequate instrumentation
$CO/CH_4/CO_2/N_2O$ (IFTS)	(4)	Adequate but requires highly skilled personnel
CO_2 (NDIR)	(3)	Adequate but performance issues during the period of the audit
Standards		
Ozone	(1)	No station standard available NOAA calibration infrequent
CO, CO_2, CH_4, N_2O	(5)	NOAA traceable standards and / or working standards available
Data Management		
Data acquisition	(4)	Fully adequate system except for ozone with manual data download
Data processing	(5)	Highly skilled staff
Data submission	(2)	Only few data (O_3, CH_4) submitted. CH_4 incomplete; only filtered data.

#0: inadequate thru 5: adequate.

Dübendorf, May 2018

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WCC-Empa

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APPENDIX

Data Review

The following figures show summary plots of LAU data accessed on 23 November 2017 from WDCGG; however, only CH_4 and O_3 were available. The plots below show time series of hourly data, frequency distribution, as well as diurnal and seasonal variations.

The main findings of the data review can be summarised as follows:

Methane:

- Data set looks sound but only one hour per day (which normally represents clean conditions) has been submitted.
- The difference between the data with wind speed > 5 m/s, which is considered to be representative for well mixed air, and the other data is small.

Ozone:

- Data looks fully plausible with regard to all aspects (mole fraction range, diurnal and seasonal variation).

Carbon monoxide, carbon dioxide and nitrous oxide:

- Data has not been submitted, and therefore no review is possible.

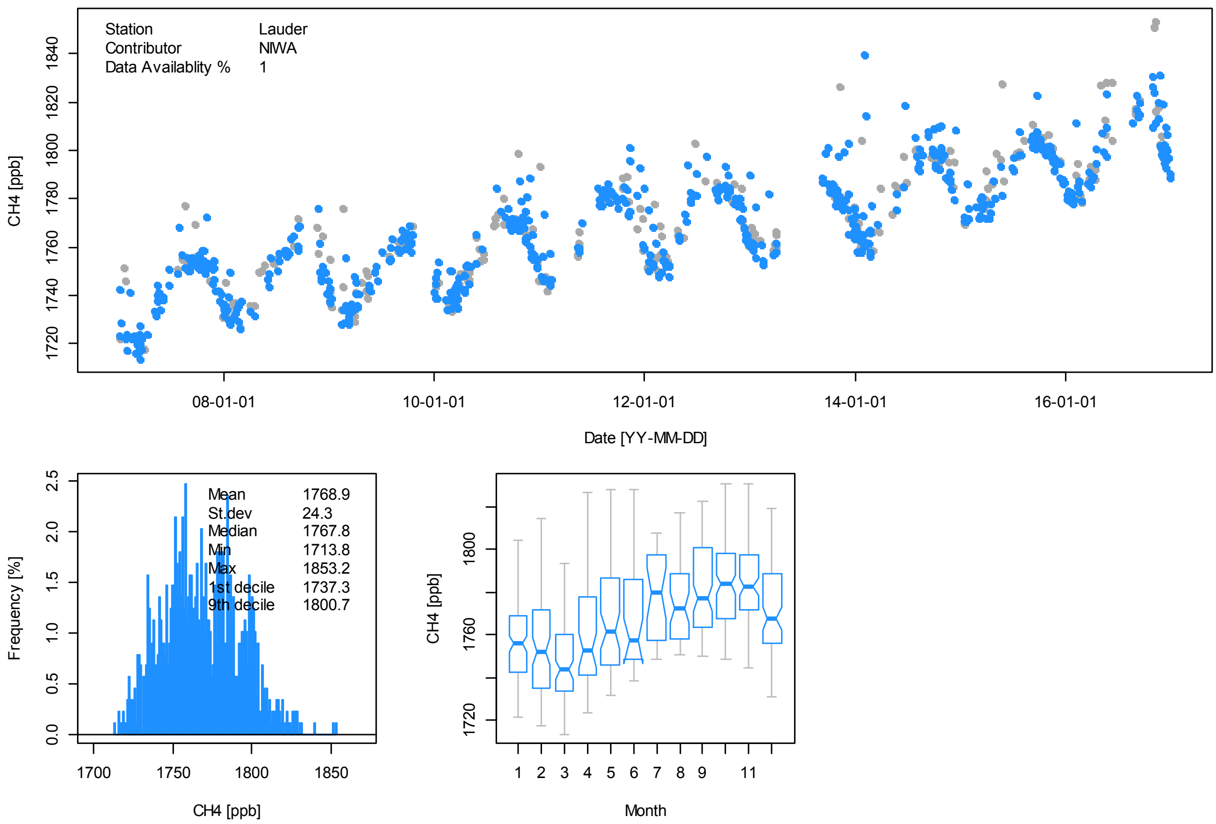

Figure 16. *CH_4 data accessed from WDCGG. Top: Time series, hourly average for 3-4 pm local time. Blue dots are for wind speed >5 m/s, which is considered to be representative of well-mixed tropospheric air. Bottom: Left: Frequency distribution. Right: Seasonal variation; the horizontal blue line denotes to the median, and the blue boxes show the inter-quartile range.*

