



Investigating the impact of control strategies on the sulfur dioxide emissions of South Korean industrial facilities using an aircraft mass balance approach

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HIGHLIGHTS

- We present an airborne observational SO₂ dataset from large industrial point sources in Taean, South Korea.
- We calculated SO₂ emission to verify bottom-up emission inventories and the continuous emission monitoring dataset.
- The comparison illustrates ~40 % SO₂ emission reduction from policy implementation between 2018 and 2020.
- We found no noticeable trend in SO₂ emissions from industrial facilities before and after the COVID-19 pandemic.

ABSTRACT

We present top-down SO₂ emission estimates from four large point sources, including two power plants, a steel mill, and a petrochemical industrial facility in Taean, South Korea. The airborne observations were conducted over two years, with three intensive observations in Fall 2019, Fall 2020, and Spring 2021. During this period, an active policy implementation to reduce air pollutant emissions from large industrial point sources in the region had been exercised, in addition to activity disruptions from the global COVID-19 pandemic. We quantify the observed SO₂ emission reduction over the time period resulting from the policy implementation. The Continuous Emission Monitoring System (CEMS) datasets from the coal power plants were 1) validated by comparing them with emissions calculated using a top-down airborne mass balance method and 2) compared with the annual bottom-up national SO₂ emission inventory. The comparisons illustrate that the policy implementation resulted in a 35% reduction in SO₂ emissions from increased implementation of SO₂ reduction technologies such as scrubbers. Finally, the COVID-19 pandemic did not cause a meaningful disruption in SO₂ emissions from industrial sectors, such as steel mills and petrochemical manufacturing facilities.

1. Introduction

Accurate inventories of anthropogenic trace gas emissions are essential for effective environmental management. These inventories become the basis for planning mitigation strategies, evaluating progress toward reduction targets, and forecasting air quality. National compilation of anthropogenic gas emissions typically relies on bottom-up (BU)

approaches, which estimate emissions using reported activity levels and emission factors associated with individual sources. However, challenges arise in accurately identifying emission sources, their activity factors, and capturing their temporal variability (López-Aparicio et al., 2017; Qu et al., 2022; Zavala-Araiza et al., 2015). Therefore, independent top-down estimates derived from atmospheric measurements are valuable tools for validating inventories and ensuring compliance with

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mitigation agreements (Nisbet and Weiss, 2010; Turnbull et al., 2011).

Continuous Emission Monitoring Systems (CEMS) directly measure concentrations of air pollutants such as NO_x and SO_x and flow rate at stack exhaust, enabling the calculation of emission rates on an ongoing basis. The high frequency of CEMS measurements of industrial emissions provides the crucial data needed to characterize the temporal variability of emissions (Tang et al., 2020; Wang et al., 2022). Data from the CEMS has been utilized for real-time, site-specific monitoring of emissions, enabling the tracking of the impact of emission standards policies on emissions at individual facilities (Tang et al., 2019; Wang et al., 2022). As the utilization of CEMS data continues to expand to monitor emissions and enforce air pollutant standards in various countries, the quality and reliability of CEMS data become increasingly crucial and require a systematic evaluation.

While the effectiveness of CEMS in the US has been validated through independent measurements, there has been limited research attention on the assessment of CEMS systems in other countries, particularly those suffering from air pollution problems (Frost et al., 2006; Peischl et al., 2010; Ryerson et al., 1998; Trainer et al., 1995). Several studies have identified various technical and management challenges associated with the CEMS system in China. Xu (2011) interviewed managers and engineers at coal power plants in China, who expressed concerns about the trustworthiness of CEMS data, citing issues with sensor location impacting readings and the potential for data manipulation. Zhang and Schreifels (2011) highlighted the need for a national standard of CEMS, including inconsistent installation certification, lack of inspections, incomplete plant monitoring, and limited quality control. Karplus et al. (2018) compared the decline in SO₂ concentrations between CEMS and satellite data following the implementation of stringent SO₂ emission standards. While the satellite data corroborated the emission reductions in areas with more lenient standards, key areas with stricter standards had larger declines in the CEMS data. This may imply possible misreporting or data manipulation when compliance was more challenging or costly, requiring substantial technological or operational changes. Therefore, a consistent and independent evaluation process is required to validate the CEMS dataset.

A range of top-down approaches has been employed to verify emission inventories of point sources, utilizing satellites (Liu et al., 2018; F. Liu et al., 2020; Wren et al., 2023) and conducting aircraft measurements (Karion et al., 2013; Liggio et al., 2019; Wren et al., 2023). However, fundamental differences in temporal coverages should be highlighted as bottom-up emissions are usually assessed in total annual emissions. On the other hand, observation efforts are limited to a much shorter time frame. Previous studies have relied on the assumption of temporal constancy in yearly emissions from the inventories (Baray et al., 2018; Fiehn et al., 2020; Turnbull et al., 2011), which cannot be well justified.

The Taean region is situated in the South Chungcheong Province on the west coast of South Korea with four major industrial facilities: Dangjin Powerplant, Taean Powerplant, Hyundai Steel Mill, and Daesan Petrochemical Facility. Dangjin and Taean Powerplants were ranked as the third and fourth largest coal power plants, respectively, as of 2018 (Grant et al., 2021). The petrochemical facility, conversely, encompasses a large-scale operation with multiple intricate sources such as steam cracking, polymer production, and refining. Similarly, the steel mill is engaged in various processes, including blast furnaces, steel-making, ironmaking, and coking. Estimating the significant SO₂ emissions from both facilities using bottom-up approaches poses a significant challenge due to the complexity and substantial size of these facilities.

This study aims to obtain instantaneous emission estimates for the industrial facilities in the Taean region using aircraft-based measurements of SO₂ to compare with emissions from the South Korean national inventory and evaluate the effect of SO₂ reduction efforts in South Korea. We present our results over a two-year period from an intensive field campaign from Oct 2019 to June 2021. The emission estimates for the individual facilities are calculated by applying a top-down mass

balance (MB) method to 92 facility flight examples. A comprehensive description of the MB method and its associated uncertainties can be found in a recently published companion paper (Wong et al., 2024). The MB emissions are compared to emissions reported by a CEMS that directly captures average SO₂ emissions from the powerplant stacks in 30-min intervals for the entire year of 2020. This comparison provides an independent assessment of the accuracy of CEMS data in South Korea. Additionally, we evaluate the emission trends over the 3 campaign periods, Fall 2019, Fall 2020, and Spring 2021, performing a comparative analysis between MB-based emissions and the BU national inventory. Our assessment delves into the impact of changes in activities and mitigation measured within South Korea's industrial facilities and their implications in the context of the COVID-19 pandemic and evolving policy measures.

