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Research Article

Keywords: Coal-fired unit, Quantification of FFCO₂ emission, Calibration, Calculation-based method, Measurement-based method

Posted Date: September 21st, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10947682/v1>

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Additional Declarations: No competing interests reported.

Reconciling calculation-based and measurement-based CO₂ emissions: a full-year calibration study for carbon accounting management in a 1000 MW unit

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Abstract

Electricity and heat generation account for 34.5%–38.6% of global greenhouse gas emissions. Accurate and timely quantification of fossil fuel CO₂ (FFCO₂) emissions from coal-fired unit is essential for carbon markets and climate policy. However, systematic discrepancies between the calculation-based method (using emission factors) and the measurement-based method (using CO₂-CEMS) undermine the reliability of emission reports and the fairness of compliance mechanisms. This study develops a calibration model for a 1000 MW ultra-supercritical coal-fired unit on multi-condition (32%–100% load) comparison tests, using a 3D pitot tube for velocity reference and a Gaussian process regression (GPR) model for CO₂ concentration calibration ($R^2=0.91$). Before calibration, the calculation-based method yielded 12.31% lower emissions than the measurement-based method; after velocity and concentration calibration, and aligning the system boundaries, the relative deviation converged to -2.66%. Further

analysis reveals that the deviation is not a fixed bias but depends on the unit's load profile: the calculation-based method tends to overestimate emissions at full-load dominated conditions, but underestimate at deep peak-shaving conditions, which is a reversal attributed to the dilution of high-carbon coals in the monthly composite sample and to the default oxidation rate (0.99) overestimating low-load combustion efficiency. This study provides the first quantitative empirical demonstration of consistency between the two methods at full operating conditions throughout the year, offering technical support for improving carbon emission data for compliance purposes.

Highlights

- A calibration framework for the measurement-based method is developed, comprising multi-condition and multi-point velocity calibration, and system boundary harmonization.
- The relative deviation between the two methods converged from 12.31% to -2.66% after applying the framework.
- The deviation direction reversed at deep peak-shaving: the calculation-based method underestimates emissions by 0.48%–0.58%, attributed to the dilution of high-carbon coals in the monthly composite sample.

Key words

Coal-fired unit, Quantification of FFCO₂ emission, Calibration, Calculation-based method, Measurement-based method

1. Introduction

The urgent need for effective emission reduction strategies is underscored by the accelerating rise in global temperature. From 2010 to 2019, the average temperature already increased by 1.06 °C above pre-industrial levels, primarily driven by human activities. Without additional mitigation, projections indicate a further warming of 3.3 °C to 5.7 °C by 2100 (IPCC 2021). Although the Paris Agreement aims to limit this increase to well below 2 °C (striving for 1.5 °C), current national pledges remain insufficient to meet these targets (Dvorak et al. 2022; Wang et

al. 2025). As a major emitter, China has committed to achieving carbon neutrality by 2060, a critical contribution to global climate goals (Kong et al. 2023). Power industry contributed about 34.54%–38.62% of global greenhouse gas emissions (Wu et al. 2025), and with electrification driving growing power demand (IEA 2018), an accurate and transparent carbon accounting system for the power sector becomes essential for designing targeted reduction policies (Tranberg et al. 2019; Gurney et al., 2024).

Two main approaches are used to quantify CO₂ emissions from power generation facilities: the calculation-based method and the CO₂-CEMS measurement method (hereafter referred to as the measurement-based method). The calculation-based method estimates emissions using fuel activity data, oxidation rate, and elemental carbon content (MEE 2022). The measurement-based method directly measures flue gas temperature, humidity, CO₂ concentration, flow velocity, and cross-sectional area via a CO₂-CEMS installed at the flue duct (NEA 2021; EPA 2026). The calculation-based method involves a longer procedural chain, with the shortest quantification interval being monthly, whereas the measurement-based method can provide hourly data (Zhang 2024).

Globally, the adoption of these methods varies significantly. The EU and most other countries primarily use the calculation-based method, while the U.S. EPA requires fossil-fuel-fired units with annual emissions exceeding 25,000 tCO₂ to use the measurement-based method under a cap-and-trade framework (EPA 2009). China currently adopts the calculation-based method as the primary approach for its power generation facilities, while the measurement-based method has been piloted since 2021. Pilot results indicate that the measurement-based method can cover all carbon emissions from power generating units, with relative deviations between the two methods within 10% for coal-fired units (Zhang 2024). However, its direct application remains constrained by the poor accuracy of S-type pitot tubes in non-ideal flow fields, their widespread use in China's power facilities, and the absence of a fully established monitoring support system (Zhang 2024).

China's energy structure is dominated by coal, and the country is required to phase out fossil fuels and achieve carbon neutrality within just 30 years. Currently, China's electricity carbon emission factors are published only as annual averages, which suffer from poor timeliness and hinder energy restructuring and coordinated industrial planning (Liu et al. 2021). Meanwhile,

China primarily implements concentration-based control over air pollutant emissions from thermal power facilities, which differs from the U.S. approach (Zhang et al. 2025). These factors together motivate the need for an improved, real-time-capable carbon accounting framework for the power sector.

The two methods have been extensively compared at various operational conditions in the literature. Pei et al. (2023) found comparable results between the two methods on a 320 MW unit. Li and Yang (2025) observed that measured results could deviate either above or below calculated values, attributing the deviations to coal sampling representativeness and default oxidation rates. Quick (2014) and Gurney et al. (2016) reported that the differences between the two methods at the national level were small but could exceed 10% at finer spatial scales. More recently, studies have begun to recognize that frequent load cycling, particularly at deep peak-shaving conditions, can significantly affect boiler combustion efficiency and carbon emission characteristics (Wang et al. 2024; Chen et al. 2025), challenging the accuracy of conventional emission factor methods. However, the specific mechanism linking the deviation between the two methods to the unit's load profile has not been systematically investigated over a full annual cycle. Despite these efforts, a systematic year-round investigation linking the deviation between the two methods to the unit's load profile remains absent.

To address this gap, semi-annual multi-condition (32%–100% load) comparison tests of flue gas velocity and CO₂ concentration were performed. A 3D pitot tube was used as the reference for velocity measurements, and multi-point sampling was conducted for CO₂ concentration calibration. A GPR model was developed to calibrate the measurement section mean CO₂ concentration from single-point CEMS measurements. The system boundaries of the two methods were aligned by deducting emissions from desulfurization, denitrification, and air introduction. After calibration, the results of the measurement-based method were validated against the calculation-based method. This approach enables a real-time, accurate reflection of the unit's carbon emission level and provides a pathway toward consistent and comparable results between the two methods.