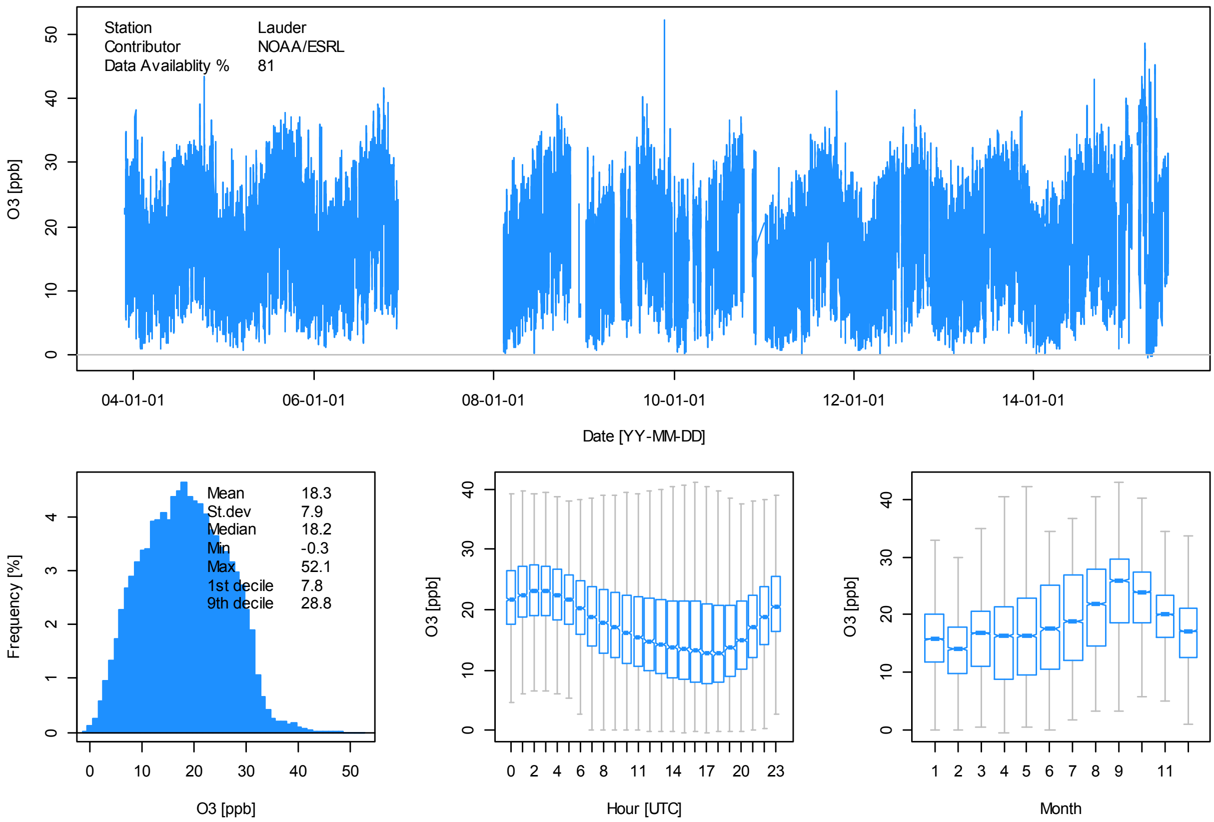

Figure 17. *Same as above for O_3, middle lower panel diurnal variation.*

Surface Ozone Comparisons

All procedures were conducted according to the Standard Operating Procedure (WCC-Empa SOP) and included comparisons of the travelling standard with the Standard Reference Photometer at Empa before and after the comparison of the analyser.

The internal ozone generator of the WCC-Empa transfer standard was used for generation of a randomised sequence of ozone levels ranging from 0 to 200 ppb. Zero air was generated using a custom built zero air generator (Nafion drier, activated charcoal, Purafil). The TS was connected to the station analyser using approx. 1.5 m of PFA tubing. Table 3 details the experimental setup during the comparisons of the travelling standard with the station analysers. The data used for the evaluation was recorded by the WCC-Empa data acquisition system.

Table 3. Experimental details of the ozone comparison.

Travelling standard (TS)	
Model, S/N	Thermo Scientific 49C-PS #0421507340 (WCC-Empa)
Settings	BKG -0.8, COEF 1.014
Pressure readings (hPa)	Ambient 972.2 TS 976.2, adjusted to 971.9 before comparison
LAU Station analyser (OA)	
Model, S/N	Thermo Scientific 49C #0326101959
Principle	UV absorption
Range	0-1 ppm
Settings	BKG 0.0 ppb, COEF 1.000 A correction based on a calibration with a NOAA travelling standard is applied: Final data = 1.029 * raw data + 0.16 ppb
Pressure readings (hPa)	Ambient 969.0; OA 967.7 (no adjustment was made)
BHD Station analyser (OA)	
Model, S/N	Thermo Scientific 49i #0532213220
Principle	UV absorption
Range	0-1 ppm
Settings	BKG 0.0 ppb, COEF 1.000
Pressure readings (hPa)	Ambient 972.2; OA 971.7 (no adjustment was made)
Future BHD Station analyser (OA)	
Model, S/N	Thermo Scientific 49i #01152220033
Principle	UV absorption
Range	0-1 ppm
Settings	Initial: BKG -0.2 ppb, COEF 1.027 Final: BKG -0.1 ppb, COEF 0.997
Pressure readings (hPa)	Ambient 972.2; OA 971.9 (no adjustment was made)

Results

Each ozone level was applied for 10 minutes, and the last 5 one-minute averages were aggregated. These aggregates were used in the assessment of the comparison. All results are valid for the calibration factors as given in Table 3 above. The readings of the travelling standard (TS) were compensated for bias with respect to the Standard Reference Photometer (SRP) prior to the evaluation of the ozone analyser (OA) values.
The results of the assessment is shown in the following Tables (individual measurement points) and further presented in the Executive Summary.

Table 4. Ten-minute aggregates computed from the last 5 of a total of 10 one-minute values for the comparison of the LAU ozone analyser (OA) Thermo Scientific 49C #0326101959 with the WCC-Empa travelling standard (TS).

Date - Time	Run #	Level (ppb)	TS (ppb)	OA (ppb)	sdTS (ppb)	sdOA (ppb)	OC-TS (ppb)	OC-TS (%)
2016-11-22 02:52	1	0	-0.20	0.15	0.11	0.10	0.35	-NA
2016-11-22 03:02	1	30	30.24	29.24	0.45	0.17	-1.00	-3.3
2016-11-22 03:12	1	60	60.16	58.55	0.08	0.08	-1.61	-2.7
2016-11-22 03:22	1	90	90.11	87.84	0.18	0.09	-2.27	-2.5
2016-11-22 03:32	1	20	20.71	19.93	0.43	0.34	-0.78	-3.8
2016-11-22 03:42	1	80	80.27	77.69	0.37	0.44	-2.58	-3.2
2016-11-22 03:52	1	50	50.25	48.88	0.24	0.21	-1.37	-2.7
2016-11-22 04:02	1	70	70.06	68.05	0.09	0.20	-2.01	-2.9
2016-11-22 04:12	1	10	12.04	11.67	0.47	0.37	-0.37	-3.1
2016-11-22 04:22	1	40	40.00	39.00	0.25	0.13	-1.00	-2.5
2016-11-22 04:32	2	0	-0.16	0.01	0.13	0.11	0.17	NA
2016-11-22 04:42	2	90	90.15	87.65	0.16	0.16	-2.50	-2.8
2016-11-22 04:52	2	10	11.85	11.58	0.56	0.34	-0.27	-2.3
2016-11-22 05:02	2	60	60.06	58.43	0.23	0.08	-1.63	-2.7
2016-11-22 05:12	2	80	80.01	78.14	0.13	0.18	-1.87	-2.3
2016-11-22 05:22	2	20	20.50	19.94	0.29	0.26	-0.56	-2.7
2016-11-22 05:32	2	30	29.83	29.05	0.21	0.13	-0.78	-2.6
2016-11-22 05:42	2	50	50.01	48.47	0.11	0.16	-1.54	-3.1
2016-11-22 05:52	2	40	40.02	38.86	0.23	0.19	-1.16	-2.9
2016-11-22 06:02	2	70	69.99	68.39	0.15	0.13	-1.60	-2.3
2016-11-22 06:12	3	0	-0.17	-0.03	0.07	0.17	0.14	NA
2016-11-22 06:22	3	90	90.08	87.71	0.13	0.19	-2.37	-2.6
2016-11-22 06:32	3	30	29.91	29.16	0.23	0.17	-0.75	-2.5
2016-11-22 06:42	3	10	11.38	10.89	0.38	0.33	-0.49	-4.3
2016-11-22 06:52	3	40	39.66	38.55	0.19	0.34	-1.11	-2.8
2016-11-22 07:02	3	50	49.98	48.64	0.18	0.18	-1.34	-2.7
2016-11-22 07:12	3	20	20.43	19.83	0.18	0.11	-0.60	-2.9
2016-11-22 07:22	3	70	70.01	68.07	0.19	0.21	-1.94	-2.8
2016-11-22 07:32	3	80	80.04	78.21	0.21	0.17	-1.83	-2.3
2016-11-22 07:42	3	60	60.07	58.63	0.15	0.18	-1.44	-2.4

Table 5. Ten-minute aggregates computed from the last 5 of a total of 10 one-minute values for the comparison of the BHD ozone analyser (OA) Thermo Scientific 49i #0532213220 with the WCC-Empa travelling standard (TS).