2. Methods

Airborne atmospheric chemistry and meteorology observations were conducted aboard the Hanseo B-1900D aircraft during an intensive two-year-long field campaign from Oct 2019 to June 2021. Aircraft flights from 3 seasons within this campaign are included in this analysis: Fall 2019, Fall 2020, and Spring 2021. The primary objective of the field campaign was to monitor seasonal variations in air pollutant concentrations and emissions at industrial facilities located in Taean, South Korea, during both Fall and Spring seasons. The Spring 2020 campaign could not sample over the industrial facilities due to the COVID-19 pandemic. The flights encompassed navigation around four key industrial sites: Dangjin Powerplant, Taean Powerplant, Hyundai Steel Mill, and Daesan Petrochemical Facility (Fig. 1). SO₂ and CO₂ was measured from the aircraft by a quadrupole chemical ionization mass spectrometer (CIMS-QMS) (Kim et al., 2007; Park et al., 2020) and cavity enhanced direct-absorption spectroscopy analyzer (CEAS, Los Gatos Research Ltd., CA) respectively. The AIMMS-30 (Aircraft Integrated Meteorological Measurement System; Aventech Research Inc., Ontario, Canada) was installed to measure meteorological parameters such as the three-dimensional field (wind directions and speed), humidity, temperature, and pressure. A detailed description of the observational system is described by (Park et al., 2020).

The flights were designed to quantify the hourly air pollutant emissions within the industrial facility area using an aircraft mass balance (MB) method that is modified from the top-down emission rate retrieval algorithm (TERRA) (Gordon et al., 2015). The aircraft flew in a circular pattern surrounding each facility at 3–8 altitudes ranging from 400 m to 1000 m above ground level (a.g.l.). The flight duration around a single facility ranges from 30 to 60 min. The stacked flight tracks form a virtual box around the facility where the pollutant mass flux through the surfaces of the box describes the total emission coming from within the box. The measurements are interpolated between the sampled altitudes (~400 m–1000 m) using radial basis function interpolation at 40 m × 20 m resolution. Mixing ratios are extrapolated from the lowest sampled altitude (~400 m a.g.l.) to the ground surface using a linear fit between the concentration sampled at the lowest altitude to a background value at the ground. A detailed methodological description can be found in (Wong et al., 2024). We refer to this method as the mass balance method (MB).

The background levels of CO₂ exhibited variability across each flight, with concentrations ranging from 408 ppm to 444 ppm. Acknowledging the impact of these background levels on emission rate calculations using the mass balance method, we opted against assuming a standard CO₂ background level of 420 ppm for all flights. Instead, individual background levels of CO₂ were determined for each instance. A histogram was made from the concentration of the sampled points where the air flux is negative, indicating an influx to the facility. The normal distribution of each histogram was assessed, and any significant outliers, possibly originating from an external source, were eliminated. The average CO₂ concentration derived from the histogram then served as

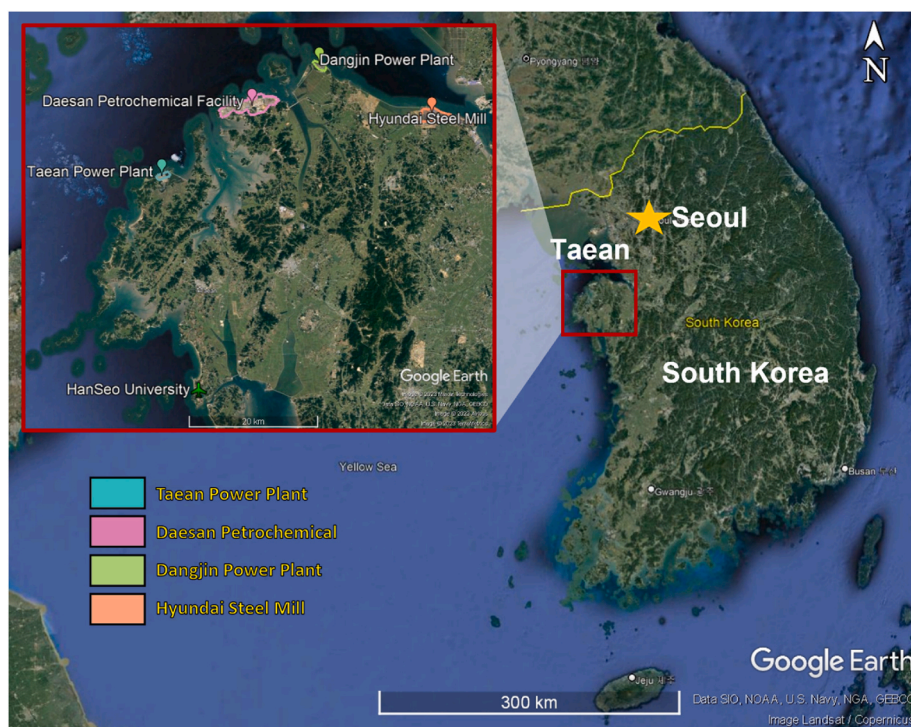


Fig. 1. Map of Tae'an County and the four industrial facilities examined during the three-year aircraft campaign in this study. The outlines define the areas associated with each facility. Map image provided by Google Earth.

the background concentration for the respective flight example. Since a substantial SO_2 background concentration was not detected during the flights, a value of 0 ppb was assigned as the background value for SO_2 .