2. Materials and methods

2.1 Unit description

This study was conducted on a 1000 MW ultra-supercritical coal-fired unit (GNQY01) in Guangdong over the full calendar year of 2025. The unit commenced commercial operation in December 2023. It is equipped with six raw coal bunkers, each with a storage capacity of approximately 500 t, storing coal of different calorific values (ranging from 3200 kcal/kg to 5800 kcal/kg). The unit operates by blending coals of varying calorific values, with load regulation achieved primarily through adjusting the coal feed rate. The unit is a double tangential circle coal-fired boiler with a dedicated flue gas exhaust duct. The stack height is 240 m, and the desulfurization outlet is at approximately 41 m. The CO₂-CEMS are installed on the vertical flue at a height of 95 m (Fig. 1), where the stack diameter is 8.4 m. The unit is equipped with desulfurization and denitrification processes, using limestone as the desulfurization agent and urea as the denitrification agent.

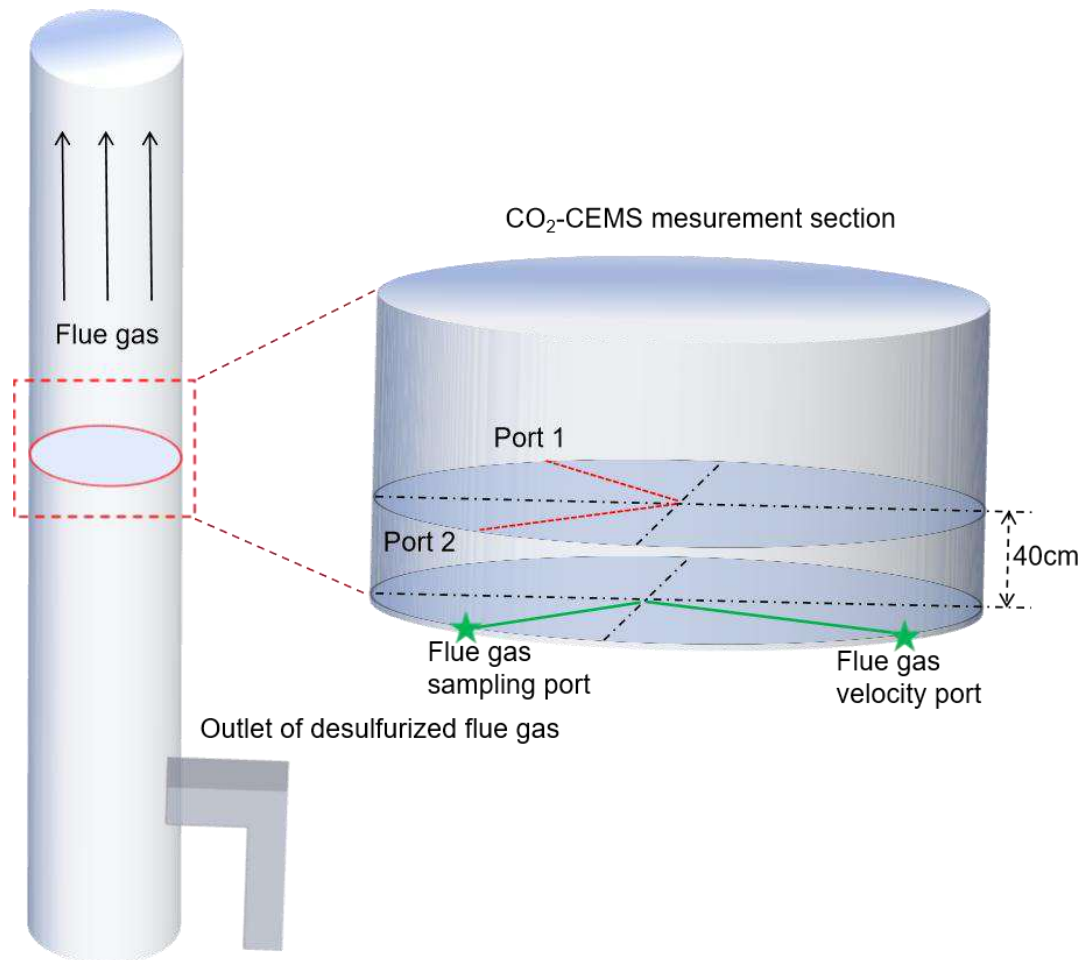


Fig. 1 Schematic diagram of the measurement ports for CO₂-CEMS and reference method

2.2 The calculation-based method

Carbon emissions are calculated as the sum of CO₂ emissions from the combustion of all fossil fuels during the statistical period:

$$E = \sum_{i=1}^n (FC_i \times C_{ar,i} \times OF_i \times \frac{44}{12}) \quad (1)$$

where E is the emissions from the combustion of fossil fuels, tCO₂; FC_i is the consumption of the i -th type of fossil fuel, t; $C_{ar,i}$ is the as-received basis elemental carbon content of the i -th type of fossil fuel, tC/t; OF_i is the carbon oxidation rate of the i -th type of fossil fuel, 0.99; 44/12 is the ratio of the relative molecular mass of carbon dioxide to that of carbon; i is fossil fuel type code.

Coal consumption was measured daily by in-feed belt scales. The as-received elemental carbon content was determined from monthly composite samples prepared by coning and quartering, and analyzed by a CNAS-accredited laboratory. The carbon oxidation rate for coal was taken as 0.99, following MEE (2022).

2.3 The measurement-based method

The measurement-based method followed DL/T 2376 (NEA 2021).

$$G_h = C_d \times 3600 \times F \times V_s \times \frac{273}{273 + t_s} \times \frac{B_a + P_s}{101325} \times (1 - X_{sw}) \quad (2)$$

where G_h is flue gas CO₂ mass flow rate by hour, t/h; C_d is CO₂ concentration in flue gas, %; F is measurement section area, m²; V_s is the flue gas velocity, m/s; t_s is flue gas temperature, °C; B_a is atmospheric pressure, Pa; P_s is flue gas static pressure (gauge), Pa; X_{sw} is flue gas moisture content.

Daily, monthly, and annual emissions were then aggregated:

$$G_d = \sum_{i=1}^{24} G_{hi} \quad (3)$$

$$G_m = \sum_{i=1}^{D_m} G_{di} \quad (4)$$

$$G_y = \sum_{i=1}^{D_y} G_{di}' \quad (5)$$

where G_d is CO₂ emission by day, t/d; G_{hi} is CO₂ emission in the i -th hour, t/h; G_m is CO₂ emission by month, t/m; G_{di} is CO₂ emission in the i -th day of the month, t/d; G_y is CO₂ emission

by year, t/a; G_{di} is CO₂ emission in the i -th day of the year, t/d; D_m is number of days in the month; D_y is days in the year.

