Two years of satellite-based carbon dioxide emission quantification at the world's largest coal-fired power plants

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12

13 Abstract

14 Carbon dioxide (CO₂) emissions from combustion sources are uncertain in many places 15 across the globe. Satellites have the ability to detect and quantify emissions from large CO₂ point 16 sources, including coal-fired power plants. In this study, we made observations with the PRecursore 17 IperSpettrale della Missione Applicativa (PRISMA) satellite imaging spectrometer and the Orbiting 18 Carbon Observatory-3 (OCO-3) instrument onboard the International Space Station at over 30 coal-19 fired power plants routinely between 2021-2022. CO₂ plumes were detected in 50% of acquired 20 PRISMA scenes, which is consistent with the combined influence of viewing parameters on detection 21 (solar illumination, surface reflectance) and unknown factors (like daily operational status). We 22 compare satellite-derived emission rates to in situ stack emission observations and find average agreement to within 27% for PRISMA and 30% for OCO-3, though more observations are needed to 23 robustly characterize the error. We highlight two examples of fusing PRISMA with OCO-2 and 24 OCO-3 observations in South Africa and India. For India, we acquired PRISMA and OCO-3 25

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observations on the same day and use the high spatial resolution capability of PRISMA (30 m spatial/pixel resolution) to partition relative contributions of two distinct emitting power plants to the net emission. Though an encouraging start, two years of observations from these satellites did not produce sufficient observations to estimate annual average emission rates within low (<15%) uncertainties. However, as the constellation of CO_2 -observing satellites is poised to significantly improve in the coming decade, this study offers an approach to leverage multiple observation platforms to better quantify and characterize uncertainty for large anthropogenic emission sources.

33

34 **1 Introduction**

35 Anthropogenic carbon dioxide (CO_2) emissions are dominated by strong discrete point sources: power and other industrial combustion are estimated to make up 59% of global 36 37 anthropogenic CO₂ emissions with transport, buildings, and other sources making up the remaining 38 20%, 9%, and 12%, respectively (Crippa et al., 2022). Fossil fuel combustion is the largest 39 contributor to warming trends globally since the pre-industrial era (IPCC, 2021). However, there 40 remains uncertainty in the total magnitude of emissions from these sectors as bottom-up emission 41 estimates rely on reported emission factors and activity data, which may vary in granularity and 42 quality across countries and provinces (Hong et al., 2017; Guan et al., 2017). Accurate CO₂ emission 43 quantification is important in light of the Paris Agreement, as participating countries must develop plans and report progress to reduce their country's greenhouse gas (GHG) emissions (UN, 2015). 44 45 Leveraging atmospheric measurements, particularly satellite remote sensing, can help reduce uncertainty in facility-level CO₂ emission estimates, provided that the observations are accurate and 46 47 sufficiently sample the facility in time (Hill and Nassar, 2019). Deployed systematically with robust 48 error characterization, this system could be an anchor towards assessing and verifying anticipated 49 CO₂ emission reductions as part of national and global GHG emission reduction plans and 50 agreements.

51 Several studies have shown that atmospheric sounding satellites can accurately quantify some 52 point source CO₂ emissions from large individual coal-fired power plants. First, the Orbiting Carbon 53 Observatory-2 (OCO-2; Crisp et al., 2017) is a space-based instrument that observes solar 54 backscattered near-infrared radiance in the oxygen A band (758-772 nm; 0.04 nm spectral resolution), 55 the weak CO2 band (1594-1619 nm; 0.08 nm spectral resolution), and strong CO2 band (2042-2082 56 nm; 0.10 nm spectral resolution). OCO-2 views in nadir mode over land, while sun glint mode 57 increases the signal over water giving measurements both land and water, and target mode to target 58 specific validation or calibration sites. With its 10-km wide swath, $\leq 1.3 \times 2.25$ km² pixel resolution, 59 and better than 1.0 ppm precision for retrievals of the column-mean dry-air mole fraction of CO_2 60 (XCO_2) (Taylor et al., 2023), OCO-2 is sensitive to single CO₂ point sources that emit sufficiently 61 close to an OCO-2 orbital track and are spatially isolated from other major CO₂ sources. Using 62 satellite observations from OCO-2, Nassar et al. (2017) detected strong CO₂ enhancements in the 63 near vicinity of seven large coal-fired power plants and employed a Gaussian plume model emission 64 quantification technique to estimate emission rates for these facilities. Further study expanded the set 65 of facilities that could be quantified by OCO-2 (Nassar et al., 2021). Other studies have leveraged the nitrogen dioxide (NO₂) retrieval capability and wide swath of the TROPOspheric Monitoring 66 67 Instrument (TROPOMI; van Geffen et al., 2020) to attribute and corroborate strong CO₂ signals seen 68 in OCO-2 observations (Hakkarainen et al., 2021; Reuter et al., 2019). The Orbiting Carbon 69 Observatory-3 (OCO-3; Eldering et al., 2019), the flight spare of OCO-2, has been on board the 70 International Space Station (ISS) since May 2019. Like OCO-2, it has been shown capable of 71 quantifying CO₂ power plant emissions. Nassar et al. (2022) analyzed nine successful OCO-3

72 acquisitions of the Belchatów Power Station and found the variability in satellite-based emission 73 estimates agreed well with the variability in independently reported hourly power generation. Guo et 74 al., (2023) estimated emissions at Chinese power plants using OCO-2/3 and found close agreement 75 with emission inventories. OCO-3 is different than OCO-2 in that it has a two-axis Pointing Mirror 76 Assembly (PMA) for more agile pointing, allowing it to rapidly point off-nadir and take Snapshot 77 Area Mapping (SAM) mode observations over the course of two minutes. These SAMs are approximately 80×80 km² collections of measurements and are typically over sites of interest 78 79 including cities, power plants, volcanoes, and flux towers.