Uncertainty estimates presented in Table S1 in our study are based on ground extrapolation and background determination methodology. The uncertainty stemming from ground extrapolation was evaluated by examining two extreme scenarios: constant extrapolation and background extrapolation from the lowest sampled altitude to the ground. In the constant extrapolation scenario, we assumed that the concentration sampled at the lowest altitude remains constant to the ground surface, providing an upper limit for emissions. Conversely, in the background extrapolation scenario, we considered the concentration below the lowest altitude sampled as the determined background value, establishing a lower limit for emissions. We quantified the uncertainty resulting from the ground extrapolation technique by comparing the percentage change in emission rates between these extreme scenarios and the default extrapolation method, a linear constant-to-background scheme. For the uncertainty associated with CO_2 background determination, we utilized the average CO_2 background value derived from the histogram constructed from measured CO_2 concentrations in the inflowing air towards the facility as the baseline. The minimum and maximum CO_2 background values were determined as one standard deviation away from the average. To quantify the uncertainty arising from background determination, we compared the percentage change in emission rates when utilizing the minimum or maximum CO_2 background values compared to the baseline CO_2 background value. Other sources of uncertainty, such as the effect of deposition, measurement error, box-height sensitivity, and box-top emission, were determined to contribute <1% to the total uncertainty and were excluded from this analysis. The flights used in this study were chosen based on the availability of multiple sampling altitudes, the continuity of directionally consistent winds over the sampling duration, and visual confirmation of plume enhancement primarily occurring within the sampled altitudes, as observed through the interpolated mixing ratio screen. A total of 92 individual facility examples were found to be suitable for identifying and quantifying emissions of the facilities (Table S1).

To compare our mass balance-based emissions (E_{MB}) with the emissions reported by the CEMS (E_{CEMS}), we utilize CEMS data from 2020 for Dangjin Powerplant, and Tae'an Powerplant. The CEMS records provide measurements of SO_2 in 30-min intervals for the entire year of 2020.

3. Results and discussion

The temporal variations of instantaneous SO_2 emissions from CEMS (E_{CEMS}) and airborne mass balance (E_{MB}) for Dangjin and Tae'an powerplants are presented in Fig. 2 and Table 1. Generally, the emission rates assessed by two independent methodologies illustrate a reasonable level of agreement, with average $E_{\text{MB}}/E_{\text{CEMS}}$ emission ratios of 0.85 for Dangjin and 1.29 for Tae'an. The agreement is observed over three weeks for nine flights. Notably, E_{MB} from the three facility flights in December (one at Dangjin and two at Tae'an) are significantly higher than the corresponding E_{CEMS} . This may suggest the presence of an additional source of SO_2 within these facilities that is not captured by the CEMS installed on the stacks during the winter month of December. Alternatively, the mass balance (MB) methodology may be less suitable for quantifying emissions during winter months due to differences in meteorological conditions, such as windspeed or boundary layer height. However, our examination of these three flights to identify any differences in meteorological conditions compared to those conducted in October and November concluded that there are no notable differences, which requires follow-up studies. Nonetheless, to our best knowledge, this is the first attempt to assess the CEMS emission rates using the airborne observational dataset in a meaningfully extended period.

As discussed, validating official emission inventories that present annual emission rates with independent top-down emission estimates is particularly important. We obtained the latest bottom-up emission inventory for SO_2 from the facilities examined in this study, encompassing emissions from 2018. It has been observed that SO_2 emissions have been steadily declining every year. To compare the inventory with our top-down emission estimates from 2020, we adjust the 2018 emissions to reflect the year 2020 using reported KEPCO coal power generation data

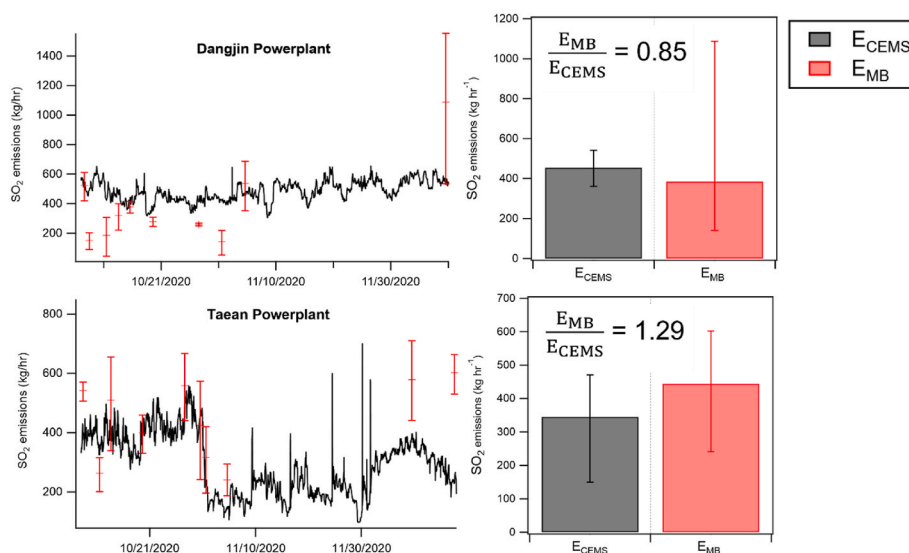


Fig. 2. (left panel) SO_2 emissions for the powerplant facilities derived from each flight using the mass balance method (E_{MB}) and from CEMS data (E_{CEMS}) in kg hr^{-1} . CEMS data is collected every half hour in grams and scaled to kg hr^{-1} for comparison to E_{MB} . Error bars on E_{MB} represent the propagated SO_2 emission change from different ground extrapolation techniques. (right panel) Average emissions during the timeframe of the flights from the CEMS (E_{CEMS}) and mass balance method (E_{MB}). Error bars denote the range in emissions among all the flights. $E_{\text{MB}}/E_{\text{CEMS}}$ for each powerplant represents the ratio of the average E_{MB} and E_{CEMS} values.

Table 1

Hourly top-down SO_2 emission estimates for 2020 using top-down aircraft mass balance (E_{MB}) and CEMS (E_{CEMS}). Mean $E_{\text{MB}}/E_{\text{CEMS}}$ is calculated from the mean E_{MB} and E_{CEMS} values.

Flight #	Date	E_{MB} (kg h^{-1})	E_{CEMS} (kg h^{-1})	$E_{\text{MB}}/E_{\text{CEMS}}$
Dangjin Powerplant				
F62	7-Oct-20	523	524	1.00
F63	8-Oct-20	150	491	0.30
F67	11-Oct-20	185	426	0.43
F68	13-Oct-20	320	475	0.67
F69	15-Oct-20	379	435	0.87
F70	19-Oct-20	279	361	0.77
F72	27-Oct-20	260	421	0.62
F74	31-Oct-20	142	418	0.34
F76	4-Nov-20	536	462	1.16
F78	9-Dec-20	1088	542	2.01
Mean		386	455	0.85
Taean Powerplant				
F63	8-Oct-20	542	408	1.33
F67	11-Oct-20	263	326	0.81
F68	13-Oct-20	510	378	1.35
F70	19-Oct-20	400	397	1.01
F72	27-Oct-20	558	468	1.19
F73	30-Oct-20	425	471	0.90
F74	31-Oct-20	317	227	1.40
F76	4-Nov-20	241	150	1.60
F78	9-Dec-20	578	385	1.50
F79	17-Dec-20	602	236	2.55
Mean		444	345	1.29

The uncertainties of E_{MB} are presented in Table S1.