Date - Time	Run #	Level (ppb)	TS (ppb)	OA (ppb)	sdTS (ppb)	sdOA (ppb)	OA-TS (ppb)	OA-TS (%)
2016-11-22 02:52	1	0	-0.20	0.15	0.11	0.10	0.35	-NA
2016-11-22 03:02	1	30	30.24	29.24	0.45	0.17	-1.00	-3.3
2016-11-22 03:12	1	60	60.16	58.55	0.08	0.08	-1.61	-2.7
2016-11-22 03:22	1	90	90.11	87.84	0.18	0.09	-2.27	-2.5
2016-11-22 03:32	1	20	20.71	19.93	0.43	0.34	-0.78	-3.8
2016-11-22 03:42	1	80	80.27	77.69	0.37	0.44	-2.58	-3.2
2016-11-22 03:52	1	50	50.25	48.88	0.24	0.21	-1.37	-2.7
2016-11-22 04:02	1	70	70.06	68.05	0.09	0.20	-2.01	-2.9
2016-11-22 04:12	1	10	12.04	11.67	0.47	0.37	-0.37	-3.1
2016-11-22 04:22	1	40	40.00	39.00	0.25	0.13	-1.00	-2.5
2016-11-22 04:32	2	0	-0.16	0.01	0.13	0.11	0.17	NA
2016-11-22 04:42	2	90	90.15	87.65	0.16	0.16	-2.50	-2.8
2016-11-22 04:52	2	10	11.85	11.58	0.56	0.34	-0.27	-2.3
2016-11-22 05:02	2	60	60.06	58.43	0.23	0.08	-1.63	-2.7
2016-11-22 05:12	2	80	80.01	78.14	0.13	0.18	-1.87	-2.3
2016-11-22 05:22	2	20	20.50	19.94	0.29	0.26	-0.56	-2.7
2016-11-22 05:32	2	30	29.83	29.05	0.21	0.13	-0.78	-2.6
2016-11-22 05:42	2	50	50.01	48.47	0.11	0.16	-1.54	-3.1
2016-11-22 05:52	2	40	40.02	38.86	0.23	0.19	-1.16	-2.9
2016-11-22 06:02	2	70	69.99	68.39	0.15	0.13	-1.60	-2.3
2016-11-22 06:12	3	0	-0.17	-0.03	0.07	0.17	0.14	NA
2016-11-22 06:22	3	90	90.08	87.71	0.13	0.19	-2.37	-2.6
2016-11-22 06:32	3	30	29.91	29.16	0.23	0.17	-0.75	-2.5
2016-11-22 06:42	3	10	11.38	10.89	0.38	0.33	-0.49	-4.3
2016-11-22 06:52	3	40	39.66	38.55	0.19	0.34	-1.11	-2.8
2016-11-22 07:02	3	50	49.98	48.64	0.18	0.18	-1.34	-2.7
2016-11-22 07:12	3	20	20.43	19.83	0.18	0.11	-0.60	-2.9
2016-11-22 07:22	3	70	70.01	68.07	0.19	0.21	-1.94	-2.8
2016-11-22 07:32	3	80	80.04	78.21	0.21	0.17	-1.83	-2.3
2016-11-22 07:42	3	60	60.07	58.63	0.15	0.18	-1.44	-2.4

Table 6. Ten-minute aggregates computed from the last 5 of a total of 10 one-minute values for the comparison of the future BHD ozone analyser (OA) (BKG -0.2 ppb, COEF 1.027) Thermo Scientific 49i #01152220033 with the WCC-Empa travelling standard (TS).

Date - Time	Run #	Level (ppb)	TS (ppb)	OA (ppb)	sdTS (ppb)	sdOA (ppb)	OA-TS (ppb)	OA-TS (%)
2016-11-22 02:52	1	0	-0.20	0.15	0.11	0.10	0.35	-NA
2016-11-22 03:02	1	30	30.24	29.24	0.45	0.17	-1.00	-3.3
2016-11-22 03:12	1	60	60.16	58.55	0.08	0.08	-1.61	-2.7
2016-11-22 03:22	1	90	90.11	87.84	0.18	0.09	-2.27	-2.5
2016-11-22 03:32	1	20	20.71	19.93	0.43	0.34	-0.78	-3.8
2016-11-22 03:42	1	80	80.27	77.69	0.37	0.44	-2.58	-3.2
2016-11-22 03:52	1	50	50.25	48.88	0.24	0.21	-1.37	-2.7

Date - Time	Run #	Level (ppb)	TS (ppb)	OA (ppb)	sdTS (ppb)	sdOA (ppb)	OA-TS (ppb)	OA-TS (%)
2016-11-22 04:02	1	70	70.06	68.05	0.09	0.20	-2.01	-2.9
2016-11-22 04:12	1	10	12.04	11.67	0.47	0.37	-0.37	-3.1
2016-11-22 04:22	1	40	40.00	39.00	0.25	0.13	-1.00	-2.5
2016-11-22 04:32	2	0	-0.16	0.01	0.13	0.11	0.17	NA
2016-11-22 04:42	2	90	90.15	87.65	0.16	0.16	-2.50	-2.8
2016-11-22 04:52	2	10	11.85	11.58	0.56	0.34	-0.27	-2.3
2016-11-22 05:02	2	60	60.06	58.43	0.23	0.08	-1.63	-2.7
2016-11-22 05:12	2	80	80.01	78.14	0.13	0.18	-1.87	-2.3
2016-11-22 05:22	2	20	20.50	19.94	0.29	0.26	-0.56	-2.7
2016-11-22 05:32	2	30	29.83	29.05	0.21	0.13	-0.78	-2.6
2016-11-22 05:42	2	50	50.01	48.47	0.11	0.16	-1.54	-3.1
2016-11-22 05:52	2	40	40.02	38.86	0.23	0.19	-1.16	-2.9
2016-11-22 06:02	2	70	69.99	68.39	0.15	0.13	-1.60	-2.3
2016-11-22 06:12	3	0	-0.17	-0.03	0.07	0.17	0.14	NA
2016-11-22 06:22	3	90	90.08	87.71	0.13	0.19	-2.37	-2.6
2016-11-22 06:32	3	30	29.91	29.16	0.23	0.17	-0.75	-2.5
2016-11-22 06:42	3	10	11.38	10.89	0.38	0.33	-0.49	-4.3
2016-11-22 06:52	3	40	39.66	38.55	0.19	0.34	-1.11	-2.8
2016-11-22 07:02	3	50	49.98	48.64	0.18	0.18	-1.34	-2.7
2016-11-22 07:12	3	20	20.43	19.83	0.18	0.11	-0.60	-2.9
2016-11-22 07:22	3	70	70.01	68.07	0.19	0.21	-1.94	-2.8
2016-11-22 07:32	3	80	80.04	78.21	0.21	0.17	-1.83	-2.3
2016-11-22 07:42	3	60	60.07	58.63	0.15	0.18	-1.44	-2.4

Table 7. Ten-minute aggregates computed from the last 5 of a total of 10 one-minute values for the comparison of the future BHD ozone analyser (OA) (BKG -0.1 ppb, COEF 0.997) Thermo Scientific 49i #01152220033 with the WCC-Empa travelling standard (TS).