80 Another class of remote sensing imaging spectrometers – sometimes also referred to as 81 hyperspectral imagers – also have been shown capable of detecting and quantifying strong CO₂ 82 signals from large point sources. Thorpe et al. (2017) flew the Next-Generation Airborne/Infrared 83 Imaging Spectrometer (AVIRIS-NG) over a coal-fired power plant in Four Corners, New Mexico, 84 and detected strong CO₂ plumes. AVIRIS-NG observes a large range of solar backscattered radiance 85 (380-2500 nm), but at much coarser spectral resolution (5 nm), and high spatial resolution (e.g., 3 m 86 when flown at 3 km altitude). The much finer spatial resolution of AVIRIS-NG allows for improved visualization of the origin of a CO₂ plume, but at the expense of fine precision for a single observed 87 88 CO₂ column. Still, Cusworth et al. (2021) analyzed a combination of AVIRIS-NG and the identically 89 built Global Airborne Observatory (GAO) at over 20 power plants in the U.S., quantified emission 90 rates, and found close agreement with continuous emissions monitoring (CEMS) hourly emission 91 observations. From space, the PRecursore IperSpettrale della Missione Applicativa (PRISMA), 92 launched in 2019, like AVIRIS-NG and GAO is sensitive to a large range of solar backscattered 93 radiance (400-2500 nm), albeit at coarser spectral and spatial resolution (10 nm spectral resolution; 94 30 m spatial resolution; Loizzo et al., 2018). PRISMA is a tasked satellite instrument potentially capable of hundreds of 30×30 km² observations per day, with equatorial crossing time of 10:30am, and target revisit of seven days, though true revisit depends on tasking priorities of the system. Cusworth et al. (2021) showed a few examples of CO₂ plumes detected and quantified with PRISMA, with quantified emissions similar in magnitude to reported CEMS emissions..

99 The capacity for satellites to be leveraged as useful tools for reducing uncertainty in the global 100 CO₂ anthropogenic emission sector requires synthesis and routine observations (i.e., tasking) of a 101 critical number of facilities. Therefore, in this study, we made observations at a subset of global coal-102 fired power plants routinely over the course of two years to probe detection limits, emission 103 quantification uncertainty, and data yields. We observed these facilities with both OCO-3 and 104 PRISMA. To our knowledge to date, this study represents the largest satellite-based facility scale 105 investigation of direct CO₂ emission quantification across a diverse set of global power plants, and 106 the first investigation to assess the capability of PRISMA to reliably detect and quantify CO₂ point 107 sources. The results, though not sufficient by themselves to reduce uncertainty relative to bottom-up 108 inventories significantly on an annual basis, show a path forward for data fusion and synthesis of 109 observations from the growing constellation of planned CO₂ sensing satellites.

110

111 **2 Methods**

Table 1 lists the locations of all power plants we targeted during this study between 2021-2022 with PRISMA. OCO-3 includes a subset of these sites as well as other fossil fuel combustion sites as part of its list of possible targets. We identified coal-fired power plants to routinely target using a combination of bottom-up and top-down information. Bottom-up coal-fired power plant CO₂ emission estimates rely on activity data, that usually includes permitted capacity of a power plant and its operational state; and emission factors, usually estimated from the composition of the coal 118 that is combusted. Inventories, like the Global Energy Monitor (GEM), include this data for a large 119 set of coal-fired power plants across the globe (GEM, 2023). From the GEM database, we gathered 120 the top 10 largest bottom-up coal-fired power plants globally. We then gathered a list of top-down 121 TROPOMI NO₂ combustion hotspots, as inferred by Beirle et al. (2021). We included an additional 122 seven unique power plants using this dataset. Because the imaging scene size of PRISMA is 30×30 123 km², some adjacent smaller power plants were imaged simultaneously along with these larger power 124 plants. In total, outside of the U.S., we made PRISMA observations at 27 power plants. In the U.S., 125 we chose 10 power plants to routinely target using reported EPA CEMS information 126 (campd.epa.gov): five of the top 30 emitting power plants, and five progressively lower emitters, 127 chosen so that we could assess satellite detection capabilities.

128

Power Plant Name	Countr y	Latitude	Longitude	Number clear-sky observatio ns	Number plume detections	Minimum quantified CO2 emission (kt CO2 h ⁻¹)	Mean quantified CO2 emission (kt CO2 h ⁻¹)	Maximum quantified CO2 emission (kt CO2 h ⁻¹)
Mundra- Adani	India	22.82	69.55	12	7	0.49 ± 0.07	1.09±0.19	1.76±0.32
Korba-Balco	India	22.40	82.74	5	1	NA*	NA	NA
PLN Paiton Baru	Indoneis a	-7.71	113.57	4	2	NA	NA	NA
Craig	USA	40.46	-107.59	5	5	$0.56{\pm}0.11$	0.69±0.16	0.8±0.22
Cumberland	USA	36.39	-87.65	1	0	NA	NA	NA
Dry Fork	USA	44.39	-105.46	6	3	0.61±0.09	0.65±0.13	0.69±0.16
H L Spurlock	USA	38.70	-83.82	5	3	1.15±0.32	1.26±0.39	1.37±0.45
Ulsan Hanju (1)	South Korea	35.49	129.33	1	0	NA	NA	NA
Hasdeo	India	22.41	82.69	5	0	NA	NA	NA
Hekinan	Japan	34.83	136.96	6	4	$0.72{\pm}0.47$	3.88±1.09	8.35±2.14
Baotou-1	China	40.66	109.66	5	2	0.19±0.07	0.27±0.07	0.35±0.07
Kendal	South Africa	-26.09	28.97	7	2	0.85±0.13	0.85±0.13	0.85±0.13
NTPC Korba	India	22.39	82.68	6	1	1.28 ± 0.27	1.28±0.27	1.28±0.27

129 Table 1. Power plants that were targeted specifically by PRISMA in this study.

Kriel	South Africa	-26.25	29.18	8	3	0.74±0.15	0.82±0.15	0.95±0.16
Labadie	USA	38.56	-90.84	4	4	0.73±0.18	0.73±0.18	0.73±0.18
Martin Lake	USA	32.26	-94.57	8	8	1.45±0.31	2±0.59	2.6±0.98
Matimba	South Africa	-23.67	27.61	11	8	0.33±0.05	0.72±0.16	1.14±0.32
Matla	South Africa	-26.28	29.14	8	3	0.33±0.05	0.77±0.15	1.37±0.27
Medupi	South Africa	-23.71	27.56	15	12	0.33±0.06	0.83±0.19	1.47±0.34
Mundra-Tata	India	22.82	69.53	12	5	0.38±0.09	0.74±0.13	$1.32{\pm}0.21$
Niederausse m	German y	51.00	6.67	1	0	NA	NA	NA
Oregon	USA	41.67	-83.44	5	1	NA	NA	NA
Paiton-3	Indonesi a	-7.71	113.58	4	4	1.54±0.37	3.16±0.69	4.78±1.02
Rihand	India	24.03	82.79	8	5	0.83±0.17	0.99±0.26	$1.36{\pm}0.38$
Sanfeng	China	40.66	109.76	6	0	NA	NA	NA
Sasan	India	23.98	82.63	9	7	0.65±0.15	1.01±0.24	1.51±0.31
Sooner	USA	36.45	-97.05	6	3	1.05±0.22	1.05±0.22	1.05±0.22
Togtoh	China	40.20	111.36	2	2	0.25±0.06	0.91±0.17	1.58±0.27
Ulsan Hanju (2)	South Korea	35.47	129.38	1	0	NA	NA	NA
Vindhyachal	India	24.10	82.68	9	7	0.33±0.1	0.72±0.15	1.24±0.23
Waigaoqiao	China	31.36	121.60	6	1	NA	NA	NA
Yeosu Hanwha	South Korea	34.84	127.69	2	0	NA	NA	NA
Yosu	South Korea	34.83	127.67	2	0	NA	NA	NA
Al Zour	Kuwait	28.71	48.37	12	0	NA	NA	NA

*A value of "NA" indicates that no plumes were detected at this power plant or that the emission quantification
 algorithm (described in Methods) failed to quantify an emission rate.