For the CO₂-CEMS on the studied unit, the velocity, temperature, and humidity measurement points were installed at 1.2 m from the inner wall of the flue, while the CO₂ concentration sampling point was located at 1.5 m from the inner wall. Comparison tests using reference methods were conducted in March and September (Table 1, Table 2).

Table 1 Measurement methods for key parameters

Key parameters	Studied unit	Reference method
Velocity	S-type pitot tube (Anrongxin APT2000)	3D pitot probe (National Institute of Metrology, China)
CO ₂ concentration	extractive NDIR (Thermo Fisher 410i)	Extractive NDIR (Laoying 3021)
Temperature	platinum resistance (An rongxin APT2000)	data from CO ₂ -CEMS
Moisture	resistance-capacitance method (Vaisala DMT143)	data from CO ₂ -CEMS
O ₂ concentration	zirconia principle (Thermo Fisher TXO-1000)	data from CO ₂ -CEMS

Table 2 The measurement conditions for the comparison testing

Key parameters	March	September
Monitoring site	5 sites in the same radius	10 sites in two different radii
Velocity		
Load factor	32%–37%, 40%–41%, 51%–55%, 68%–74%; 99%	52%–59%, 98%–100%
CO ₂ concentration	extractive NDIR (Thermo Fisher 410i)	extractive NDIR (Laoying 3021)

2.4 Calibration method

The novel calibration procedure comprised three main components, flue gas velocity calibration, CO₂ concentration calibration, and harmonization of the quantification boundaries between the two methods, respectively.

For gas **velocity calibration**, 3D pitot tube was used as the reference method for velocity tests. According to GB/T 16157 (MEE 1996), 10 measurement points were arranged along two perpendicular diameters (port 1 and port 2) at the measurement section (Fig. 1). The velocity

field coefficient K_V , defined as the ratio of the measurement section mean velocity to the single-point measured velocity, was determined from the multi-point tests. Following DL/T 2376 (NEA 2021) and 40 CFR Part 75 (EPA 2002), comparison testing was conducted every six months. The resulting K_V values were applied to calibrate the CEMS single-point velocity measurements to the measurement section mean velocity for each load range.

For **CO₂ concentration**, for the January–June period, calibration was performed using a single-point reference method measurement compared against the CO₂-CEMS data. For the July–December period, multi-point CO₂ concentration measurements were conducted at 10 points according to GB/T 16157 (MEE 1996). A GPR model was developed using these multi-point data to predict the measurement section mean CO₂ concentration from the single-point CEMS measurement. The model was established using 10 sets of multi-point comparison test results conducted in September, each comprising 10 measurement points distributed along different radii. The model inputs were time, load factor, coordinate position, and point-measured CO₂ concentration; the output was the measurement section mean. The kernel combined RBF (local non-linear fitting), DotProduct (global linear extrapolation), ConstantKernel, and WhiteKernel. Based on the observed load-dependent behavior of the CO₂ concentration field, a larger noise term ($\alpha=0.05$) was assigned to low-load ($<70\%$) and a smaller term ($\alpha=0.01$) to high-load ($\geq 70\%$). GroupKFold cross-validation grouped by spatial region ensured that points from the same region did not appear across different folds. Cross-validation yielded an R^2 of 0.91 and a mean absolute error (MAE) of 0.15% (volume fraction). The model was trained on September data and applied to calibrate the July to December CEMS data.

For **boundary harmonization**, the calculation-based method accounts only for CO₂ emissions from fuel combustion and does not include emissions from desulfurization and denitrification agents. The measurement-based method, however, captures these emissions because the flue gas stream contains CO₂ released from these agents during the process. To align the system boundaries, the CO₂ emissions attributable to desulfurization and denitrification agents were deducted from the measurement-based results after velocity and concentration calibration.

$$G_{ch}=C_{cd} \times 3600 \times F \times V_{cs} \times \frac{273}{273+t_s} \times \frac{B_a+P_s}{101325} \times (1 - X_{sw}) - E_{nh} - G_{airh} \quad (6)$$

where G_{ch} is calibrated flue gas CO₂ mass flow rate, t/h; C_{cd} is calibrated CO₂ concentration by comparison testings; V_{cs} is flue gas velocity calibrated by the checkout, m/s; E_{nh} is CO₂ emission form limestone and other sorbent used in the hour; G_{airh} is CO₂ emission form the air.

$$E_{nh}=\sum_k CAL_k \times EF_k + \sum_k URE_k \times EF_{kure} \quad (7)$$

where CAL_k is carbonate consumption of the k -th desulfurization agent, t; EF_k is carbonate emission factor for the k -th desulfurizer agent, tCO₂/t, and the EF of CaCO₃ is 0.440; URE_k is urea consumption of the k -th denitration agent, t; EF_{kure} is urea emission factor for the k -th denitration agent, tCO₂/t, and the EF_{ure} of urea is 0.73.

2.5 Statistical analysis

Relative difference (RD) for the calculation-based method results and the measurement-based method results were calculated:

$$RD=\frac{G-E}{E} \times 100 \quad (8)$$

where RD is relative difference for the calculation-based method results and the measurement-based method results, %; E is emissions from the calculation-based method, tCO₂; G is emissions from measurement-based method, tCO₂.

$$RD_c=\frac{G_c-E}{E} \times 100 \quad (9)$$

where RD_c is relative difference for the calculation-based method results and calibrated the measurement-based method results, %; G_c is calibrated emissions from measurement-based method, tCO₂.

3. Results

3.1 Unit operating conditions

During 2025, the unit generated 60.37×10^5 MW·h of electricity and operated for 8853 hours, with four start-up and shut-down cycles using plasma ignition. The hourly average load factor ranged from 10.1% to 101.3%, with a mean of 72.6% and a median of 82.2%, indicating a substantial number of low-load periods (mainly during late night and holidays) (Fig. 2). The

load distribution in 2025 exhibited a bimodal pattern, with a large concentration of operating hours near full load (>99%) and at low load (<50%), and relatively few transition periods. On workdays, loads rose in the morning and fell at night, with a pronounced peak-to-valley difference; on weekends, the load remained lower throughout the day and the peak hours shifted later. Monthly loads were highest in February and December, and lowest in September, closely followed by November.