Date - Time	Run #	Level (ppb)	TS (ppb)	OA (ppb)	sdTS (ppb)	sdOA (ppb)	OA-TS (ppb)	OA-TS (%)
2016-11-23 00:10	1	0	0.38	0.12	0.22	0.23	-0.26	NA
2016-11-23 00:20	1	30	29.98	29.72	0.10	0.14	-0.26	-0.9
2016-11-23 00:30	1	60	60.07	59.80	0.12	0.18	-0.27	-0.4
2016-11-23 00:40	1	90	90.16	89.84	0.14	0.04	-0.32	-0.4
2016-11-23 00:50	1	20	20.41	20.24	0.41	0.32	-0.17	-0.8
2016-11-23 01:00	1	80	80.03	79.91	0.15	0.30	-0.12	-0.1
2016-11-23 01:10	1	50	50.03	49.90	0.10	0.26	-0.13	-0.3
2016-11-23 01:20	1	70	69.90	70.01	0.10	0.43	0.11	0.2
2016-11-23 01:30	1	10	11.70	11.64	0.85	0.72	-0.06	-0.5
2016-11-23 01:40	1	40	39.76	39.45	0.34	0.43	-0.31	-0.8
2016-11-23 01:50	2	0	0.39	0.19	0.12	0.18	-0.20	NA
2016-11-23 02:00	2	90	90.02	89.62	0.08	0.30	-0.40	-0.4
2016-11-23 02:10	2	10	11.91	11.56	0.52	0.55	-0.35	-2.9
2016-11-23 02:20	2	60	59.95	59.64	0.17	0.12	-0.31	-0.5
2016-11-23 02:30	2	80	79.93	79.57	0.13	0.29	-0.36	-0.5
2016-11-23 02:40	2	20	20.45	20.23	0.32	0.17	-0.22	-1.1
2016-11-23 02:50	2	30	29.62	29.27	0.49	0.40	-0.35	-1.2

Date - Time	Run #	Level (ppb)	TS (ppb)	OA (ppb)	sdTS (ppb)	sdOA (ppb)	OA-TS (ppb)	OA-TS (%)
2016-11-23 03:00	2	50	50.02	49.76	0.13	0.28	-0.26	-0.5
2016-11-23 03:10	2	40	39.51	39.34	0.41	0.20	-0.17	-0.4
2016-11-23 03:20	2	70	70.07	69.70	0.20	0.19	-0.37	-0.5
2016-11-23 03:30	3	0	0.36	0.13	0.10	0.14	-0.23	NA
2016-11-23 03:40	3	90	90.00	89.66	0.16	0.29	-0.34	-0.4
2016-11-23 03:50	3	30	30.18	29.68	0.17	0.27	-0.50	-1.7
2016-11-23 04:00	3	10	11.68	11.42	0.47	0.36	-0.26	-2.2
2016-11-23 04:10	3	40	39.88	39.35	0.18	0.46	-0.53	-1.3
2016-11-23 04:20	3	50	50.08	49.87	0.23	0.10	-0.21	-0.4
2016-11-23 04:30	3	20	20.57	20.27	0.17	0.39	-0.30	-1.5
2016-11-23 04:40	3	70	70.15	69.71	0.09	0.13	-0.44	-0.6
2016-11-23 04:50	3	80	80.04	79.56	0.18	0.28	-0.48	-0.6
2016-11-23 05:00	3	60	59.99	59.82	0.10	0.10	-0.17	-0.3
2016-11-23 06:00	4	0	0.49	0.20	0.11	0.10	-0.29	NA
2016-11-23 06:20	4	70	69.98	69.79	0.18	0.28	-0.19	-0.3
2016-11-23 06:20	4	90	90.05	89.84	0.11	0.15	-0.21	-0.2
2016-11-23 06:25	4	60	60.01	59.61	0.13	0.15	-0.40	-0.7
2016-11-23 06:35	4	30	29.89	29.44	0.36	0.40	-0.45	-1.5
2016-11-23 06:45	4	20	20.70	20.31	0.19	0.29	-0.39	-1.9
2016-11-23 06:45	4	80	80.00	79.64	0.20	0.36	-0.36	-0.4
2016-11-23 06:50	4	10	11.91	11.65	0.52	0.43	-0.26	-2.2
2016-11-23 07:05	4	50	49.93	49.66	0.14	0.23	-0.27	-0.5
2016-11-23 07:25	4	40	39.79	39.48	0.29	0.18	-0.31	-0.8

Carbon Monoxide Comparisons

All procedures were conducted according to the Standard Operating Procedure (WMO, 2007) and included comparisons of the travelling standards at Empa before the comparison of the analysers. Details of the traceability of the travelling standards to the WMO/GAW Reference Standard at NOAA/ESRL are given in the appendix.

Table **8** shows details of the experimental setup during the comparison of the transfer standard and the station analysers. The data used for the evaluation was recorded by the LAU data acquisition system. The standards used for the calibration of the LAU analyser are shown in Table 9.

Table 8. Experimental details of LAU CO comparison.

Travelling standard (TS)	
WCC-Empa Travelling standards (6 l aluminium cylinder containing a mixture of natural and synthetic air), assigned values and standard uncertainties see Table 17.	
Station Analyser LAU (AL)	
Model, S/N	IFTS instrument, originally prototype analyser, software and hardware modified/upgraded in 2013 to bring it into line with the 2013 commercial version of the instrument manufactured by Ecotech.
Principle	FTIR
Drying system	FTS internal Nafion dryer operated in reflux mode in series and followed by chemical drying ($Mg(ClO_4)_2$)
Comparison procedures	
Connection	The TS were connected to spare calibration gas ports. All measurements were done in flow mode (dynamic).

Table 9 Reference standards available at LAU. Calibration scales: CH4-WMOX2004A, N2O-NOAA2006A, CO-WMOX2014A, CO2-WMOX2007.

Cylinder ID	CH_4 (ppb)		N_2O (ppb)		CO (ppb)		CO_2 (ppb)	
REF13171	1733.24	0.13	339.02	0.11	95.90	0.13	412.69	0.01
REF13172	1687.32	0.27	320.08	0.10	131.01	0.03	398.51	0.03
REF13173	1742.82	0.22	307.38	0.13	51.32	0.28	457.68	0.06
REF13174	2019.30	0.13	338.94	0.15	107.77	0.20	380.42	0.01

Results

The results of the assessment are shown in the Executive Summary, and the individual measurements of the TS are presented in the following Tables.

Table 10. CO aggregates computed from single analysis (mean and standard deviation of mean) for each level during the comparison of the IFTS instrument (AL) with the WCC-Empa TS (WMO-X2014A CO scale).

Date / Time	TS Cylinder	TS (ppb)	sdTS (ppb)	AL (ppb)	sdAL (ppb)	N	AL-TS (ppb)	AL-TS (%)
(16-11-24 22:31:24)	160922_FB03930	68.8	0.1	67.0	0.4	10	-1.8	-2.6
(16-11-24 23:31:30)	160825_FB03365	171.1	0.2	165.2	0.5	10	-6.0	-3.5
(16-11-25 00:31:24)	130819_FB03855	161.1	0.2	155.6	0.5	10	-5.5	-3.4
(16-11-25 01:31:30)	160922_FF30491	53.4	0.3	53.0	0.4	10	-0.4	-0.8
(16-11-25 02:31:36)	160922_FA02785	45.5	0.1	45.2	0.7	10	-0.3	-0.7
(16-11-25 03:31:36)	160926_FB03346	86.6	0.1	84.1	0.5	10	-2.5	-2.8

Methane Comparisons

All procedures were conducted according to the Standard Operating Procedure (WMO, 2007) and included comparisons of the travelling standards at Empa before the comparison of the analysers. Details of the traceability of the travelling standards to the WMO/GAW Reference Standard at NOAA/ESRL are given in the appendix. Information on instrumentation and standards is given above (same system as for CO).