132

133 2.1 PRISMA observations and quantification

PRISMA is a tasked satellite instrument, capable of collecting around $200 \ 30 \times 30 \ \text{km}^2$ targets per day and has 20° pointing capability off nadir. Authenticated users can program single observation requests through PRISMA's web portal (prisma.asi.it), which currently allows for 13 concurrent requests at a time per user. We specified two-week observing windows for each request, and configured requests to collect if the scene-averaged solar zenith angle (SZA) was less than 70° and forecast meteorology anticipated less than 20% cloud cover. If the orbital configuration, weather,
SZA align and there are no other conflicting or higher priority requests, PRISMA images a target.

141 For each acquired PRISMA image, we performed XCO₂ retrievals on all pixels within a 2.5 142 km radius around the power plant. We retrieve XCO₂ using the Iterative Maximum A Posteriori – 143 Differential Optical Absorption Spectroscopy (IMAP-DOAS) algorithm, as implemented in 144 Cusworth et al. (2021). This approach estimates XCO_2 by decomposing an observed radiance 145 spectrum into high and low frequency features between 1900-2100 nm. For high-frequency features, 146 we simulate atmospheric transmission of CO₂, H₂O, and N₂O using a Beer-Lambert approximation. 147 For low-frequency features (e.g., surface reflectance, aerosol scattering), we use an 8-degree 148 polynomial. The forward model that drives IMAP-DOAS therefore has the following form:

149
$$F^{h}(\mathbf{x}) = I_{0}(\lambda) \exp\left(-\sum_{n=1}^{6} s_{n} \sum_{l=1}^{72} A_{l} \tau_{n,l}\right) \sum_{k=0}^{K} a_{k} \lambda^{k} \quad (1)$$

Where F^h is simulated backscattered radiance at wavelength λ , I_0 is incident solar intensity, A_l is the 150 151 airmass factor at vertical level $l \in [1,72]$, $\tau_{n,l}$ is the optical depth for each trace gas element, s_n is the 152 scaling factor applied to the optical depth, and a_k is a polynomial coefficient (K=8). Optical depths 153 are computed by querying meteorological information for pressure and temperature from the 154 MERRA-2 reanalysis (Gelaro et al., 2017), and using that information to select proper HITRAN 155 absorption cross sections for each trace gas (Kochanov et al., 2016). To compare the model from Equation 1 against PRISMA observed radiance (y), we compute $F^{h}(\mathbf{x})$ between 1900-2100 nm at 156 157 0.02 nm resolution, then convolve the output using the PRISMA full-width half maximum, and 158 sample at PRISMA wavelength positions. This results in vector F(x) that is comparable to y. The 159 vector \mathbf{x} , also called the state vector, includes scale factors for CO₂, H₂O, N₂O, and polynomial fficients: V - (c c 160

160 coefficients:
$$\mathbf{x} = (s_{CO2}, s_{H2O}, s_{N2O}, a_0, ..., a_8).$$

161 XCO₂ is retrieved from PRISMA radiance using a Bayesian optimal estimation approach 162 (Rodgers, 2000). Here, the optimized state vector solution, or posterior, is solved through Gauss-163 Newton iteration:

164
$$\mathbf{x}_{i+1} = \mathbf{x}_{A} + (\mathbf{K}_{i}^{T} \mathbf{S}_{0}^{-1} \mathbf{K}_{i} + \mathbf{S}_{A}^{-1})^{-1} \mathbf{K}_{i}^{T} \mathbf{S}_{0}^{-1} [y - \mathbf{F}(\mathbf{x}_{i}) + \mathbf{K}_{i} (\mathbf{x}_{i} - \mathbf{x}_{A})]$$
(2)

Where $\mathbf{S}_{O} = [\mathbf{\epsilon}\mathbf{\epsilon}^{T}]$ is the observation error covariance matrix defined by the instrument signal to noise ratio (SNR), \mathbf{x}_{A} is the prior estimate of the state vector, and \mathbf{S}_{A} is the prior error covariance matrix. The matrix **K**, or Jacobian matrix, represents the first derivative of the $\mathbf{F}(\mathbf{x})$ with respect to the state vector:

169
$$\mathbf{K}_{i} = \frac{\partial \mathbf{F}}{\partial \mathbf{x}}\Big|_{\mathbf{x}=\mathbf{x}_{i}}$$
(3)

170 The posterior error covariance matrix can be computed explicitly to quantify retrieval precision:

171
$$\hat{\mathbf{S}} = \left(\mathbf{K}_i^T \mathbf{S}_0^{-1} \mathbf{K}_i + \mathbf{S}_A^{-1}\right)^{-1} \quad (4)$$

Across the scenes we acquired with PRISMA, using this retrieval approach, we quantify an average 3.3 ppm precision for an XCO_2 column. Absolute biases in PRISMA XCO_2 retrievals are less important for CO_2 plume detection and quantification: systematic retrieval biases are removed from a scene through the quantification and removal of a local background, as described below. To characterize bias in emission quantification, we compare emission rates derived from PRISMA to stack-level CEMS measurements (Section 3.2).

We quantified emissions for each PRISMA plume detection using the Integrated Mass Enhancement (IME) approach (Cusworth et al., 2021). However, we updated the masking scheme for this analysis to produce more reliable masks for each CO₂ plume. Figure 1 shows the plume masking procedure for a plume detected at the Hekinan, Japan power plant on July 19, 2021. First, we apply a background threshold to differentiate candidate plume pixels from the background (method to quantify background threshold described in Section 3.2). We then group enhanced XCO₂ pixels into clusters of at least 20 connected pixels. These groups are then buffered with a one-pixel dilation filter to smooth edges and any gaps that exist in a group (Dougherty, 1992). Finally, each cluster is considered part of the plume if at least one of its pixels is within 500 m of an exhaust stack.