Table 2

Coal power generation from Dangjin and Taean Powerplants in MWh retrieved from KEPCO, 2022. Bottom-up (BU) inventory SO_2 emissions are presented with 2018 values reported from CAPSS inventory. 2019–2021 values are scaled using the annual percent change trends in KEPCO coal power generation.

	KEPCO coal power generation (MWh)				BU inventory SO_2 emissions (kg h^{-1})			
	2018	2019	2020	2021	2018	2019	2020	2021
Dangjin	3.44E+07	3.37E+07	2.94E+07	2.74E+07	620	607	530	494
Taean	3.69E+07	3.34E+07	2.81E+07	3.00E+07	1120	1013	854	908

to account for changes in activity (Table 2) (KEPCO, 2022). Electricity generation decreased in 2020 by 15 % and 24 % when compared to 2018 at Dangjin and Taean Powerplants, respectively.

Additionally, we attempt to account for the difference in 2020 annual average emission rates with those during our sampling period in Fall 2020. For both Dangjin and Taean Powerplants, the average hourly CEMS SO_2 emission rates conducted during our Fall 2020 campaign (E_{CEMS}) was consistently 3% lower than the average annual SO_2 emission rates reported by CEMS ($E_{\text{CEMS, annual}}$) (Table 3). We adjust the average annual BU inventory emissions to average emissions during our measurements during the Fall 2020 campaign using this 3% adjustment.

Table 3

Comparison of average hourly SO_2 emissions rates from facilities in Taean. The 2018 annual BU inventory emission rates were reported as a baseline. 2020 BU activity adjusted scales the 2018 baseline using coal power generation trends from KEPCO. 2020 BU scrubber adjusted accounts for the average 35% decrease in SO_2 emissions as a result of increased scrubber enforcement. 2020 Fall BU adjusted scaled the annual emission rates to average emission rates during the sampling timeframe of the campaign. 2020 Fall Mass balance is the average SO_2 emissions from the 2020 flights. $E_{\text{CEMS, annual}}$ is the average emissions from CEMS over the entire year of 2020. E_{CEMS} is the average emissions from CEMS during the corresponding mass-balance flight sampling periods.

	SO_2 emissions (kg h^{-1})			
	Dangjin	Taean	Hyundai	Daesan
2018 annual BU baseline	620	1120	1770	1606
2020 BU activity adjusted	530	854	n/a	n/a
2020 BU scrubber adjusted	n/a	n/a	1151	1044
2020 Fall BU adjusted	513	828	n/a	n/a
2020 Fall Mass balance	386	444	1056	523
$E_{\text{CEMS, annual}}$	470	356	n/a	n/a
E_{CEMS}	455	345	n/a	n/a

The results allow us to quantitatively compare SO₂ emission rates from the Clean Air Policy Support System (CAPSS), an official emission inventory in South Korea assessed by a bottom-up (BU) methodology, to those from the airborne mass balance method for both power plants (Fig. 3 and Table 3). The results clearly illustrate a meaningful decrease (25 % and 46% for Dangjin and Taean powerplants, respectively) in SO₂ emissions rates from the adjusted 2020 Fall BU emission inventory that is independent of activity change and temporal adjustment.

Given the absence of reported changes in industrial combustion processes, such as equipment upgrades, since the 2018 reporting of recent CAPSS emission rates, we assume that the change in electricity generation reflects the SO₂ emission activity change. The additional SO₂ reduction we observe aligns with policy initiatives of both the South Korean Federal Government and the municipal authorities to reduce air pollutants from major emission sources over this period. Due to concerns regarding air pollution and international commitments for emissions reduction, the South Korean government has implemented policies to shift away from coal. The 9th Basic Plan for Power Supply and Demand, introduced in 2020, aimed to mitigate SO₂ pollution through a series of measures (Republic of Korea, 2020). In addition to suppressing electricity generation from coal power plants, the governments implemented ‘the best available technologies’ to prevent the release of SO₂ into the atmosphere, such as scrubbing technologies.

At these facilities, SO₂ and CO₂ are co-emitted from coal burning. Hence, analyzing the trends of these emissions can provide valuable insights into regional activity changes. It is particularly notable that no known scrubbing technology has been applied in contrast to SO₂. Therefore, the comparisons of temporal changes in CO₂ and SO₂ emission provide key empirical constraints to assess SO₂ emission reduction efforts. We compare the average hourly SO₂ and CO₂ emission rates retrieved using the aircraft mass balance method at Dangjin and Taean Powerplants during the 3 different sampling periods: Fall 2019, Fall 2020, and Spring 2021 (Fig. 4). At Dangjin Powerplant, we observed a 45% reduction in SO₂ emissions between Fall 2019 and Fall 2020.

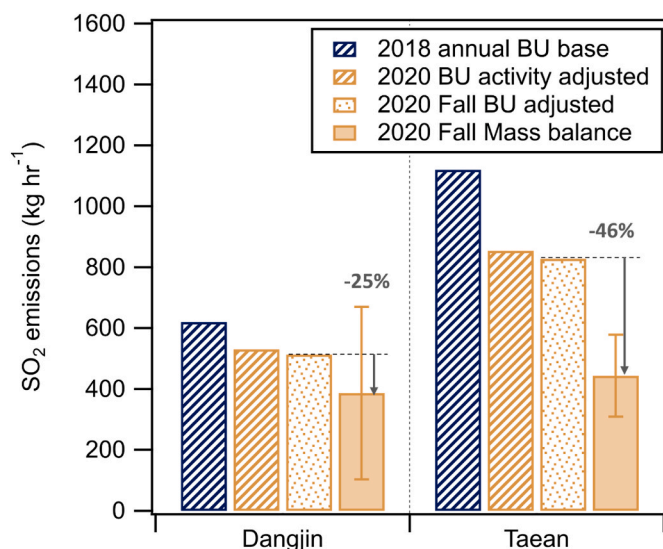


Fig. 3. Average hourly SO₂ emission comparison between the 2018 annual bottom-up baseline inventory (blue striped) and the 2020 Fall mass balance flights (solid orange). The 2018 annual inventory is adjusted to 2020 based on activity trends in coal power generation reported by KEPCO (orange striped). The 2020 annual bottom-up emissions are adjusted to the average emissions during the timeframe of the mass balance flight campaign in Fall 2020 (dotted orange) using the ratio between the average CEMS emissions for the entire year and during the campaign. The arrows and percentages represent the estimated change in emissions due to increased enforcement of SO₂ control factors at the powerplants such as scrubbers. Error bars represent the standard deviation of the emission rates from the flights.