Results

The results of the assessment are shown in the Executive Summary, and the individual measurements of the TS are presented in the following Tables.

Table 11. CH_4 aggregates computed from single analysis (mean and standard deviation of mean) for each level during the comparison of the Picarro G1301 #143-CFADS040 instrument (AL) with the WCC-Empa TS (WMO-X2004A CH_4 scale).

Date / Time	TS Cylinder	TS (ppb)	sdTS (ppb)	AL (ppb)	sdAL (ppb)	N	AL-TS (ppb)	AL-TS (%)
(16-11-24 22:31:24)	160922_FB03930	1795.38	0.08	1797.33	0.55	10	1.95	0.11
(16-11-24 23:31:30)	160825_FB03365	1920.93	0.11	1922.85	0.47	10	1.92	0.10
(16-11-25 00:31:24)	130819_FB03855	1890.19	0.04	1892.04	0.58	10	1.85	0.10
(16-11-25 01:31:30)	160922_FF30491	1766.97	0.12	1770.20	0.51	10	3.23	0.18
(16-11-25 02:31:36)	160922_FA02785	1721.73	0.07	1723.22	0.49	10	1.49	0.09
(16-11-25 03:31:36)	160926_FB03346	1883.52	0.12	1885.53	0.44	10	2.01	0.11

Carbon Dioxide Comparisons

All procedures were conducted according to the Standard Operating Procedure (WMO, 2007) and included comparisons of the travelling standards at Empa before the comparison of the analysers. Details of the traceability of the travelling standards to the WMO/GAW Reference Standard at NOAA/ESRL are given in the appendix. Information on instrumentation and standards is given above (same system as for CO).

Results

The results of the assessment are shown in the Executive Summary, and the individual measurements of the TS are presented in the following Table.

Table 12. CO_2 aggregates computed from single analysis (mean and standard deviation of mean) for each level during the comparison of the IFTS instrument (AL) with the WCC-Empa TS (WMO-X2007A CO_2 scale).

Date / Time	TS Cylinder	TS (ppm)	sdTS (ppm)	AL (ppm)	sdAL (ppm)	N	AL-TS (ppm)	AL-TS (%)
(16-11-24 22:31:24)	160922_FB03930	407.47	0.04	407.55	0.06	10	0.08	0.02
(16-11-24 23:31:30)	160825_FB03365	412.96	0.05	413.17	0.07	10	0.21	0.05
(16-11-25 00:31:24)	130819_FB03855	387.03	0.02	387.11	0.08	10	0.08	0.02
(16-11-25 01:31:30)	160922_FF30491	400.30	0.08	400.46	0.05	10	0.16	0.04
(16-11-25 02:31:36)	160922_FA02785	396.37	0.10	396.47	0.05	10	0.10	0.03
(16-11-25 03:31:36)	160926_FB03346	417.06	0.03	417.27	0.06	10	0.21	0.05

Table 13. CO_2 aggregates computed from single analysis (mean and standard deviation of mean) for each level during the comparison of the NDIR instrument (AL) with the WCC-Empa TS (WMO-X2007A CO_2 scale).

Date / Time	TS Cylinder	TS (ppm)	sdTS (ppm)	AL (ppm)	sdAL (ppm)	N	AL-TS (ppm)	AL-TS (%)
(17-03-07 08:48:44)	160926_FB03346	417.06	0.03	416.60	0.03	23	-0.46	-0.11
(17-03-07 09:17:46)	130819_FB03855	387.03	0.02	386.83	0.03	22	-0.20	-0.05
(17-03-07 09:47:32)	160825_FB03365	412.96	0.05	412.58	0.04	19	-0.38	-0.09
(17-03-07 10:22:23)	160922_FA02785	396.37	0.10	395.85	0.04	18	-0.52	-0.13
(17-03-07 10:52:56)	160922_FB03930	407.47	0.04	406.94	0.02	16	-0.53	-0.13
(17-03-07 11:25:43)	160922_FF30491	400.30	0.08	399.88	0.01	7	-0.42	-0.10

Nitrous Oxide Comparisons

All procedures were conducted according to the Standard Operating Procedure (WMO, 2007) and included comparisons of the travelling standards at Empa before the comparison of the analysers. Details of the traceability of the travelling standards to the WMO/GAW Reference Standard at NOAA/ESRL are given in the appendix. Information on instrumentation and standards is given above (same system as for CO).

Results

The results of the assessment are shown in the Executive Summary, and the individual measurements of the TS are presented in the following Tables.

Table 14. N_2O aggregates computed from single analysis (mean and standard deviation of mean) for each level during the comparison of the IFTS instrument (AL) with the WCC-Empa TS (WMO-X2006A N_2O scale).

Date / Time	TS Cylinder	TS (ppb)	sdTS (ppb)	AL (ppb)	sdAL (ppb)	N	AL-TS (ppb)	AL-TS (%)
(16-11-24 22:31:24)	160922_FB03930	328.34	0.09	329.27	0.26	10	0.93	0.28
(16-11-24 23:31:30)	160825_FB03365	318.53	0.02	319.52	0.27	10	0.99	0.31
(16-11-25 00:31:24)	130819_FB03855	319.54	0.05	320.74	0.17	10	1.20	0.38
(16-11-25 01:31:30)	160922_FF30491	327.16	0.06	328.47	0.17	10	1.31	0.40
(16-11-25 02:31:36)	160922_FA02785	324.60	0.10	325.66	0.22	10	1.06	0.33
(16-11-25 03:31:36)	160926_FB03346	342.12	0.11	343.30	0.30	10	1.18	0.34

WCC-Empa Traveling Standards

Ozone

The WCC-Empa travelling standard (TS) was compared with the Standard Reference Photometer before and after the audit. The following instruments were used:

WCC-Empa ozone reference: NIST Standard Reference Photometer SRP #15 (Master)

WCC-Empa TS: Thermo Scientific 49C-PS #0421507340, BKG -0.8, COEF 1.014

Zero air source: Pressurized air - Dryer – Breitfuss zero air generator – Purafil – charcoal – outlet filter

The results of the TS calibration before the audit and the verification of the TS after the audit are given in Table 15. The TS passed the assessment criteria defined for maximum acceptable bias before and after the audit (Klausen et al., 2003) (cf. Figure 18). The data were pooled and evaluated by linear regression analysis, considering uncertainties in both instruments. From this, the unbiased ozone mixing ratio produced (and measured) by the TS can be computed (Equation 6a). The uncertainty of the TS (Equation 6b) was estimated previously (cf. equation 19 in (Klausen et al., 2003)).

$$X_{TS} \text{ (ppb)} = ([TS] - 0.07 \text{ ppb}) / 1.0001 \quad (6a)$$

$$u_{TS} \text{ (ppb)} = \text{sqrt}((0.43 \text{ ppb})^2 + (0.0034 * X)^2) \quad (6b)$$