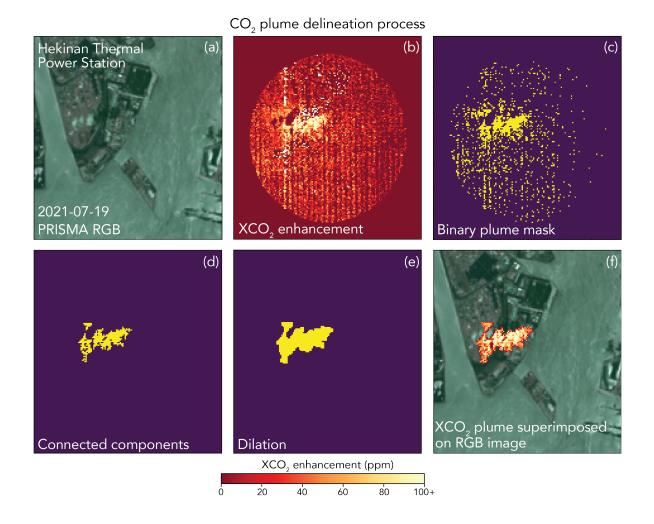


Figure 1. Example of the plume delineation and masking process performed on XCO₂ retrievals derived from PRISMA observations. Panel (a) shows the simultaneously observed RGB PRISMA imagery, panel (b) shows retrieved XCO₂ above the background, panels (c)-(e) show the plume masking procedure to isolate enhanced pixels and remove noise, and panel (f) shows the resulting CO₂ plume superimposed on the RGB imagery.

193

194 IME is calculated for a plume using the following equation:

$$IME = \sum_{i=1}^{N} \Delta \Omega_i \Lambda_i$$
 (5)

196 where $\Delta \Omega_i$ is the XCO₂ mass enhancement in pixel *i* relative to background (kg m⁻²), Λ_i is the pixel 197 area (900 m²), and *N* is the number of pixels in the plume. The CO₂ emission rate *Q* is estimated from 198 the IME using the following relationship:

199
$$Q = \frac{U_{eff}}{L} \text{ IME (6)}$$

where $L = \sqrt{\sum_{i=1}^{N} \Lambda_i}$ is the plume length and U_{eff} is the effective wind speed. The parameter *L* is an 200 201 operational parameter that needs to be related to the extent of the plume. Since a plume dissipates in 202 all directions due to turbulent diffusion, an explicit scaling function (i.e., an effective wind speed 203 U_{eff}) that relates L and 10 m wind speed (U_{10}) to the true emission can be derived through large eddy 204 simulations (Varon et al., 2018): The effective wind speed relates IME and plume length 205 parameterizations to true emission rates. This relationship can be empirically estimated through large 206 eddy simulations using the 10-m wind speed (U_{10}). Here we apply the U_{eff} relationship derived from 207 Varon et al., 2018:

$$U_{eff} = 1.1 \log U_{10} + 0.6. \quad (7)$$

where U_{eff} and U_{10} are in units of [m s⁻¹]. We query the ERA5-Land reanalysis using the Open-Meteo Application Programming Interface (open-meteo.com), which provides hourly wind information globally at 0.1° spatial resolution (Muñoz-Sabater et al., 2021). Uncertainty due to winds is calculated by generating an ensemble of U_{10} values assuming 50% error (Cusworth et al., 2021). Uncertainty due to the CO₂ background is calculated by generating many emission estimates and calculating a standard deviation using an ensemble of background thresholds. Background thresholds are set to vary with scene-averaged CO₂ retrieval precision. Total emission uncertainty is estimated by adding in quadrature the contribution of wind and background uncertainties.

- 217
- 218 2.2 OCO-3 observations and quantification

219 OCO-3 is also a tasked mission: it can take SAMs over any place of interest within the latitude 220 range of the ISS orbit (about 52° S to 52° N). In addition to the SAM locations we supplied to OCO-221 3 to overlap with PRISMA targets, there are many other power plant and fossil fuel combustion 222 sources that make up its set of mission targets. However, unlike PRISMA, OCO-3 does not consider 223 cloud forecasts, snow cover, or viewing geometry when planning SAMs and thus the majority of 224 observations fail to produce useful maps of XCO₂. Additionally, aerosol- and albedo-induced XCO₂ 225 artifacts are present in many SAMs (Bell et al., 2023) and thus make the detection of plumes even 226 more difficult.

227 For all cloud-free soundings, OCO-3 XCO2 concentrations are derived using the 228 Atmospheric Carbon Observations from Space (ACOS; O'Dell et al., 2012; Crisp et al., 2012; O'Dell 229 et al., 2018) v10 optimal estimation retrieval, which employs the Levenberg-Marquardt modification 230 of the Gauss-Newton method. In this work, bias corrected XCO₂ from the OCO-3 Lite files is used 231 but the official data quality flag is not applied. This was done because often the quality flag removes 232 XCO₂ retrievals within the plume and makes emission estimation more difficult or impossible 233 (Nassar et al., 2022). For SAMs where we visually identified CO₂ plumes (e.g., Figure 2), emission 234 rates are estimated using two approaches: (1) a Gaussian plume model and (2) the IME method. For 235 the Gaussian plume model approach, we follow the algorithm outlined in Nassar et al. (2022):

236
$$V(x,y) = \frac{Q}{\sqrt{2\pi}\sigma_y(x)u} e^{-(\frac{1}{2})(\frac{y}{\sigma_y(x)})^2}$$
(8)

237
$$\sigma_y(x) = a \cdot \left(\frac{x}{x_o}\right)^{0.894} \tag{9}$$

238 Where V represents the vertical columns within the plume (g/m^2) , Q is the CO₂ emission rate (g/s), 239 y is the wind direction perpendicular to the plume (m), u is the wind speed at the height of the plume at its midline (m/s) assuming plume rise of 250 m above the stack height, $\sigma_v(x)$ is the standard 240 deviation of the y-direction, x_o is a characteristic plume length (1000 m), and a is a stability 241 242 parameter (Nassar et al., 2021). Following Nassar et al. (2022), wind speed and direction inputs are 243 estimated by taking the average of ERA-5 (Bell et al., 2020) and MERRA-2 reanalysis data. The wind direction is optimized by rotating the plume, typically between -30° to 30° away from the mean 244 245 ERA-5/MERRA-2 direction, and calculating the correlation coefficient (R) of the modeled and 246 observed XCO_2 . The optimized wind direction is the direction that produces the largest R. The 247 background is typically estimated by averaging OCO-3 footprints within a radius of 30 km, excluding 248 the plume itself and a narrow 3 km buffer zone. However, if there are visible artifacts in the XCO_2 249 background that are unrelated to the power plant plume, the background field is modified to avoid 250 them. For example, decreasing the radius of footprints used from 30 km to 20 km. The uncertainty 251 in wind speed is calculated by taking the difference of the emission estimate using two different 252 models (ERA-5 and MERRA2). The background concentration uncertainty is calculated by 253 estimating Q using three different background radii of 30, 40, and 50 km. Q is also calculated for a 254 30 km radius background but only using the left and right halves of the background, relative to the 255 direction of the plume. The standard deviation of both these methods is calculated and the larger of 256 the two is the background uncertainty. The plume rise uncertainty is calculated by estimating Q using 257 plume rise values of 100, 200, 250, 300, and 400 m and taking the standard deviation of those values.