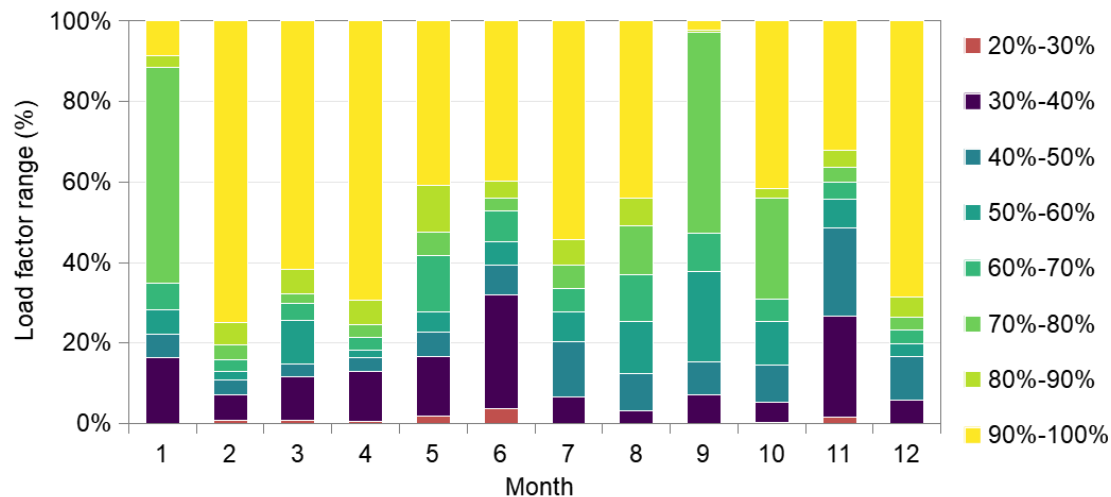


Fig. 2 The distribution of load factor in each month during 2025 for the studied unit

3.2 Carbon oxidation rate

The monthly carbon oxidation rates were calculated following the Guidelines for Accounting and Reporting Greenhouse Gas Emissions for Power Generation Enterprises in China (Trial) (NDRC 2013). The monthly oxidation rates ranged from 0.992 to 0.998, with a mean of 0.996, all exceeding the default value of 0.99 specified in the guidelines. The oxidation rate showed a moderate negative correlation with the monthly average load factor ($r=-0.47$), suggesting a tendency toward lower combustion efficiency during low-load periods. However, it showed only weak correlations with monthly emissions from both the calculation-based method ($r=-0.12$) and the calibrated measurement-based method ($r=-0.06$). This might reflect that the oxidation rate, derived from intermittent fly ash and bottom slag sampling, is not temporally aligned with the load fluctuations, limiting its ability to capture efficiency variations across different load regimes.

3.3 Flue gas velocity distribution

The hourly averaged flue gas velocity measured by CO₂-CEMS showed a strong positive correlation with unit load ($r=0.96$), ranging from 7.23 m/s to 21.01 m/s across the full load range. Multi-condition comparison testings conducted in March and September revealed the following velocity field characteristics (Table 3). First, the relative root mean square (RMS) of velocities across all measuring points was generally ≤ 0.15 , except at low-load conditions (below 40%), where it exceeded 0.15, for instance, reaching 0.20 at 32%–35% load. Second, the velocity field coefficient K_V decreased with increasing load: from 1.032 at ca.32% load to 0.915 at 100% load (based on the March tests). Third, across all operating conditions, the maximum velocity consistently occurred at the 2/7 radius position, with velocities decaying towards both the centre and the wall (Fig. 3). Fourth, at high load (100%), the mean velocity at monitoring port 1 was 4.1% higher than that at port 2, whereas at medium load (55%), the difference was only 0.7%, indicating the presence of distinct high- and low-velocity zones at high load, which was not evident at low load.

These load-dependent variations necessitate the use of condition-specific calibration coefficients for accurate emission quantification (EPA 2002). Based on the March test results, the following K_V values were applied: 0.914 for load $\geq 70\%$; 0.924 for $40\% \leq \text{load} < 70\%$; and 1.032 for load $< 40\%$. Using these coefficients, flue gas velocity data from January to June were calibrated. For the July–December period, coefficients derived from the September tests were used: 0.925 for load $\geq 70\%$ and 0.965 for load $< 70\%$ (Table 3).

Table 3 Flue gas velocity comparison at different conditions

Load factor	Date	Unit CEMS flow velocity mean (m/s)	Reference method flow velocity mean (m/s)	Relative root mean square of flow velocities	Velocity field coefficient K_V
98%–100%	31 March 2025	18.53	16.95	0.067	0.914
60%–66%	4 March 2025	12.70	12.02	0.067	0.946
55%	19 March 2025	11.99	11.13	0.060	0.928
40%–41%	20 March 2025	9.64	9.04	0.094	0.938
32%–37%	4 March 2025	7.18	7.41	0.149	1.032

100%	September 2025	20.47	18.93	0.117	0.925
51%–53%	September 2025	12.45	12.01	0.140	0.965

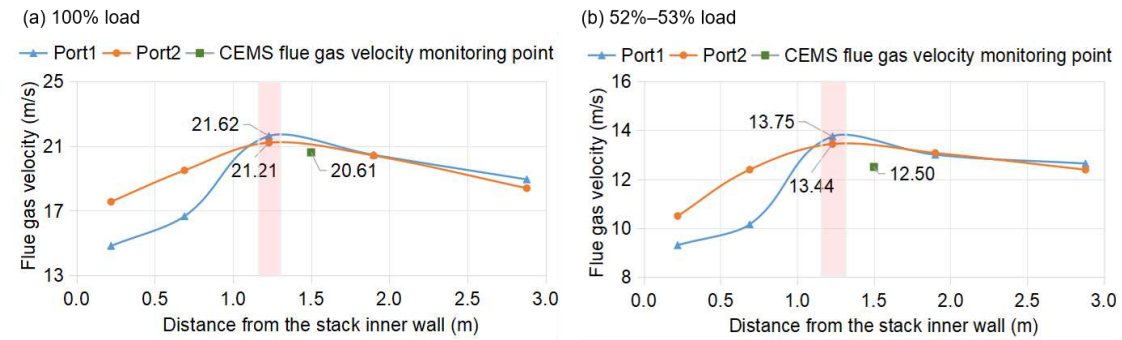


Fig. 3 Flue gas velocity distribution at 95 m height from September comparison tests. (a) 100% load (September 2025); (b) 52%–53% load (September 2025). The solid line represents the CEMS single-point measurement; the symbols represent reference method measurements at 10 points along two radii. The red shade indicates the 2/7 radius position where the maximum velocity consistently occurs.

3.4 Concentration distribution

The hourly averaged CO₂ concentration measured by CO₂-CEMS increased with load, ranging from 7.02% to 18.61%, and was positively correlated with both load ($r=0.86$) and flue gas velocity ($r=0.85$). During the comparison testings, the CO₂ concentration measured by the reference method was consistently lower than that from the CO₂-CEMS (Table 4). Multi-point CO₂ concentration comparison testings were conducted in September 2025, yielding 10 valid datasets at high and medium load conditions. Following the U.S. EPA stratification criteria, at high load (100%), two of the four test runs indicated mildly stratified conditions and two indicated stratified conditions. At medium load (52%–58%), among the six test runs, one was non-stratified, three were mildly stratified, and two were stratified (Fig. 4). Comparing the mean CO₂ concentrations along the two radii, at high load the means differed markedly: 14.62% and 15.01%, respectively, with a relative deviation of 2.6%, a trend consistent with the flue gas velocity distribution. At medium load, however, the means along the two radii were essentially

consistent (13.27% and 13.37%, relative deviation: 0.74%), contrary to the velocity distribution pattern.