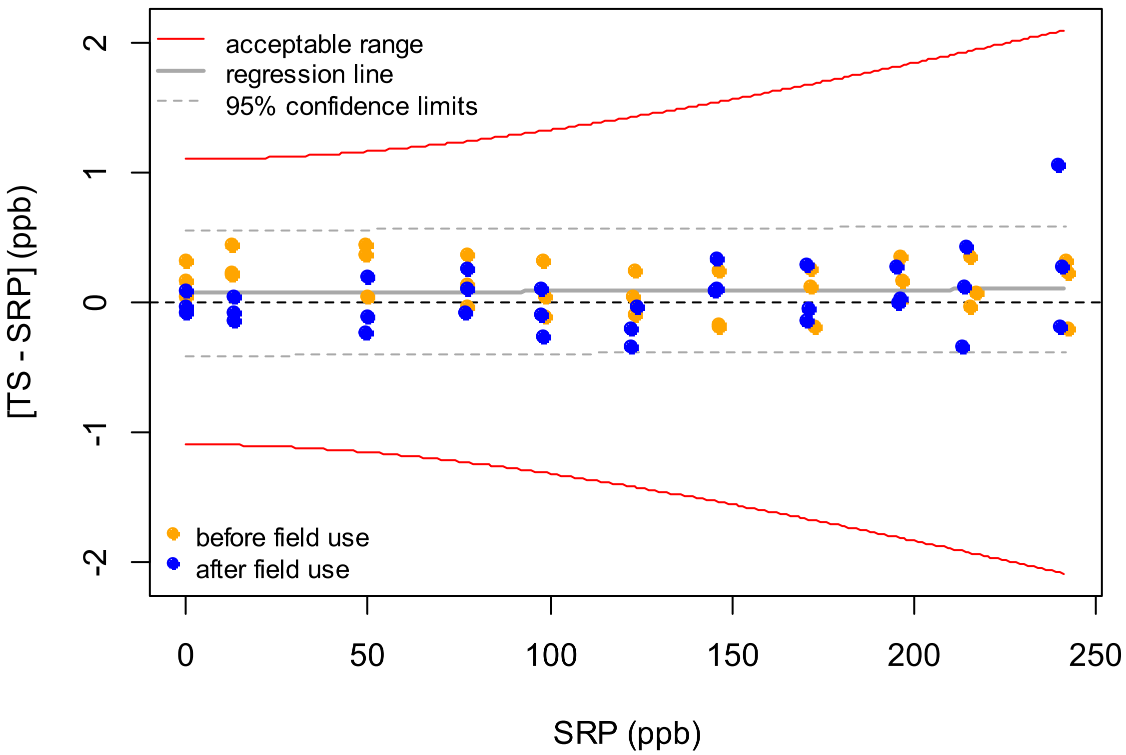

Figure 18. *Deviations between traveling standard (TS) and Standard Reference Photometer (SRP) before and after use of the TS at the field site.*

Table 15. Five-minute aggregates computed from 10 valid 30-second values for the comparison of the Standard Reference Photometer (SRP) with the WCC-Empa traveling standard (TS).

Date	Run	Level#	SRP (ppb)	sdSRP (ppb)	TS (ppb)	sdTS (ppb)
2016-09-21	1	100	98.30	0.36	98.62	0.28
2016-09-21	1	75	77.10	0.13	77.23	0.19
2016-09-21	1	215	217.16	0.35	217.23	0.21
2016-09-21	1	0	-0.02	0.34	0.30	0.14
2016-09-21	1	175	172.62	0.38	172.43	0.27
2016-09-21	1	125	122.81	0.32	122.85	0.17
2016-09-21	1	10	12.47	0.24	12.91	0.22
2016-09-21	1	50	49.71	0.25	49.75	0.19
2016-09-21	1	195	196.09	0.28	196.11	0.18
2016-09-21	1	145	146.23	0.30	146.06	0.18
2016-09-21	1	240	241.87	0.44	241.67	0.24
2016-09-21	2	75	77.28	0.47	77.24	0.22
2016-09-21	2	0	0.09	0.25	0.13	0.41
2016-09-21	2	215	215.43	0.36	215.39	0.22
2016-09-21	2	125	123.24	0.26	123.14	0.13
2016-09-21	2	170	171.73	0.22	171.84	0.11
2016-09-21	2	195	195.90	0.30	196.24	0.14
2016-09-21	2	15	12.51	0.21	12.72	0.19
2016-09-21	2	50	49.51	0.20	49.88	0.11
2016-09-21	2	100	98.39	0.27	98.43	0.09
2016-09-21	2	145	146.26	0.13	146.50	0.20
2016-09-21	2	240	241.98	0.37	242.21	0.13
2016-09-21	3	75	76.97	0.25	77.34	0.14
2016-09-21	3	170	171.64	0.27	171.89	0.11
2016-09-21	3	15	12.53	0.20	12.77	0.27
2016-09-21	3	100	98.67	0.30	98.56	0.19
2016-09-21	3	215	215.42	0.28	215.78	0.16
2016-09-21	3	145	146.29	0.17	146.10	0.17
2016-09-21	3	50	49.38	0.38	49.83	0.28
2016-09-21	3	0	0.02	0.15	0.18	0.10
2016-09-21	3	195	196.82	0.27	196.98	0.26
2016-09-21	3	125	123.20	0.22	123.43	0.24
2016-09-21	3	240	241.44	0.46	241.77	0.19
2017-05-04	4	100	77.05	0.29	77.31	0.23
2017-05-04	4	75	145.67	0.38	145.77	0.23
2017-05-04	4	215	13.13	0.23	12.99	0.18
2017-05-04	4	0	196.03	0.17	196.05	0.40
2017-05-04	4	175	170.96	0.18	170.91	0.18
2017-05-04	4	125	97.42	0.26	97.52	0.18
2017-05-04	4	10	49.82	0.25	50.01	0.22
2017-05-04	4	50	-0.06	0.35	0.03	0.12
2017-05-04	4	195	123.60	0.32	123.56	0.17
2017-05-04	4	145	214.31	0.25	214.74	0.27
2017-05-04	4	240	240.48	0.24	240.76	0.11
2017-05-04	5	75	76.97	0.38	77.08	0.29
2017-05-04	5	15	13.19	0.30	13.11	0.12
2017-05-04	5	50	49.86	0.20	49.74	0.21
2017-05-04	5	100	97.72	0.25	97.63	0.12
2017-05-04	5	195	195.03	0.26	195.30	0.16
2017-05-04	5	120	122.22	0.20	122.02	0.24

Date	Run	Level#	SRP (ppb)	sdSRP (ppb)	TS (ppb)	sdTS (ppb)
2017-05-04	5	170	170.43	0.29	170.71	0.12
2017-05-04	5	145	145.19	0.53	145.27	0.20
2017-05-04	5	0	0.06	0.29	-0.02	0.11
2017-05-04	5	215	213.84	0.23	213.96	0.49
2017-05-04	5	240	239.82	0.51	239.62	0.34
2017-05-04	6	120	122.06	0.28	121.72	0.26
2017-05-04	6	170	170.13	0.68	169.98	0.26
2017-05-04	6	215	213.20	0.22	212.85	0.21
2017-05-04	6	50	49.26	0.29	49.02	0.27
2017-05-04	6	0	0.02	0.18	-0.01	0.12
2017-05-04	6	100	98.09	0.27	97.83	0.15
2017-05-04	6	195	195.35	0.34	195.36	0.23
2017-05-04	6	15	13.01	0.16	13.05	0.22
2017-05-04	6	145	145.66	0.56	146.00	0.25
2017-05-04	6	75	76.96	0.21	76.88	0.21
2017-05-04	6	240	239.51	0.48	240.57	0.43

#the level is only indicative.