Total uncertainty on the emission rate Q using the Gaussian plume method is estimated by adding in quadrature the contribution of wind speed, background concentration, and plume rise uncertainties.

260 To obtain another estimate of emission rate, we also apply an IME quantification approach to 261 the CO₂ plumes, which to our knowledge is the first time the IME method has been applied to OCO-3 SAMS at coal power plants. We first interpolate the XCO₂ retrievals in a SAM to a uniform 2×2 262 263 km^2 grid to account for occasional OCO-3 footprint overlap. Similar to Varon et al. (2018), 3×3 264 pixel neighborhoods are sampled and the distributions are compared to the background using a 265 Student's *t*-test. The default confidence level for the *t* test is 95% but this is often lowered to visually 266 capture most of the plume. The plume is then smoothed using a 3×3 pixel median filter and a 267 Gaussian filter with a standard deviation of 0.5. The U_{eff} calculation is done using an equation 268 approximately equal to Equation 7 ($U_{eff} = 1.0 \log U_{10} + 0.55$). Other recent studies have used various 269 methods (Lin et al., 2023; Brunner et al., 2023), but further research is needed to determine the most 270 accurate way to estimate U_{eff} for an OCO-3-like instrument. The wind direction is the optimized direction determined by the Gaussian plume model. The background XCO₂ estimate is taken from 271 272 the Gaussian plume model methodology and the plume is typically required to be within 50 km 273 downwind and 8 km crosswind of the source, although these parameters are modified if the plume 274 curves outside of the 8 km crosswind threshold or there are XCO₂ artifacts that should be avoided.

The uncertainty for the IME method is estimated similarly to the Gaussian plume model method. The uncertainty in wind speed is calculated by taking the standard deviation of the emission estimates using wind speed from two different models (ERA-5 and MERRA2). The background concentration uncertainty is calculated by estimating Q using the different backgrounds calculated in the Gaussian plume model method: a 20 km radius, 30 km radius, 40 km radius, left half, full circle, and right half. The standard deviation of the three radii estimates and left half, full circle, and right half estimates are calculated and the larger of the two is the background uncertainty. Uncertainty of the Student's *t*-test confidence level is also estimated. The confidence level and -10% and +10% of the confidence level are used to find Q. For example, if the confidence level needed to visually capture the XCO₂ plume is 85%, Q is calculated for 75%, 85%, and 95% and the standard deviation of those three values represents the confidence level uncertainty. Total uncertainty on the emission rate Q using the IME method is estimated by adding in quadrature the contribution of wind speed, background concentration, and Student's *t*-test confidence level uncertainties.

Figure 2 shows IME methodology successfully identifying an XCO₂ plume from an OCO-3
SAM taken over the Colstrip power plant.

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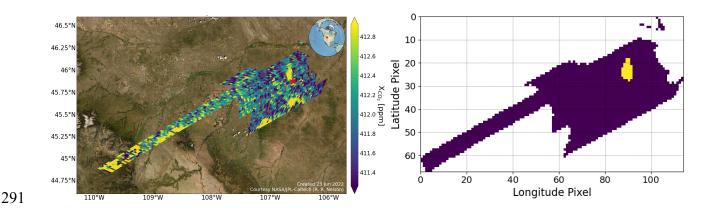


Figure 2. IME plume identification approach applied to an example OCO-3 SAM at the Colstrip power plant on 18 August 2021. Left panel: OCO-3 SAM bias corrected XCO₂. Right panel: yellow pixels indicate the final plume mask.

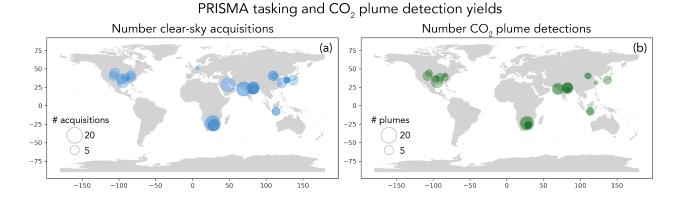
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3 Results

297 *3.1 Global yields from two years of observations*

298 Figure 3a shows a global map of power plants we targeted with PRISMA, with the marker 299 for each power plant's location (latitude, longitude) scaled to represent the number of successful 300 acquisitions between 2021-2022. In total, we acquired 181 PRISMA images, which corresponds to 301 314 unique power plant observation scenes. Of these scenes, 210 were of sufficient quality to attempt 302 CO₂ retrieval and plume detection, with quality mostly determined by visual inspection for clouds 303 and haze. Of these 210 scenes, 104 were determined to have CO₂ plumes (Figure 3b). Scenes were 304 marked as containing CO₂ plumes through inspection of XCO2 and visible imagery: if a large cluster 305 of pixels with elevated XCO₂ above the background were also in the vicinity of a power plant exhaust 306 stack, an analyst would mark the scene as containing a CO₂ plume. Routine tasking observations with 307 PRISMA resulted in an average of 6 acquisitions for each power plant (maximum 15), roughly one 308 image acquired per quarter. Of these acquisitions, plumes were detected on average four times per 309 facility (maximum 12).

For OCO-3, 1363 power plant SAMs were taken during September 2019 to December 2022. Of these, 139 XCO₂ plumes emanating from power plants were visually identified. However, only 14 were for power plants that were also observed by PRISMA and have CEMS validation (nine Colstrip cases, two Martin Lake cases, and three Craig cases). The acquisition rates are low relative to PRISMA because OCO-3 does not account for scene favorability when planning its SAMs. For example, OCO-3 took 66 Colstrip SAMs from 2019-2022 yet only yielded nine high-quality XCO₂ plume cases.



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Figure 3. Data yields from PRISMA continually between 2021-2022. Panel (a) shows the number of clear-sky acquisitions for each power plant. Panel (b) shows the number of plumes detected by an analyst for each of the observed power plants.