However, during the same period, there was only a 9% reduction in CO₂ emissions. This confirms significant SO₂ reduction efforts in the time frame. Between Fall 2020 and Spring 2021, Both SO₂ and CO₂ emissions increased by 55%. The concurrent and equal increase in emissions for both species suggests that no additional measures were implemented to mitigate SO₂ emissions during this period. This is consistent with the policy implementation, mostly exercised before 2020.

At Taean Powerplant, we observed a 47% increase in CO₂ emissions between Fall 2019 and Fall 2020, indicating increased activity. Conversely, SO₂ emissions decreased by 19% during this period, suggesting that an expanded implementation of SO₂ reduction measures led to a reduction in SO₂ emissions even as activity levels contributing to emissions rose. Subsequently, observed consistent SO₂ and CO₂ emission increases by 72% and 42%, respectively, indicate a resurgence in activity in 2021. The difference in the amount of reduction can be accounted for within one standard deviation, illustrated by the error bars.

In summary, the decline in SO₂ emissions in 2020 at these two powerplants can be attributed to the implementation of scrubber technology, as it specifically targets SO₂ reduction, leaving CO₂ emissions unchanged. This suggests that additional SO₂ reduction technology was adopted between 2019 and 2020. Thus, the assessed reduction in SO₂ emissions, as quantified by the airborne observational dataset, can be attributed to the effective implementation of policies that led to the SO₂ emission reduction at these facilities. The 9% decrease in CO₂ emissions at Dangjin Powerplant and the 47% increase in CO₂ emissions at Taean Powerplant in 2020 suggest that the COVID-19 pandemic did not significantly suppress activity at these facilities between Fall 2019 and Fall 2020, and, on average, activity increased. While it's possible that activity might have been affected earlier in the year when the pandemic began in Spring 2020, our findings indicate that activity had normalized mainly by the time of our sampling period in Fall 2020. Our study underscores a noteworthy average SO₂ reduction of 35% resulting from enhanced emission reduction technology enforcement measures at these facilities in South Korea (Fig. 3).

Hyundai Steel Mill and Daesan Petrochemical Facility last provided emissions or activity reports in 2018. Consequently, we rely on our observations and the assessed 35% reduction in SO₂ emissions from scrubber implementation in the region to monitor activity trends over the three-year period from 2019 to 2021. This methodology can be justified because consistent SO₂ reduction efforts were exercised during the time frame to all the regional large point sources. This specific time frame is particularly interesting as it offers valuable insights into industrial SO₂ emissions before and after the global COVID-19 pandemic. Notably, previous studies, particularly to investigate CO₂ emission changes during the pandemic, suggested that industrial CO₂ emissions quickly bounced back after an initial decrease in the first few months of the pandemic (e.g. March to May 2020, Z. Liu et al., 2020). Fig. 5 and Table S1 illustrate the SO₂ emission rates observed in the three sampling periods of the campaign from 2019 to 2021 Hyundai Steel Mill and Daesan Petrochemical Facility. Additionally, both facilities' 2018 baseline CAPSS SO₂ emission rates are denoted in striped-blue. We apply the 35% SO₂ reduction observed from 2018 to 2020 due to scrubber implementation to the 2018 bottom-up inventory emission rates to estimate the 2020 bottom-up emission rates, assuming activity remains unchanged (striped-orange).

The SO₂ emissions from the 2018 BU emission inventory fell within the uncertainty of our 2019 Fall emissions from the mass balance flights. A substantial decrease of 50% in SO₂ emissions was observed at Hyundai Steel Mill between 2019 and 2020 from the mass balance flights. This reduction coincided with the implementation of the government's SO₂ emission reduction policy, making it challenging to definitively attribute the observed decline from 2020 and onwards to a single cause. However, we see that the 2020 inventory emissions, which have been adjusted assuming a 35% SO₂ reduction, are within the uncertainty of our observed 2020 emission rates. The average observed emissions in 2020