Based on the March comparison testing results, bias adjustment factors (BAFs) for CO₂ concentration were determined for three load ranges: 0.930 for load $\geq 70\%$; 0.920 for $40\% \leq$ load $< 70\%$; and 0.871 for load $< 40\%$. These BAFs were applied to calibrate CO₂ concentration data from January to June. For the July–December period, a GPR model was developed to predict the measurement section mean CO₂ concentration from the CEMS point measurements.

Table 4 CO₂ concentration comparison at different conditions

Date	Load factor	CEMS CO ₂ (%)	Reference method CO ₂ (%)	Relative root mean square of CO ₂	CO ₂ adjustment factor (BAF)
31 March 2025	98%–100%	16.37	15.22	—	0.930
4,19,20 March 2025	47%–55%	13.01	11.97	—	0.920
4,20 March 2025	32%–39%	11.97	10.42	—	0.871
25 September 2025	100%	15.96	14.80	0.047	0.928
26 September 2025	51%–53%	14.21	13.32	0.016	0.938

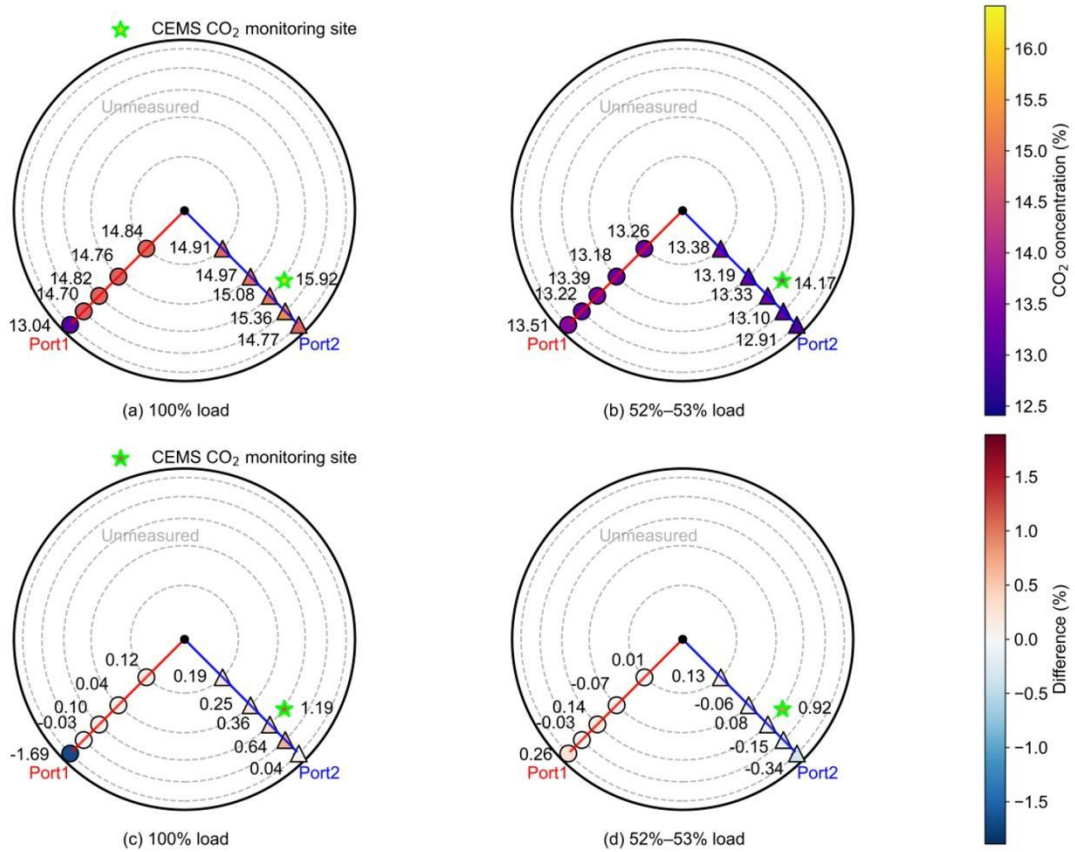


Fig. 4 CO₂ concentration distribution at 95 m height from September comparison tests. (a) Concentration field at 100% load (September 2025); (b) concentration field at 52%–53% load; (c) deviation of each point from the measurement section mean at 100% load; (d) deviation of each point from the measurement section mean at 52%–53% load.

3.5 FFCO₂ emission

Following MEE (2022), carbon emissions were calculated monthly based on as-received elemental carbon content. The total annual carbon emission was 44.82×10^5 tCO₂. Monthly carbon emissions were positively correlated with electricity generation, approximately linearly. December had the highest electricity generation (6.31×10^5 MW·h) and the highest emissions (4.53×10^5 tCO₂); June had the lowest (2.95×10^5 MW·h and 2.18×10^5 tCO₂). The carbon emission intensity ranged from 704.8 gCO₂/kW·h (February) to 771.2 gCO₂/kW·h (November), with an annual average of 743.3 gCO₂/kW·h.

Following DL/T 2376, the measurement-based method provides CO₂ emission data at 1-minute resolution, aggregated into hourly and monthly totals. The annual emissions ranked as: measurement-based > calculation-based > calibrated measurement-based (Fig. 5). The

contributions of four bias sources were quantified. The respective contribution of CO₂ concentration and velocity were derived from the difference between each single-factor calibrated emission and the uncalibrated emission. The contributions from desulfurization/denitrification sorbents and air-derived CO₂ were calculated as their respective proportions of the total bias. The resulting contributions were: CO₂ concentration(45.9%), velocity(49.7%), desulfurization and denitrification sorbents (2.2%), and air-derived CO₂ (2.2%).