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322 The low observed average detection rate of CO_2 plumes is a result of three primary factors: 323 (1) observing conditions at each facility including solar zenith angle (SZA) and surface reflectance; 324 (2) local meteorology; and (3) operational status at each power plant at the time of acquisition. To 325 test how well these factors predict the presence of a plume for PRISMA, we fit a logistic regression 326 classification function with a sparse (L1) penalty to our dataset (Fan et al., 2008). This algorithm fits 327 a logit function to the plume detection outcome of each scenes (i.e., detected plume = TRUE, no 328 detected plume = FALSE) using a set of predictor variables that are likely candidates to explain 329 prediction results. In this setup, the statistical model is fit using the following predictor variables – 330 SZA, U_{10} , average single-sounding retrieval precision across the scene, annual bottom-up emission 331 estimate for the power plant using GEM, and average observed radiance between 1900-2100 nm 332 within the scene normalized by the cosine of the SZA. This last factor is a simple proxy for surface 333 reflectance, although it does not take into account other factors that influence radiance observations 334 (e.g., water vapor, aerosols, other atmospheric constituents). We split the data so that 50% was used 335 to train the model and 50% was reserved as a test set. The predictor variables were all standardized 336 by their mean and standard deviation before the model was fit. The results of classification can be 337 summarized using two statistics: precision (ratio of true positives to sum of true positives and false 338 positives) and recall (ratio of true positives to sum of true positives and false negatives). The results 339 of fitting a logistic regression model to the data show minor prediction performance, with precision 340 = 0.60 and recall = 0.69 for positive plume detection. The regression coefficients are shown in Figure 4a. The coefficient with the highest weight is normalized radiance. Figure 4b shows SZA against 341 342 normalized radiance, with red dots indicating no plume detection and blue dots representing positive 343 plume detection. Though no clear separation exists, there is a cluster of no plume detection at high 344 SZA and low normalized radiance. This is a consistent and expected relationship, as SZA and surface 345 reflectance are principal drivers of the quantity of light that is observed by the satellite, and therefore 346 SNR of the observation.

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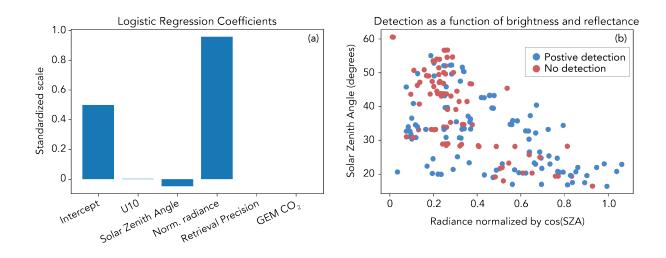


Figure 4. CO₂ plume prediction using various atmospheric, retrieval, and bottom-up information.
Panel (a) shows the results of fitting a logistic regression classification model to the set of PRISMA

- acquisitions where an analyst identified the presence or lack of a plume. Panel (b) shows the top two
 explanatory variables (SZA and normalized radiance) along with plume classification.
- 353

354 The logistic regression model performed better on the test data set than predictions made at 355 random, though the prediction performance was still low. Missing from the model is sub-annually 356 resolved information regarding operating status. For most of the power plants outside the U.S., we 357 do not have information on daily operations of a power plant. In many cases of non-detects, we could simply be observing a power plant temporarily not in operation. Another possibility is that at the time 358 359 of acquisition, some power plants were operating at reduced capacity, meaning that CO₂ emission 360 rates were lower than those predicted by annual emission factors or activity data. If the true CO_2 361 emission rate was below the minimum detection limit (MDL) possible by the PRISMA satellite, then 362 it would show as a non-detect. However, even if the emission were near or slightly above the 363 PRISMA MDL, the probability of detection would still be low as slight variations in atmospheric 364 properties, as seen in Figure 4, would then influence the ability to detection a CO₂ plume.

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366 *3.2 Validation of PRISMA and OCO-3 emission rates against CEMS*

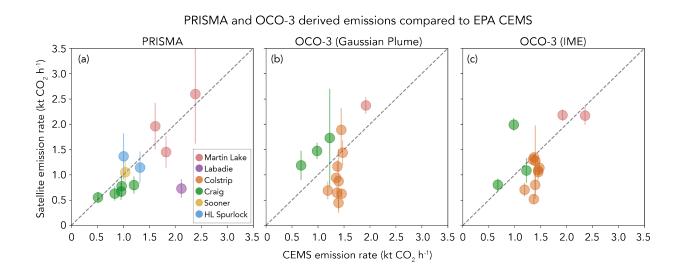
For each power plant where a CO₂ plume was identified, we quantify emissions using the IME approach described by Equations 5-7. In order to estimate the XCO₂ mass enhancement ($\Delta\Omega$ in Equation 1), a local background must be quantified and subtracted from total XCO₂ retrievals across the scene. To do this, we apply a concentration threshold β to initiate the plume masking and segmentation process (described in Methods section). Once we have a plume mask, we apply another concentration threshold γ to the remaining XCO₂ pixels that exist outside of the plume. This value γ represents the XCO₂ background that we use to calculate the XCO₂ enhancement that is used in the IME formulation of Equation 1. Thresholds β and γ largely influence the magnitude of the emission rate and are not known a priori. For global generalizability, we wish to estimate β and γ such that they do not vary across power plants, seasons, regions, etc. Therefore, we parameterize β and γ as percentiles under the assumption that the local contrast between enhanced CO₂ plume pixels and the background should be similar across PRISMA scenes.

379 To estimate values for β and γ , we compare EPA CEMS data for power plants in the U.S. 380 with estimated emission rates from PRISMA. In total, we have 12 scenes in the U.S. with CEMS 381 information that pertain to 5 power plants. We then optimize β and γ such that the output of an 382 ordinary least squares regression produces a slope near unity. Figure 5a shows the results of this 383 optimization which produces an optimal β percentile of 94% and a γ percentile of 62%. The results also show decent correlation between CEMS data and PRISMA-derived emission rates ($R^2 = 0.43$). 384 385 A single outlier at the Labadie power plant (imaged July 10, 2022) shows the largest discrepancy from CEMS data (69%), but the remaining plumes show average 27% relative difference from CEMS 386 data. If we remove the one data point at Labadie, the R^2 improves to 0.75. Though a limited sample 387 size, between PRISMA and OCO-3, these scenes represent variability in solar geometries (20-40° 388 389 SZA), surface reflectance (0.09-0.90 normalized radiance), and reported emission rates (0.51 - 2.39)390 kt CO2 h⁻¹). Therefore, we use these optimal parameters and apply them to our global dataset of 391 PRISMA detections. These emission rates are reported in Table 1. There are some instances when 392 performing IME emission calculations using these thresholds and plume masking technique do not 393 result in emission rates (e.g., the plume masking procedure produces a mask with no pixels). In these 394 cases, we report a detection but not an emission quantification.