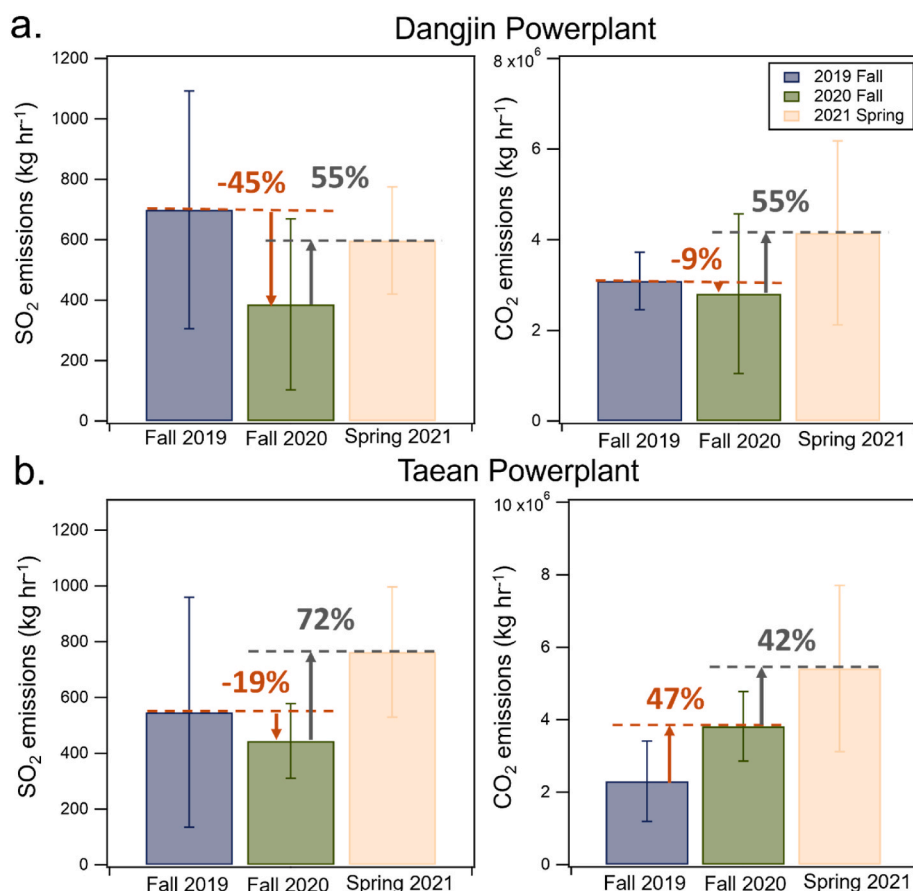


Fig. 4. Average hourly SO₂ emissions (left) and CO₂ emissions (right) retrieved from the aircraft mass balance method (E_{MB}) from each of the 3 campaign sampling periods. The average emission rates are composed for 48 individual facility flights over the 3 sampling periods. Error bars represent the standard deviation of the emission rates from the flights.

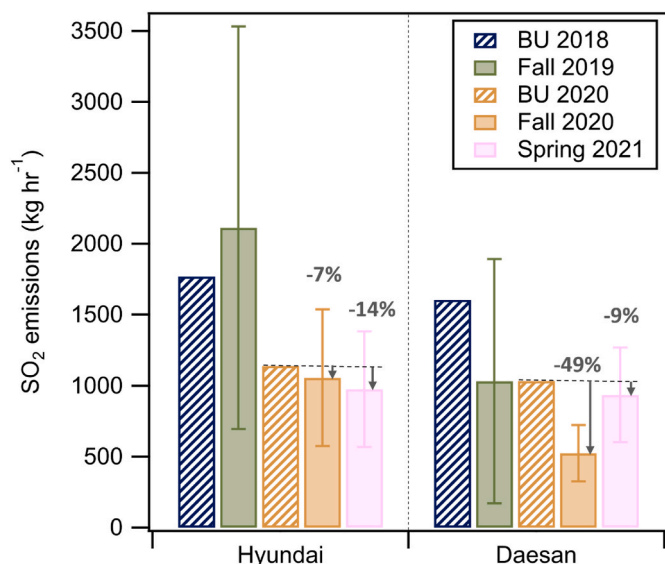


Fig. 5. Hourly SO₂ emissions from the national bottom-up inventory (striped) and from the mass balance flights (solid). The 2020 BU emissions are estimated by applying an average 35% decrease from scrubber control efficiency to the 2018 baseline bottom-up inventory. The arrow percentages represent the estimated difference in SO₂ emissions compared to the 2020 scrubber adjusted emissions. Error bars are expressed as the standard deviation for multiple flights over a single facility.

and 2021 are 7% and 14% lower than the adjusted 2020 inventory emissions. Thus, we presume that the decrease in SO₂ emissions in 2020 at Hyundai Steel Mill is primarily due to the implementation of the SO₂ reduction policy.

At Daesan Petrochemical Facility, our analysis also assesses a substantial 49% decrease in average SO₂ emissions between 2019 and 2020 from the mass balance flights. This reduction exceeded the expected 35% decrease attributed to increased scrubber implementation. This leads us to infer that decreased operational activity may have resulted in reduced SO₂ emissions. Notably, the SO₂ emissions observed in Fall 2020 were 49% lower than the adjusted 2020 inventory. This variance could, in part, be due to an overestimation in the inventory, as evidenced by the average BU inventory emission rates from 2018 being 56% higher than our observed emission rates from 2019.

The SO₂ emissions in 2021 from the petrochemical industrial facility rebounded to be 9% lower than the adjusted 2020 emission inventory. This suggests that activity at this site experienced a temporary decrease in 2020 and returned to normal in 2021. In contrast, the SO₂ emissions from Hyundai Steel Mill continued to decrease in 2021 compared to the values observed in 2020.

These observations lead to two key conclusions: First, industrial pollutant emissions over the global pandemic may not exhibit a consistent downward trend, contrary to the CO₂ emission trend noted in other studies (Liu et al., 2020). Second, while bottom-up trace gas emission inventories provide a general overview of large industrial facilities' emission capacities, the emission intensities primarily hinge on the specific industrial activities of each facility. It is also worth noting that South Korea did not implement mandatory lockdown during the COVID-19 pandemic, allowing uninterrupted industrial activities, which

may explain the absence of a distinct SO₂ signature in the SO₂ emission change before and during the pandemic.

4. Summary and conclusions

The multiyear airborne observational campaign allows us to compile temporal variations of top-down SO₂ emission estimates from Fall 2019 to Spring 2021 for four major industrial facilities in Taean, South Korea. Our evaluation results for the performance of CEMS of two of the largest power plants in the world demonstrate an acceptable agreement between airborne assessed SO₂ and CEMS-reported SO₂ emission rates. The verification provides us the confidence to use an annual CEMS dataset to scale the annual-based national emission inventory estimated by a bottom-up methodology to our hourly aircraft mass balance emission estimates for comparison. The quantitative analysis illustrates that the studied coal-burning power plants increased scrubber efficiency in 2020 which reduced SO₂ emissions by an average of 35%. This analysis allows us to quantitatively evaluate changes in SO₂ emission from Hyundai Steel Mill and Daesan Petrochemical Industrial Facility. The analysis results suggest that SO₂ emissions from the industrial sector did not illustrate any consistent trend. Instead, they are determined by industrial activities of each individual facility. This study demonstrates a prudent methodology to verify emission changes from policy implementation and societal interruption, such as the global pandemic, using existing observational datasets and emission inventories.

CRedit authorship contribution statement

Gracie Wong: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. **Minwoo Park:** Writing – original draft, Visualization, Investigation, Formal analysis. **Jinsoo Park:** Writing – original draft, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Joon-Young Ahn:** Writing – original draft, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **Minyoung Sung:** Writing – original draft, Investigation, Formal analysis. **Jinsoo Choi:** Writing – original draft, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Taehyun Park:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Jihee Ban:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Seokwon Kang:** Data curation, Formal analysis, Investigation, Writing – original draft. **Taehyoung Lee:** Data curation, Formal analysis, Investigation, Writing – original draft. **Jongho Kim:** Data curation, Funding acquisition, Investigation, Writing – original draft. **Beom-Keun Seo:** Data curation, Formal analysis, Investigation, Writing – original draft. **Jeong-Hun Yu:** Data curation, Formal analysis, Investigation, Writing – original draft. **Jeongho Kim:** Data curation, Formal analysis, Investigation, Writing – original draft. **Jung-Hun Woo:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Saewung Kim:** Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2024.120496>.