Greenhouse gases and carbon monoxide

WCC-Empa refers to the primary reference standards maintained by the Central Calibration Laboratory (CCL) for Carbon Monoxide, Carbon Dioxide and Methane. NOAA/ESRL was assigned by WMO as the CCL for the above parameters. WCC-Empa maintains a set of laboratory standards obtained from the CCL that are regularly compared with the CCL by way of traveling standards and by addition of new laboratory standards from the CCL. For the assignment of the mole fractions to the TS, the following calibration scales were used:

CO: WMO-X2014A scale (Novelli et al., 2003)
CO_2: WMO-X2007 scale (Zhao and Tans, 2006)
CH_4: WMO-X2004A scale (Dlugokencky et al., 2005)
N_2O: WMO-X2006A scale (http://www.esrl.noaa.gov/gmd/ccl/n2o_scale.html)

More information about the NOAA/ESRL calibration scales can be found on the GMD website (www.esrl.noaa.gov/gmd/ccl). The scales were transferred to the TS using the following instruments:

CO and N_2O: Aerodyne mini-cw (Mid-IR Spectroscopy using a Quantum Cascade Laser). CO_2 and CH_4: Picarro G1301 (Cavity Ring Down Spectroscopy).

Table 16 gives an overview of the WCC-Empa laboratory standards that were used for transferring the CCL calibration scales to the WCC-Empa TS. The results including estimated standard uncertainties of the WCC-Empa TS are listed in Table 17, and Figure 19 shows the analysis of the TS over time. Usually, a number of individual analysis results dating from before and after the audit was averaged. During these periods, the standards remained usually stable with no significant drift. If drift is present, this will lead to an increased uncertainty of the TS.

Table 16. NOAA/ESRL laboratory standards at WCC-Empa.

Cylinder	CO (ppb)	CH_4 (ppb)	N_2O (ppb)	CO_2 (ppm)
CC339478	463.76	2485.25	357.19	484.39
CB11499	141.03	1933.77	329.15	407.33
CB11485	110.88	1844.78	328.46	394.30

Table 17. Calibration summary of the WCC-Empa travelling standards.

TS	CO (ppb)	sdCO (ppb)	CH_4 (ppb)	sdCH_4 (ppb)	CO_2 (ppm)	sdCO_2 (ppm)	N_2O (ppb)	sdN_2O (ppb)
130819_FB03855	161.10	0.24	1890.19	0.04	387.03	0.02	319.54	0.05
160825_FB03365	171.12	0.19	1920.93	0.11	412.96	0.05	318.53	0.02
160922_FA02785	45.47	0.10	1721.73	0.07	396.37	0.10	324.60	0.10
160922_FB03930	68.82	0.13	1795.38	0.08	407.47	0.04	328.34	0.09
160922_FF30491	53.40	0.29	1766.97	0.12	400.30	0.08	327.16	0.06
160926_FB03346	86.60	0.13	1883.52	0.12	417.06	0.03	342.12	0.11

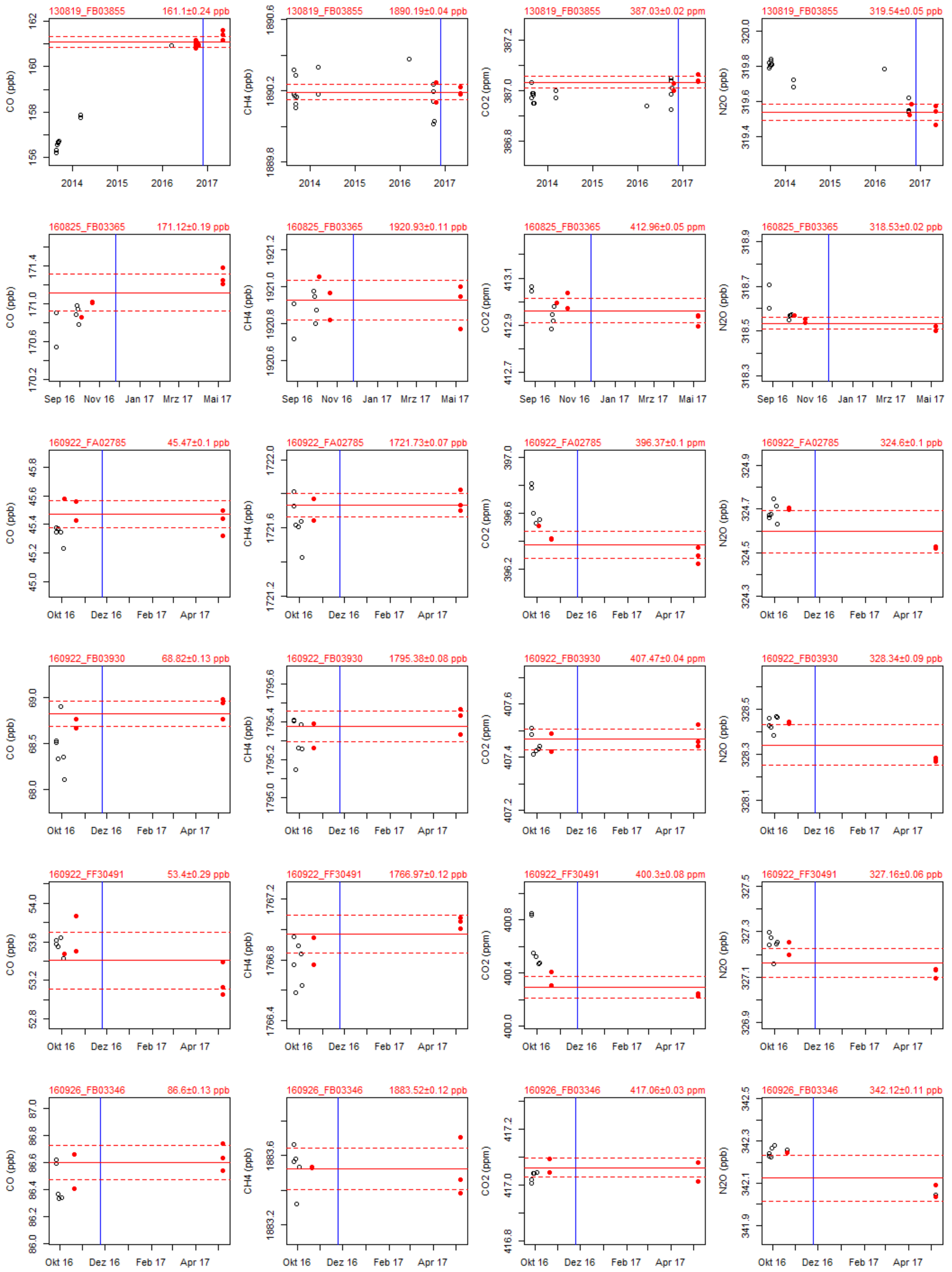

Figure 19. *Results of the WCC-Empa TS calibrations. Only the values of the red solid circles were considered for averaging. The red solid line is the average of the points that were considered for the assignment of the values; the red dotted line corresponds to the standard deviation of the measurement. The blue vertical line refers to the date of the audit.*

Calibration of the WCC-Empa travelling instrument

The calibration of the WCC-Empa travelling instrument is shown in the following figures. For CH_4 and CO_2, the Picarro G2401 SN # 1497-CFKADS2098 was calibrated every 1805 min using one WCC-Empa TS as a working standard, and two TS were used as targets. Based on the measurements of the working standard, a drift correction using a loess fit was applied to the data, which is illustrated in the figure below. The maximum drift between two WS measurements was approx. 2 ppb for CH_4 and 0.1 ppm for CO_2. Both target cylinders were within half of the WMO GAW compatibility goals for all measurements.