Figures 5b and 5c shows the comparison between OCO-3 and CEMS at some power plants that overlap with PRISMA observations (14 scenes total). OCO-3 Gaussian plume model emission

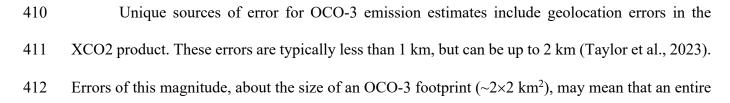
rates (Fig. 5b) have an improved correlation compared to PRISMA ($R^2 = 0.51$), although with greater bias (average 47% relative difference from CEMS). The OCO-3 IME estimates (Fig. 5c) have worse R^2 (0.32) but a better RMSE (0.45 kt CO₂/hr) compared to the Gaussian plume model estimates (0.84 kt CO₂/hr), with 9 of the 14 cases being within 30% of the reported CEMS emission and an average relative difference of 30% for all 14 cases. Additionally, the least squares fit for IME is closer to the 1-to-1 line than for the Gaussian plume model.





404

Figure 5. Comparison of emission rates in the U.S. between satellite-derived estimates and CEMS information. Panel (a) shows a comparison between PRISMA derived emission rates and CEMS (R^2 = 0.43), panel (b) shows a comparison between OCO-3 and CEMS using the Gaussian plume model (R^2 = 0.51), and panel (c) shows a comparison between OCO-3 and CEMS using IME (R^2 = 0.32).

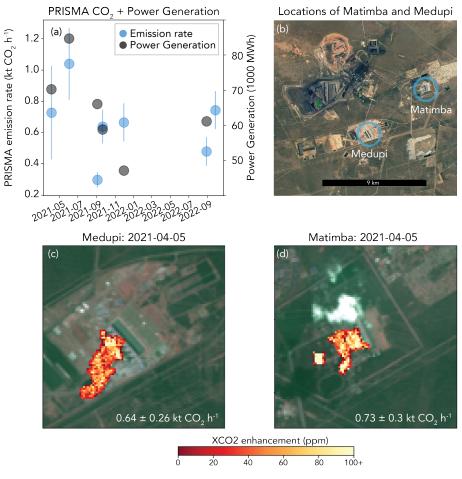


footprint is not included when estimating emissions using the Gaussian plume method, which assumes that the plume only extends downwind of the known source location. The Gaussian plume model is also susceptible to wind direction errors, and requires the plume to be Gaussian in shape, which is often not true. IME, while not suffering from wind direction or geolocation-induced errors, assumes that the entire plume is captured in a given SAM, which is sometimes not true and results in an underestimation of emissions. IME is also sensitive to errors in U_{eff} parameterization.

419

420 3.3 Comparison and fusion of PRISMA and OCO

421 Outside the U.S., PRISMA observed the Matimba power station in South Africa 11 times and 422 quantified emission rates 7 times. Emissions from Matimba have previously been quantified and 423 validated using OCO-2 (Hakkarainen et al., 2021). This station does not report hourly emission rates, 424 but does report daily power generation (Eskom, 2023). Though not a direct comparison, we can use 425 this information to check if the emission quantification approach we describe above captures some 426 variability in activity at this power plant. Figure 6a shows the emission rates we quantified compared 427 against reported power generation. We see rough agreement in variability - the high power 428 generation reported between Apr to July 2021 (70000-85000 MWh) drop for subsequent dates (47000-66000 MWh) between Sep 2021 to Sep 2022, a drop which is also seen in the PRISMA-429 430 derived CO2 emission rate. Across all observations, we estimate an emission rate range of 0.30-1.04 431 kt CO2 h⁻¹ (average 0.66 kt CO₂ h⁻¹). This average emission rate is substantially lower than the average 2.50 kt CO₂ h⁻¹ emission rate estimated from OCO-2 and TROPOMI between 2018-2020, 432 but within the range of emissions estimates directly quantified with OCO-2 (0.30-7.20 kt CO_2 h⁻¹; 433 Hakkarainen et al., 2021). However, this discrepancy could be result of (1) changes in activity or 434 435 variability or (2) existence of other nearby emission sources. For changes in activity, during August 436 2020, the Matimba reported a large range of power generation (65000-94000 MWh) and emission 437 estimates derived directly from OCO-2 were also highly variable (0.88-4.33 kt CO₂ h⁻¹). Given that 438 maximum power generation at the time of a PRISMA observation was 85000 MWh, some of the 439 discrepancy in maximum CO₂ quantification between PRISMA and OCO-2 could be due to activity. 440 Nearby (7 km) the Matimba Power Station is the Medupi Power Plant (Figure 6b). Figure 6c 441 show the Medupi CO₂ plume observed during the same PRISMA overpass on Apr 5, 2021. The PRISMA derived emission rate for Medupi is 0.64 ± 0.26 kt CO2 h⁻¹ and for Matimba is 0.73 ± 0.30 442 443 kt CO₂ h⁻¹. Given the proximity of the two power plants, the higher derived emission rate reported 444 for Matimba from previous studies could actually be a result of a net emission from these two 445 facilities. The OCO-2 flight track is located tens of kilometers downwind from Matimba and Medupi, 446 making a clear delineation between potentially co-emitted distinct emission plumes near impossible. 447 If we sum emission rates from both Medupi and Matimba, we quantify a range of 0.89-1.73 kt CO₂ h^{-1} (1.30 ± 0.28 kt CO2 h^{-1}), which is still lower, but closer to the average emissions quantified by 448 449 OCO-2.



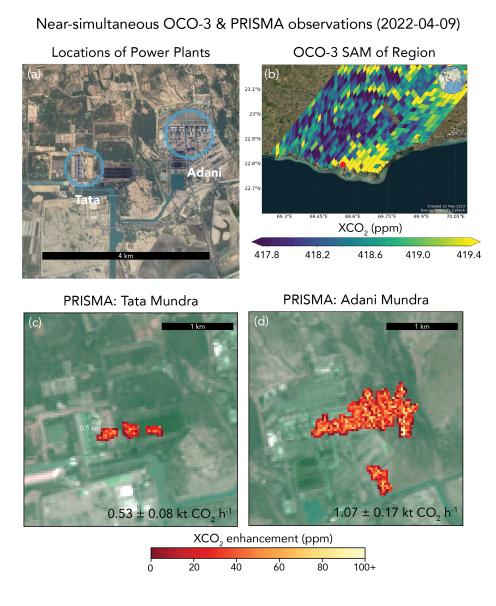
Quantification results at Matimba and Medupi Power Plants

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Figure 6. Emission rates and reported power generation at the Matimba and Medupi power plants in South Africa. Panel (a) shows the CO₂ emission rates derived from PRISMA and the reported daily power generation for the day of PRISMA overpass. Panel (b) shows the locations of the Medupi and Matimba power plants (base imagery provided by Google Earth; © Google Earth 2023). Panels (c) and (d) show plume imagery and emission rates for a PRISMA overpass on Apr 5, 2021.