References

- Baray, S., Darlington, A., Gordon, M., Hayden, K.L., Leithead, A., Li, S.-M., Liu, P.S.K., Mittermeier, R.L., Moussa, S.G., O'Brien, J., Staebler, R., Wolde, M., Worthy, D., McLaren, R., 2018. Quantification of methane sources in the Athabasca oil sands region of Alberta by aircraft mass balance. *Atmos. Chem. Phys.* 18, 7361–7378. <https://doi.org/10.5194/acp-18-7361-2018>.
- Fiehn, A., Kostinek, J., Eckl, M., Klausner, T., Galkowski, M., Chen, J., Gerbig, C., Röckmann, T., Maazallahi, H., Schmidt, M., Korben, P., Nečki, J., Jagoda, P., Wildmann, N., Mallau, C., Bun, R., Nickl, A.-L., Jöckel, P., Fix, A., Roiger, A., 2020. Estimating CH₄, CO₂ and CO emissions from coal mining and industrial activities in the Upper Silesian Coal Basin using an aircraft-based mass balance approach. *Atmos. Chem. Phys.* 20, 12675–12695. <https://doi.org/10.5194/acp-20-12675-2020>.
- Frost, G.J., McKeen, S.A., Trainer, M., Ryerson, T.B., Neuman, J.A., Roberts, J.M., Swanson, A., Holloway, J.S., Sueper, D.T., Fortin, T., Parrish, D.D., Fehsenfeld, F.C., Flocke, F., Peckham, S.E., Grell, G.A., Kowal, D., Cartwright, J., Auerbach, N., Habermann, T., 2006. Effects of changing power plant NO_x emissions on ozone in the eastern United States: proof of concept. *J. Geophys. Res. Atmos.* 111 <https://doi.org/10.1029/2005JD006354>.
- Gordon, M., Li, S.M., Staebler, R., Darlington, A., Hayden, K., O'Brien, J., Wolde, M., 2015. Determining air pollutant emission rates based on mass balance using airborne measurement data over the Alberta oil sands operations. *Atmos. Meas. Tech.* 8, 3745–3765. <https://doi.org/10.5194/amt-8-3745-2015>.
- Grant, D., Zelinka, D., Mitova, S., 2021. Reducing CO₂ Emissions by Targeting the World's Hyper-Polluting Power Plants.
- Karion, A., Sweeney, C., Pétron, G., Frost, G., Michael Hardesty, R., Kofler, J., Miller, B. R., Newberger, T., Wolter, S., Banta, R., Brewer, A., Dlugokencky, E., Lang, P., Montzka, S.A., Schnell, R., Tans, P., Trainer, M., Zamora, R., Conley, S., 2013. Methane emissions estimate from airborne measurements over a western United States natural gas field. *Geophys. Res. Lett.* 40, 4393–4397. <https://doi.org/10.1002/grl.50811>.
- Karplus, V.J., Zhang, S., Almond, D., 2018. Quantifying coal power plant responses to tighter SO₂ emissions standards in China. *Proc. Natl. Acad. Sci. USA* 115, 7004–7009. <https://doi.org/10.1073/pnas.1800605115>.
- KEPCO, 2022. In: Korea Electric Power Statistics [WWW Document]. URL: https://home.kepco.co.kr/kepco/KO/ntcob/list.do?boardCd=BRD_000099&menuCd=FN05030103. (Accessed 15 April 2023).
- Kim, S., Huey, L.G., Stichel, R.E., Tanner, D.J., Crawford, J.H., Olson, J.R., Chen, G., Brune, W.H., Ren, X., Leshner, R., Wooldridge, P.J., Bertram, T.H., Perring, A., Cohen, R.C., Lefer, B.L., Shetter, R.E., Avery, M., Diskin, G., Sokolik, I., 2007. Measurement of HO₂NO₂ in the free troposphere during the intercontinental chemical transport experiment - North America 2004. *J. Geophys. Res. Atmos.* 112, 1–10. <https://doi.org/10.1029/2006JD007676>.
- Liggio, J., Li, S.M., Staebler, R.M., Hayden, K., Darlington, A., Mittermeier, R.L., O'Brien, J., McLaren, R., Wolde, M., Worthy, D., Vogel, F., 2019. Measured Canadian oil sands CO₂ emissions are higher than estimates made using internationally recommended methods. *Nat. Commun.* 10, 1–9. <https://doi.org/10.1038/s41467-019-09714-9>.
- Liu, F., Choi, S., Li, C., Fioletov, V.E., McLinden, C.A., Joiner, J., Krotkov, N.A., Bian, H., Janssens-Maenhout, G., Darmenov, A.S., da Silva, A.M., 2018. A new global anthropogenic SO₂ emission inventory for the last decade: a mosaic of satellite-derived and bottom-up emissions. *Atmos. Chem. Phys.* 18, 16571–16586. <https://doi.org/10.5194/acp-18-16571-2018>.
- Liu, F., Duncan, B.N., Krotkov, N.A., Lamsal, L.N., Beirle, S., Griffin, D., McLinden, C.A., Goldberg, D.L., Lu, Z., 2020. A methodology to constrain carbon dioxide emissions from coal-fired power plants using satellite observations of co-emitted nitrogen dioxide. *Atmos. Chem. Phys.* 20, 99–116. <https://doi.org/10.5194/acp-20-99-2020>.