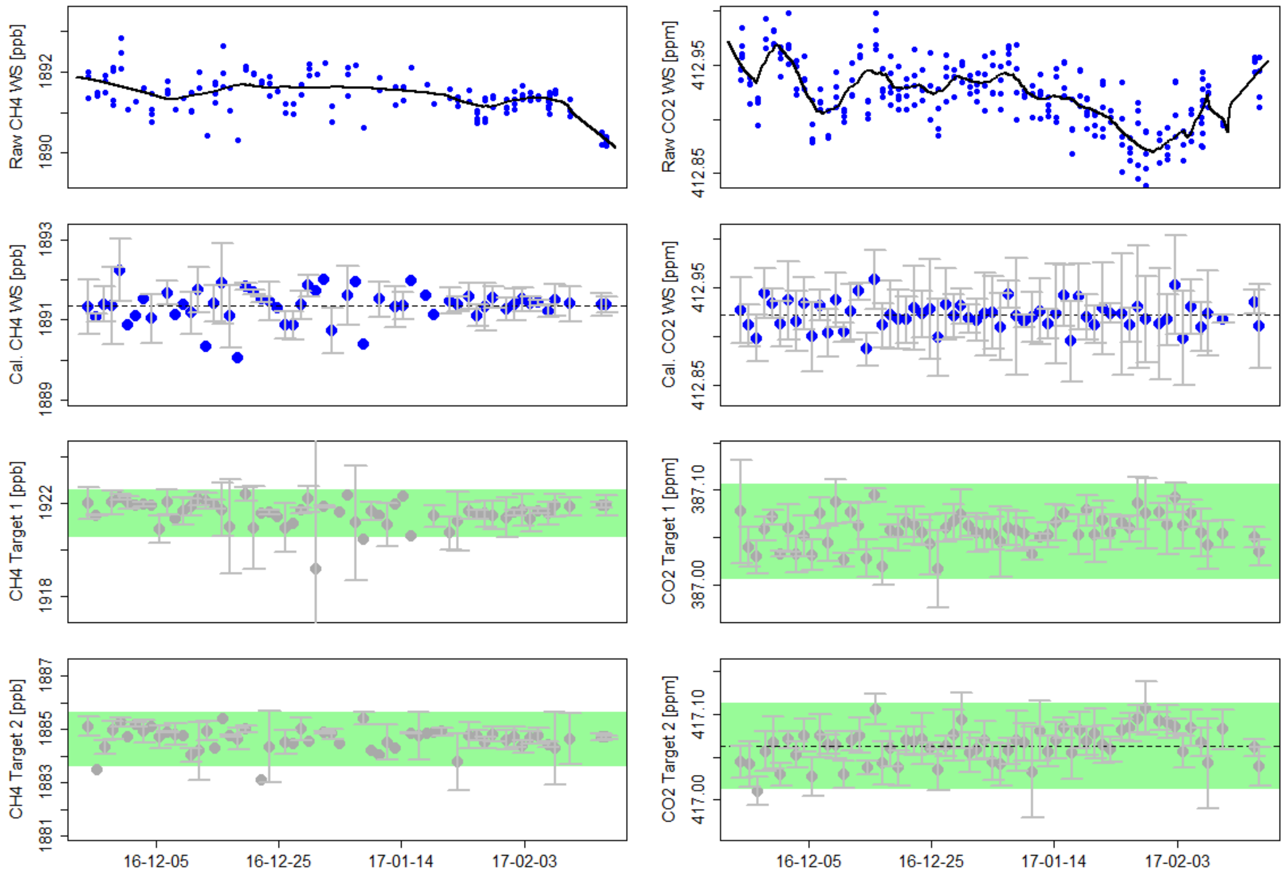

Figure 20. *CH_4 (left panel) and CO_2 (right panel) calibrations of the WCC-Empa-TI. The upper panel shows raw 1 min values of the working standard and the loess fit (black line) used to account for drift. The second panel shows the variation of the WS after applying the drift correction. The two lower most panels show the results of the two target cylinders. Individual points in the three lower panels are 5 min averages, and the error bars represent the standard deviation. The green area represents half of the WMO/GAW compatibility goals.*

For CO, the Picarro G2401 was calibrated every 1805 min three WCC-Empa TS as a working standards. Based on the measurements of the working standards, a drift correction using a loess fit was applied to the data, which is illustrated in the figure below.

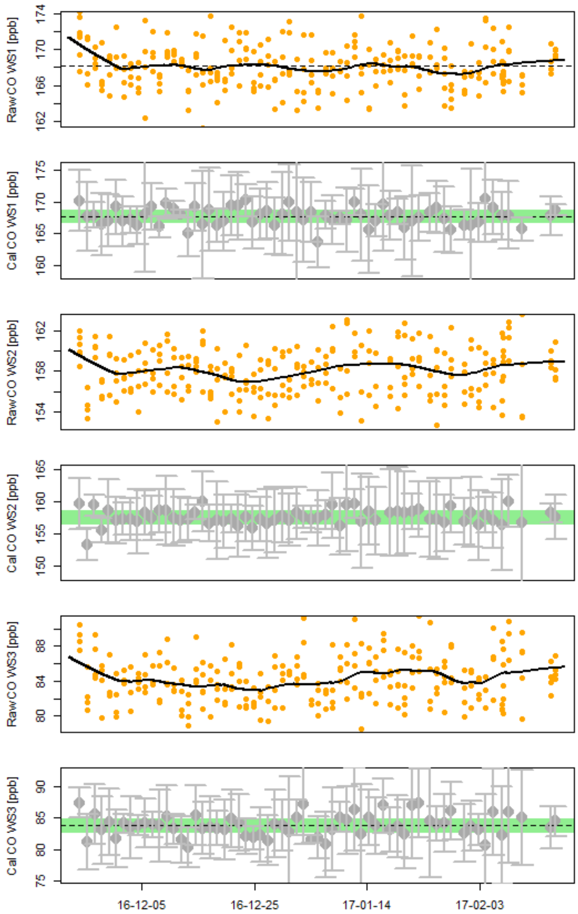

Figure 21. *CO calibrations of the WCC-Empa-TI. The panels with the orange dots show raw 1 min values of the working standards and the loess fit (black line) used to account for drift. The other panels show the variation of the WS after applying the drift correction. Individual points in these panels are 5 min averages, and the error bars represent the standard deviation. The green area represents half of the WMO/GAW compatibility goals.*

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LIST OF ABBREVIATIONS

a.s.l	above sea level
BKG	Background
COEF	Coefficient
CRDS	Cavity Ring-Down Spectroscopy
DQO	Data Quality Objective
ECD	Electron Capture Detector
ESRL	Earth System and Research Laboratory
GAW	Global Atmosphere Watch
GAWSIS	GAW Station Information System
GHG	Greenhouse Gases
LAU	Lauder GAW Station
LS	Laboratory Standard
NA	Not Applicable
NDIR	Non-Dispersive Infrared
NIWA	National Institute of Water and Atmospheric Research
NOAA	National Oceanic and Atmospheric Administration
PI	Principle Investigator
QCL	Quantum Cascade Laser
SOP	Standard Operating Procedure
SRP	Standard Reference Photometer
TI	Travelling Instrument
TS	Traveling Standard
WCC-Empa	World Calibration Centre Empa
WDCGG	World Data Centre for Greenhouse Gases
WMO	World Meteorological Organization