The ability to differentiate the contribution of unique point sources to a regional total is an application made possible by joint observing of imaging spectrometers and atmospheric sounders. Figure 7 shows observations that were made at the Tata Mundra Ultra Mega Power Plant and the

460	Adani Mundra Thermal Power Project: two power plants less than 3 km apart. Both OCO-3 and
461	PRISMA imaged the power plants on Apr 9, 2022. Figure 7b shows the OCO-3 SAM (taken 04:41
462	UTC) – large CO_2 enhancements appear along the coastline likely associated with emission from
463	these power plants. PRISMA imaged the power plants less than two hours later (06:02 UTC) and
464	detected CO2 plumes at each facility (Figure 7b-c). The OCO-3 derived emission rate using Gaussian
465	plume approaches is 5.5 ± 0.7 kt CO ₂ h ⁻¹ , but the emission rate derived using the IME approach is
466	much lower (3.0 kt CO_2 h ⁻¹). For this case, the IME approach may be more appropriate as the shape
467	of the OCO-3 plume (Figure 7b) is more diffuse in nature and does not visibly resemble a Gaussian
468	structure. The PRISMA emission rate for the Adani plant is 1.07 ± 0.17 kt CO ₂ h ⁻¹ and for the Tata
469	Mundra plant is 0.53 ± 0.08 kt CO ₂ h ⁻¹ . We can use this information to estimate that 67% of the net
470	CO ₂ emission came from Adani, and the remaining 33% came from the Tata plant. The combined
471	emission rate $(1.60 \pm 0.25 \text{ kt CO}_2 \text{ h}^{-1})$ is lower than the OCO-3 IME emission rate. Like the Matimba
472	power plant, some of this discrepancy may partially be explained by bias or uncertainty in retrievals,
473	background, and wind information. Also, lower estimates of CO2 emissions from PRISMA are
474	consistent with the fact that PRISMA is only sensitive to emissions at two exhaust stacks, while the
475	OCO-3 observation includes all CO2 sources in the industrial area around Mundra. Continued
476	validation of retrieved emission rates against ground standards (e.g., CEMS) will help better quantify
477	bias and uncertainty. However, even with lingering uncertainty, the near simultaneous observations
478	of OCO-3 and PRISMA can help us disentangle the relative contributions from each power plant.
479	



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Figure 7. Near-simultaneous observation of two power plants in Mundra, India on Apr 9, 2022. Panel (a) shows the locations of two power plants spaced less than 3 km apart: Tata Mundra and Adani Mundra Power Stations (base imagery provided by Google Earth; © Google Earth 2023). Panel (b) shows the OCO-3 SAM with a red dot showing the location of the power plants. Panel (c) and (d) show the PRISMA acquisition (less than 2 hours after OCO-3) over the two power plants with associated emission rates.

488 Conclusion

489 We observed a global set of power plants for two years between 2021-2022 with both 490 PRISMA and OCO-3 to test the ability of these satellite platforms to do routine operational 491 monitoring of CO₂ emissions. When PRISMA observations were of sufficient quality to perform 492 XCO_2 retrievals, we detected CO_2 plumes nearly half of the time. We fit a logistic regression 493 classification using plume detections and find that there is some relationship between SZA and 494 surface reflectance that partially explains plume prediction; consistent given that these factors are 495 major drivers of SNR. The remaining non-plume detections may be due to operational status of a 496 power plant at the time of observation. We compared emission rates from both PRISMA and OCO-497 3 to power plants in the U.S. where we have access to hourly *in situ* CEMS emission information. 498 We find significant correlation between satellite and *in situ* estimates, though some significant biases 499 may exist for some of the observations for both PRISMA and OCO-3. Also, the quantity of CEMS 500 observations was limited (~10 for each instrument), so robust calibration is not yet possible. Still, 501 early results show that under the right conditions, satellites can provide reliable estimates of CO₂ 502 emissions at discrete point source locations. This is consistent with the close agreement between 503 airborne imaging spectrometer emissions and CEMS information (Cusworth et al., 2021).

Fusion of information from atmospheric sounders like OCO-3 and imaging spectrometers like PRISMA is valuable for cross-validation and source attribution. We see this particularly for our examples at the Matimba and Medupi power plants in South Africa and the Tata and Adani power plants in Mundra, India. In these cases, and particularly at Mundra where near-simultaneous PRISMA and OCO-3 observations were taken, OCO-2/3 provides a local, but coarse resolution emission constraint for a complex of facilities that emit large CO₂ quantities. PRISMA, with its 30 m pixel resolution, then can help refine relative contributions of single emitters against the net 511 emission flux. More work is needed to refine cross-validation between instruments, but initial 512 observation shows one avenue for data from multiple observing systems to be complementary 513 aggregated and analyzed.

514 Even when combining information from both satellites, there is still too little sampling to 515 constrain facility emissions within low uncertainties. Cusworth et al. (2021), using arguments from 516 Hill and Nassar (2019), suggested that nearly 30 unbiased observations from a PRISMA-class 517 instrument is needed per year at each power plant to reduce annual uncertainties below 14% (i.e., 518 reduce emission uncertainty from Non-Annex I countries below 1 Gt CO₂ per year). No power plant 519 in this study met this minimum sampling requirement. However, there will be a significant increase 520 in data volumes and observation performance of satellite remote sensing capabilities for CO2, from 521 both recently launched and planned imaging spectrometers including EMIT (launched 2022; Thorpe 522 et al., in revision); EnMAP (launched 2022; Guanter et al., 2015); Carbon Mapper/Tanager 1-2 523 (Planned launch 2024; Duren et al., 2021), and atmospheric sounders including CO2M (Sierk et al., 524 2019). Improved observation of global power plants and emission quantification with robust error 525 characterization will be vital to reduce global uncertainty of anthropogenic emissions from fossil fuel 526 combustion sources.

527

528 Data Availability.

The OCO-3 XCO2 and other retrieval properties are publicly available at the NASA Goddard Earth Science Data and Information Services Center (GES-DISC). The full suite of retrieval products in the standard per-orbit format can be obtained at OCO Science Team et al., 2021, https://doi.org/10.5067/D9S8ZOCHCADE. The lightweight per-day format data (Lite files), which includes the bias corrected estimates of XCO2, can be obtained at OCO Science Team et al., 2022,

534	https://doi.org/10.5067/970BCC4DHH24. PRISMA data including radiance for each scene and
535	XCO2 retrievals is available at https://doi.org/10.5281/zenodo.8083596.
536	
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541	
542	Author Contributions. DHC designed the study. DHC, AKA, RJ tasked and acquired PRISMA
543	data. DHC performed PRISMA emission quantification and validation. RRN performed OCO-3
544	quantification and validation. RN and JPM helped implement OCO-3 quantification algorithms. All
545	authors provided feedback on results and the manuscript.
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547	
548	Competing interests. The authors declare no conflicts of interest.
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