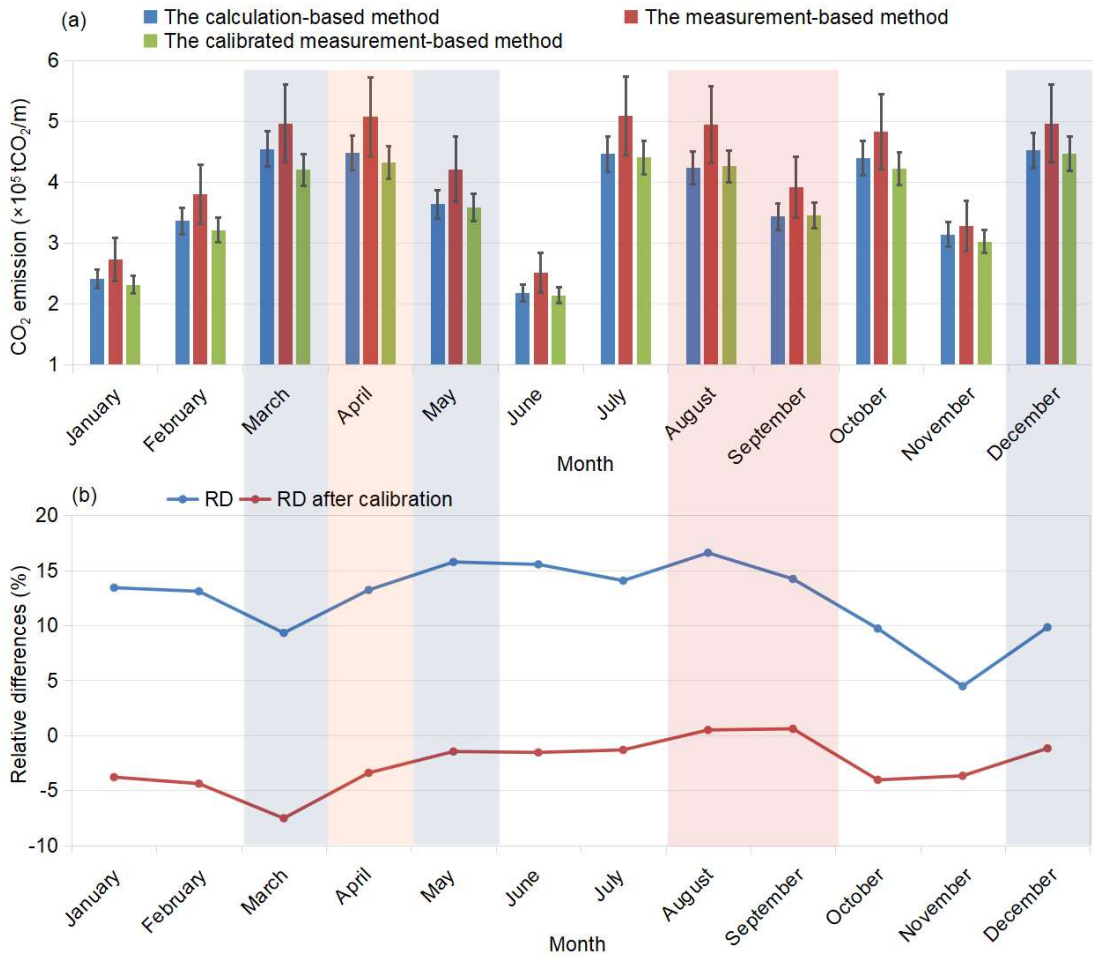


Fig. 5 The CO₂ emissions from different methods and relative differences during 2025. (a) Monthly CO₂ emissions estimated by the calculation-based method, the uncalibrated measurement-based method, and the calibrated measurement-based method. Error bars represent expanded uncertainties ($k=2$). (b) Relative deviations between the calculation-based method and the measurement-based method (RD), and between the calculation-based method and the calibrated measurement-based method (RDc). The orange shade indicates the month (March) with the largest negative RDc after calibration. The red shade indicates the months (August, September) where the RDc reverses from

negative to positive. The blue shade indicates the months (February, April, and December) dominated by full-load steady-state operation.

3.6 Relative difference of carbon emission from different methods

The difference between the calculation-based and measurement-based results in 2025 was 5.52×10^5 tCO₂, corresponding to an RD of 12.31%. The difference between the calibrated measurement-based result and the calculation-based result was 1.19×10^5 tCO₂, corresponding to an RD_c of -2.66%. Before calibration, the monthly relative deviations were all positive (4.45%–16.58%). After applying the velocity and concentration calibrations, the relative deviations for all months except August and September turned negative (-7.55%–-1.19%), while August and September retained slight positive deviations (0.48% and 0.58%, respectively).

4 Discussions

4.1 Discrepancies between the calculation-based and measurement-based methods

4.1.1 General patterns of reported discrepancies

Previous studies have extensively compared the two methods for CO₂ emission accounting in coal-fired power plants. As summarized in Table 5, the calculation-based method typically yields higher results, though the opposite trend has also been reported, indicating that the deviation direction is not fixed but depends on site-specific factors including coal sampling representativeness, CEMS measurement accuracy, and operating conditions. The RD (12.31%) and RD_c (-2.66%) obtained in this study fall within this reported range.

Table 5 Comparison of calculated and measured results across different studies

Unit(s)	Load conditions	Quantification methods	Key findings	References
1 unit, 320 MW	3 loads (54.7%– 63.2%), 1-hour tests	Calc.: measured or default C content, oxidation rate 0.99;	Calc. > Meas.; mean RD: 4.9%	Pei et al. 2023
8 units, 330MW-	Feb., Mar., Aug. 2022		Meas.>Calc.;	Zhi 2020

1060 MW		Meas.: CO ₂ -CEMS, S-type pitot tube	RD:-20.27% to 8.32% (mean -4.56%)	
4 units	Dec. 2023–Apr. 2024		both directions observed; RD: -15.28% to 2.90%	Li and Y 2025
1 unit	Aug. 2011		Calc. < Meas.; 12%–19% lower	Lee et al. 2015
210 U.S. plants	2009	Calc.: EIA dataset; Meas.: EPA dataset	±10.8% deviation range; mean diff.-0.7%	Quick 2014
Major U.S. coal-fired plants	2001–2009		monthly deviations: >±6% for ca. 50% of plants; >±13% for ca. 20%	Gurney et al. 2016
1 unit, 1000 MW	full year 2025, 32%–100% load	Calc.: monthly composite C content, oxidation rate 0.99; Meas.: CO ₂ -CEMS, 3D pitot tube, GPR calibration	RD: 12.31%; RDc: -2.66% (after calibration)	This study

4.1.2 Sources of uncertainty in the two methodologies

The discrepancies originate from fundamentally different sources of uncertainty inherent in each method. For the calculation-based method, uncertainties arise primarily from three aspects (Liu et al. 2022; Fan et al. 2025; Li et al. 2025): (1) coal consumption recording, due to process operations and coal bunkers buffering; (2) elemental carbon content, where monthly composite sample may not capture time-resolved variations during coal switching or low-load operation; and (3) the oxidation rate, where the default value of 0.99 may overestimate efficiency at low-load or transient conditions. For the measurement-based method, accuracy depends on two sequential requirements (EPA 2022; Im et al. 2023; Tian et al. 2026): (1) the precision of individual sensors; and (2) the measurement section representativeness, as the flue gas velocity and CO₂ concentration fields are inherently non-uniform. This study employed a multi-point

matrix to characterize both fields, providing a more representative measurement section average than single-point configurations.