- Liu, Z., Ciaïis, P., Deng, Z., Lei, R., Davis, S.J., Feng, S., Zheng, B., Cui, D., Dou, X., Zhu, B., Guo, Rui, Ke, P., Sun, T., Lu, C., He, P., Wang, Yuan, Yue, X., Wang, Yilong, Lei, Y., Zhou, H., Cai, Z., Wu, Y., Guo, Runtao, Han, T., Xue, J., Boucher, O., Boucher, E., Chevallier, F., Tanaka, K., Wei, Y., Zhong, H., Kang, C., Zhang, N., Chen, B., Xi, F., Liu, M., Bréon, F.-M., Lu, Y., Zhang, Q., Guan, D., Gong, P., Kammen, D.M., He, K., Schellnhuber, H.J., 2020. Near-real-time monitoring of global CO₂ emissions reveals the effects of the COVID-19 pandemic. *Nat. Commun.* 11, 5172. <https://doi.org/10.1038/s41467-020-18922-7>.
- López-Aparicio, S., Guevara, M., Thunis, P., Cuvelier, K., Tarrasón, L., 2017. Assessment of discrepancies between bottom-up and regional emission inventories in Norwegian urban areas. *Atmos. Environ.* 154, 285–296. <https://doi.org/10.1016/j.atmosenv.2017.02.004>.
- Nisbet, E., Weiss, R., 2010. Top-down versus bottom-up. *Science* 328, 1241–1243. <https://doi.org/10.1126/science.1189936>.
- Park, J., Choi, J., Moon, K., Kim, D., Kim, H.-J., Ahn, J., Lee, S., Seo, B.-K., Kim, J., Park, S., Kim, S., 2020. Application of chemical ionization mass spectrometry in airborne SO₂ observation on Hanseo Beechcraft 1900 D. *Asian J. Atmospheric Environ.* 14, 10.
- Peischl, J., Ryerson, T.B., Holloway, J.S., Parrish, D.D., Trainer, M., Frost, G.J., Aikin, K. C., Brown, S.S., Dubé, W.P., Stark, H., Fehsenfeld, F.C., 2010. A top-down analysis of emissions from selected Texas power plants during TexAQS 2000 and 2006. *J. Geophys. Res. Atmos.* 115 <https://doi.org/10.1029/2009JD013527>.
- Qu, Z., Henze, D.K., Worden, H.M., Jiang, Z., Gaubert, B., Theys, N., Wang, W., 2022. Sector-based top-down estimates of NO_x, SO₂, and CO emissions in East Asia. *Geophys. Res. Lett.* 49, e2021GL096009 <https://doi.org/10.1029/2021GL096009>.
- Republic of Korea, 2020. 9th Basic Plan for Power Supply and Demand.
- Ryerson, T.B., Buhr, M.P., Frost, G.J., Goldan, P.D., Holloway, J.S., Hübler, G., Jobson, B. T., Kuster, W.C., McKeen, S.A., Parrish, D.D., Roberts, J.M., Sueper, D.T., Trainer, M., Williams, J., Fehsenfeld, F.C., 1998. Emissions lifetimes and ozone formation in power plant plumes. *J. Geophys. Res. Atmos.* 103, 22569–22583. <https://doi.org/10.1029/98JD01620>.
- Tang, L., Qu, J., Mi, Z., Bo, X., Chang, X., Anadon, L.D., Wang, S., Xue, X., Li, S., Wang, X., Zhao, X., 2019. Substantial emission reductions from Chinese power plants after the introduction of ultra-low emissions standards. *Nat. Energy* 4, 929–938. <https://doi.org/10.1038/s41560-019-0468-1>.
- Tang, L., Xue, X., Qu, J., Mi, Z., Bo, X., Chang, X., Wang, S., Li, S., Cui, W., Dong, G., 2020. Air pollution emissions from Chinese power plants based on the continuous emission monitoring systems network. *Sci. Data* 7, 325. <https://doi.org/10.1038/s41597-020-00665-1>.
- Trainer, M., Ridley, B.A., Buhr, M.P., Kok, G., Walega, J., Hübler, G., Parrish, D.D., Fehsenfeld, F.C., 1995. Regional ozone and urban plumes in the southeastern United States: Birmingham, A case study. *J. Geophys. Res. Atmos.* 100, 18823–18834. <https://doi.org/10.1029/95JD01641>.
- Turnbull, J.C., Karion, A., Fischer, M.L., Faloona, I., Guilderson, T., Lehman, S.J., Miller, B.R., Miller, J.B., Montzka, S., Sherwood, T., Saripalli, S., Sweeney, C., Tans, P.P., 2011. Assessment of fossil fuel carbon dioxide and other anthropogenic trace gas emissions from airborne measurements over Sacramento, California in spring 2009. *Atmos. Chem. Phys.* 11, 705–721. <https://doi.org/10.5194/acp-11-705-2011>.
- Wang, J.L., Daniels, W.S., Hammerling, D.M., Harrison, M., Burmaster, K., George, F.C., Ravikumar, A.P., 2022. Multiscale methane measurements at oil and gas facilities reveal necessary frameworks for improved emissions accounting. *Environ. Sci. Technol.* 56, 14743–14752. <https://doi.org/10.1021/acs.est.2c06211>.
- Wong, G., Wang, H., Park, M., Park, J., Ahn, J.-Y., Sung, M., Choi, J., Park, T., Ban, J., Kang, S., Lee, T., Kim, Jongho, Seo, B.-K., Yu, J.-H., Kim, Jeongho, Woo, J.-H., Kim, S., 2024. Optimizing an airborne mass-balance methodology for accurate emission rate quantification of industrial facilities: a case study of industrial facilities in South Korea. *Sci. Total Environ.* 912, 169204 <https://doi.org/10.1016/j.scitotenv.2023.169204>.
- Wren, S.N., McLinden, C.A., Griffin, D., Li, S.-M., Cober, S.G., Darlington, A., Hayden, K., Mihele, C., Mittermeier, R.L., Wheeler, M.J., Wolde, M., Liggio, J., 2023. Aircraft and satellite observations reveal historical gap between top-down and bottom-up CO₂ emissions from Canadian oil sands. *PNAS Nexus* 2, pgad140. <https://doi.org/10.1093/pnasnexus/pgad140>.
- Xu, Y., 2011. Improvements in the operation of SO₂ scrubbers in China's coal power plants. *Environ. Sci. Technol.* 45, 380–385. <https://doi.org/10.1021/es1025678>.
- Zavala-Araiza, D., Lyon, D., Alvarez, R.A., Palacios, V., Harris, R., Lan, X., Talbot, R., Hamburg, S.P., 2015. Toward a functional definition of methane super-emitters: application to natural gas production sites. *Environ. Sci. Technol.* 49, 8167–8174. <https://doi.org/10.1021/acs.est.5b00133>.
- Zhang, X., Schreifels, J., 2011. Continuous emission monitoring systems at power plants in China: improving SO₂ emission measurement. *Energy Pol.* 39, 7432–7438. <https://doi.org/10.1016/j.enpol.2011.09.011>.