The uncertainties of the three approaches were evaluated using the GUM (Guide to the Expression of Uncertainty in Measurement) framework (Supplementary Tables S1 to S9). For the calculation-based method, the relative combined uncertainty was estimated as $\pm 6.40\%$ ($k=2$), following Pan et al.(2026), with the oxidation rate and the representativeness of coal sampling identified as the largest contributors. For the measurement-based method, two estimates were derived following (Korellis and Dene 2015). The single-point CEMS configuration yielded $\pm 12.77\%$ ($k=2$), attributable to three components: comparison test itself (shared with the calibrated measurement-based method), spatial representativeness of the single-point measurement (estimated via a t -test-based factor), and CEMS sensor uncertainty (Supplementary Tables S2 to S5). The calibrated method (after multi-point calibration) gave $\pm 6.26\%$ ($k=2$), where the spatial component was derived from the relative root mean square of the multi-point measurements, and the instrumentation uncertainty corresponds to the reference method instruments (Supplementary Tables S6 to S9). The reduction from 12.77% to 6.26% confirmed the effectiveness of the multi-point calibration strategy in improving measurement reliability. The overlap of the expanded uncertainty intervals ($k=2$) indicated the two methods were consistent within measurement uncertainty.

4.1.3 Implications of present findings

While the GUM uncertainty analysis confirms the reliability of both methods, it alone does not explain the observed deviation patterns. The following two sections address this gap: section 4.2 examines the rationale for multi-point calibrations, and section 4.3 investigates how operational conditions influence the deviation characteristics.

4.2 Main factors affecting the measurement-based method

This study mainly discussed the influence of flue gas velocity and CO₂ concentration on the measurement-based method.

4.2.1 Distribution characteristics and calibration of flue gas velocity

Due to bends and reducers upstream, flue gas generates tangential velocity components and forms non-uniform low fields (Tian et al. 2026). Conventional S-type pitot tubes can not identify incoming flow direction, causing measured velocity, especially at high-load conditions,

to deviate from the true axial component (Korellis and Dene 2015). In contrast, 3D pitot tube can recognize flow direction and calculate the axial velocity component converted from the tangential velocity, more accurate single-point measurements (Johnson et al. 2020). Therefore, the decreasing trend of K_v with increasing load reflects the larger measurement errors of the S-type pitot tube at high-load conditions.

Comparison tests along two radii in September revealed distinct high- and low-velocity zones, but not at low-load conditions. This was likely because, at high loads, enhances fluid inertia amplifies vortices induced by upstream structures; at medium and low loads, weaker inertia allows viscous dissipation to dominate, producing a more uniform flow field.

The load-dependent variations have direct implications for emission quantification. A single-point configuration cannot capture the heterogeneous velocity field, and the bias varies with load, being most severe at high loads. A multi-point matrix was therefore employed to characterize the velocity distribution across the measurement section, capturing the high- and low-velocity zones that single-point measurements inevitably miss. This approach is consistent with the U.S. EPA Method 2 and similar standards that recommend multi-point traverses for velocity field characterization (EPA 2002).

4.2.2 Distribution characteristics and calibration of CO₂ concentration

The U.S. EPA requires stratification testing for CO₂ sampling point selection in coal-fired units (EPA 2002; NDRC 2013). Under EPA Method 3A, a single point is acceptable if deviations from the measurement section mean are $\leq 0.3\%$; three points are required if deviations exceed 0.3% but are $\leq 0.5\%$. In practice, however, few plants perform such tests, and most use only two fixed sampling points (Korellis and Dene 2015). In China, standards have evolved: earlier ones (GB/T 16157-1996, HJ/T 397-2007) allow single-point sampling, while recent standards (GB/T 10184-2025, GB/T 45869-2025) recommend or require stratification testing (Table 6). Nevertheless, single-point sampling without stratification remains the mainstream practice.

Table 6 Relevant standards and specifications for the setting of flue gas CO₂ concentration sampling points in Chinese thermal power plants

Standard/Specification	Sampling point position	Stratification test	Key feature
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GB/T 16157-1996 (MEE 1996)	near the measurement section center	not mentioned	foundational standard; single-point allowed
HJ/T 397-2007 (MEE 2007)	near the measurement section center	not mentioned	follows GB/T 16157
HJ 75-2017 (MEE 2017)	not specified (focus on straight pipe length)	not mentioned	CEMS technical spec; focuses on SO ₂ , NO _x , and particulates
DL/T 2376-2021 (NEA 2021)	not specified (focus on straight pipe length)	not mentioned	first CO ₂ -specific CEMS standard; sampling rules inherited from HJ 75
HJ 1405-2024 (MEE 2024)	not specified (focus on straight pipe length)	not mentioned	emission outlet spec; follows HJ 75
GB/T 10184-2025 (SAMR 2025a)	grid method for acceptance tests; single-point allowed only if velocity/O ₂ fields are uniform	conditional	boiler performance test code; allows single- point only at verified uniform conditions
GB/T 45869-2025 (SAMR 2025b)	recommended: 1/3 to 1/2 of diameter; or choose point closest to measurement section mean after test	recommended	first standard to recommend stratification testing for CO ₂ sampling point selection

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419 Multi-point CO₂ measurements at high and medium loads revealed two key findings. First,
420 unlike velocity, CO₂ concentration showed no fixed high- or low-concentration points; the
421 distribution along two radii was non-uniform, with some points exceeding the mean by 0.3%-
422 0.5%. Second, at high load, concentrations along the two radii differed markedly, suggesting
423 distinct zones; at medium load, these differences were less pronounced. This behavior may
424 reflect inertial effects carrying heavier CO₂ molecules into high-velocity zones, while the
425 absence of fixed high- or low-concentration points suggests the concentration field is more
426 sensitive to instantaneous mixing than to velocity. Without stratification testing, single-point
427 sampling can deviate from the mean by 0.3% or more, leading to proportional errors in emission
428 quantification that vary with load and cannot be corrected by a simple fixed offset.

429 Existing standards address such deviations through calibration factors: U. S. CFR 75 adopts

a conservative approach ($BAF \geq 1$) to avoid underestimation (EPA 2022), while China's DL/T 2376 adopts an unbiased approach (E_{ac} may be > 1 or < 1) to minimize deviation from the true value (Table 7). Both share a critical limitation: the calibration factor relies on the arithmetic mean deviation between the reference method and the CEMS, without verifying whether the reference sampling points are truly representative of the measurement section mean. If the reference method is biased due to poor representativeness, as is likely given the absence of stratification testing, the resulting calibration factor will incorporate that same bias, effectively calibrating the CEMS to a biased reference.

Table 7 Comparison of CO₂ concentration calibration methods

	CFR 75	DL/T 2376
CO ₂ concentration calibration method	$BAF = 1 + \frac{ \bar{d} }{CEM_{avg}} \quad (10)$ where BAF is bias adjustment factor, calculated to the nearest thousandth; $\bar{d}$ is arithmetic mean of the difference during measurement; CEM_{avg} is mean of the data values provided by the CO₂-CEMS during measurement.	$E_{ac} = 1 + \frac{\bar{d}}{CEM_{avg}} \quad (11)$ where E_{ac} is bias adjustment factor, calculated to the nearest thousandth; $\bar{d}$ is arithmetic mean of the difference between data from CO₂-CEMS and data from checkout method during measurement; CEM_{avg} is mean of the data values provided by the CO₂-CEMS during measurement.

Unlike the velocity field, which has a relatively stable spatial structure enabling simple regression between point and mean velocities, CO₂ concentration distribution lacks a persistent pattern. The absence of fixed high- or low-concentration points and its sensitivity to instantaneous mixing make simple linear or polynomial regressions inadequate: they either fail to capture the load-dependent nonlinearity or overfit the limited data. A GPR model was therefore developed to predict the measurement section mean from the point-measured CO₂ concentration (see section 2.4 for model details). GPR was selected because it requires no a priori assumptions about the functional form and performs well with limited data. Cross-validation confirmed the model's ability to reliably estimate the measurement section mean concentration from single-point CEMS measurements across varying load conditions.

4.2.3 Effect of air on the measurement-based method results

During unit operation, ambient air is introduced as combustion air. Since atmospheric air contains a small fraction of CO₂ (≈ 437 ppm)(Li et al. 2026), the CEMS-measured CO₂ concentration includes both fuel-derived and air-borne CO₂, requiring calibration to avoid systematic overestimation. Palstra and Meijer (2010) quantified this air-derived CO₂ contribution in coal-fired power plants as approximately $0.34\% \pm 0.06\%$ of the total flue gas CO₂. Accurate air velocity measurements are not available at the target unit to calculate this contribution directly; therefore, the literature-derived value of 0.34% is adopted as a reasonable approximation. Given the small proportion and narrow uncertainty range ($\pm 0.06\%$), the potential error introduced by using this literature value is considered negligible relative to other measurement uncertainties. The measurement-based results were accordingly adjusted by subtracting this contribution.

4.3 Factors influencing the variation of calibrated relative deviations

The RD_c showed a distinct pattern: negative for most months, but positive in August and September, with March exhibiting the largest negative deviation (Table 8). As summarized in Table 8, RD_c was governed by two competing factors: the conservative bias of the default oxidation rate, and dynamic changes in coal consumption and carbon content during low-load and transient operation. The former dominated at full-load steady conditions; the latter progressively offset, and in extreme cases reversed, the conservative bias when low-load operation became sufficiently severe.

Table 8 Classification of monthly operating conditions and corresponding calibrated deviations

Category	Representative months	RD _c	Dominant mechanism
I. full-load dominated steady state	February, April	-3.42%—4.40%	oxidation rate of 0.99 without reflecting low-load oxidation rate; overestimation of elemental carbon content
Ia. full-load with elevated emission intensity	March	-7.55%	oxidation rate of 0.99 without reflecting low-load oxidation rate; overestimation of elemental carbon content; both amplify overestimation

II. balanced regime with mixed operation	January, May, June, July, October, November	-1.19%—4.06%	elemental carbon content underestimated; oxidation rate overestimated. Both partially offset conservative bias
III. deep peak-shaving reversal	August, September	0.48%, 0.58%	elemental carbon content underestimated (dilution of high-carbon coals); oxidation rate overestimated

The anomalies in August-September and March are likely explained by the inherent limitation of the monthly composite sampling strategy. The sampling point is located at the coal bunker inlet, while the boiler consumes coal from the bunker outlet with a time lag of hours to tens of hours. During deep peak-shaving (August–September), low-load operation typically requires firing high-calorific coals for flame stability. The high-carbon signal from these coals was diluted in the monthly composite by low-calorific coals used during other periods, causing the measured carbon content to be systematically lower than the coal actually fired during low-load periods, leading to underestimation. Conversely, in March, if a specific batch of high-carbon coal accounted for a disproportionately large share of the composite sample, the monthly carbon content would be biased upward, amplifying overestimation. A similar temporal mismatch existed for the oxidation rate: monthly averages from fly ash and bottom slag samples did not reflect instantaneous combustion efficiency during low-load periods, reinforcing underestimation in August–September and potentially contributing to overestimation in March.

Direct coal quality measurements corresponding to specific operating conditions were not available to verify these mechanisms definitively. Nevertheless, the combination of the time-lag effect, the operational practice of low-load coal switching, and the observed load-dependent deviation pattern provided a coherent explanation for the month-to-month variation in RDc.

5 Conclusions

The measurement-based method provides continuous real-time carbon emission data and thus offers critical support for emission reduction in coal-fired units, but remains underdeveloped. This study conducted a calibrated framework for a 1000 MW coal-fired unit based on multi-condition (32%–100% load) comparison tests over a full year, including load-dependent

velocity calibration coefficients derived from 3D pitot tube measurements and a GPR model for CO₂ concentration calibration ($R^2=0.91$, $MAE=0.15\%$). System boundaries were aligned by deducting emissions from desulfurization, denitrification, and air introduction. Before calibration, the measurement-based method yielded 12.31% higher emissions than the calculation-based method. After calibration, the relative deviation converged to -2.66%, with overlapping expanded uncertainties ($\pm 6.40\%$ for the calculation-based method, $\pm 6.26\%$ for the calibrated measurement-based method, $k=2$), confirming consistency between the two methods.

The deviation is not a fixed bias but depends on the unit's operating profile, traced to a temporal mismatch in the monthly composite strategy. Sampling occurs at the bunker inlet, while the boiler fires coal from the bunker outlet with a lag of hours. Thus, high-carbon coals used during low-load periods are diluted in the monthly composite, causing underestimation; conversely, a high-carbon batch can bias the composite upward, leading to overestimation.

This study provides a practically feasible pathway to achieve comparability between the two methods. The findings are based on a single 1000 MW unit, and the generalizability of the calibration coefficients and the GPR model to other unit types, coal qualities, or flue duct configurations remains to be further validated. Further research on a wider range of units is needed to assess the long-term stability of the calibration approach under varying operational and maintenance conditions.

Author's contributions

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Funding

This work is supported by the Science and Technology Program of the State Administration for Market Regulation (Grant no. 2024MK093, and 2025MK098).

Declarations

Competing interest

The authors declare that they have no conflicts of interest in this work.

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