



*Supplement of*

## **Performance validation of the GHGSat methane constellation via controlled releases**

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## **S1 Controlled releases at the Carbon Management Canada facility**

Controlled release measurements were conducted at Carbon Management Canada's field research station, located near Brooks, Alberta, Canada, shown in Fig. S1. The surrounding landscape is mostly flat open grassland. Compressed natural gas (CNG) was transported to the controlled release site using a trailer that was outfitted with 16 high-pressure cylinders. Each canister has a seamless aluminum liner and is entirely overwrapped with high-strength carbon fibre embedded in an epoxy resin matrix allowing for a pressure rating of 3,600 PSIG. After each trailer refill the CNG composition was analyzed at the Isotope Science Lab and the University of Calgary. The released gas consisted of an average of ~94% CH<sub>4</sub> by volume, with remaining constituents being ethane (~3.5% by volume), propane (~1% by volume), and other minor components (~1.5% by volume) such as heavier hydrocarbons, N<sub>2</sub>, and O<sub>2</sub>.

The gas was released from the pressurized cylinder via a regulator valve. The temperature of the expanding gas decreases due to the Joule–Thomson effect; the gas temperature was conditioned to atmospheric temperature by means of a heat exchanger. The release rate was controlled by adjusting the pressure regulator and measured using the FloBoss™ 103 (a calibrated flow computer) coupled with a McCrometer v-cone flow meter (½” and 1”, depending on the desired flow rate). The flow rate was visually inspected and logged digitally during the releases. Finally, the conditioned CNG was vented to atmosphere via a modular stack at two heights: 3.4 m and 4.8 m. All components were connected using high-pressure red polyurethane jacketed flowlines constructed of electrically conductive nylon core and steel braided reinforcement layer. The local meteorological conditions were measured using a portable 81000-L RM Young 3D ultrasonic anemometer, situated 2.25 m above ground.

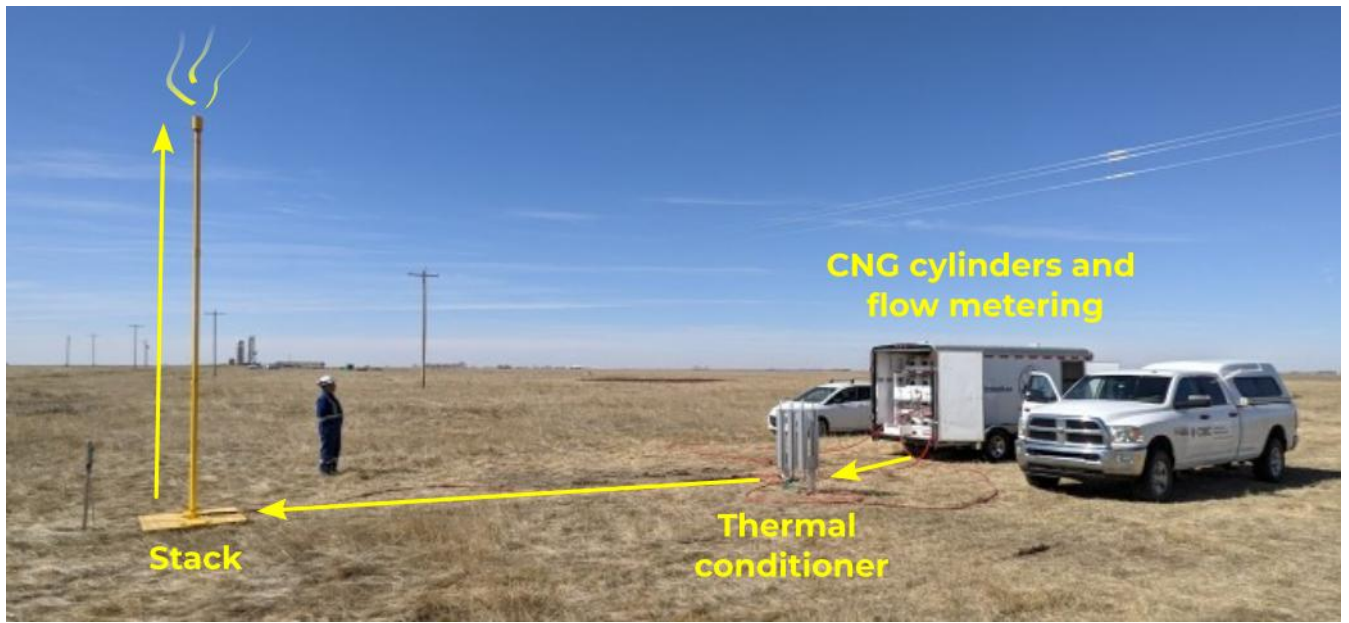


**Figure S1.** Overview of the controlled release set-up. Visible are the yellow-painted modular stack (left), the conditioner (centre), and the trailer housing the cylinders with CNG, pressure regulators, and flow meters (right). All components are connected using high-pressure gas lines (in red).

## S2 Height-adjusted wind speed

The IME method estimates emission rates based on an effective wind speed function that is calibrated for an input wind at  $z_{\text{ref}} = 10$  m above ground. In most controlled release campaigns considered in this study, the anemometer was already deployed at the 10 m reference height: no adjustment is required and the measured wind is used directly. However, two campaigns measured below 10 m: the GHGSat/CMC campaigns at Brooks and the FluxLab campaign at Petrolia. In those cases, the measured wind is adjusted to the reference height assuming a logarithmic profile:

$$u(z_{\text{ref}}) = u(z_{\text{ane}}) \frac{\ln((z_{\text{ref}} - d)/z_0)}{\ln((z_{\text{ane}} - d)/z_0)}, \quad (S1)$$

where  $z_0$  is the roughness length and  $d$  is the zero-plane displacement. Table S1 lists, for each campaign, the anemometer height  $z_{\text{ane}}$ , and surface parameters used to scale the measured wind to the 10 m reference height. Surface parameters were queried from the global wind atlas (<https://globalwindatlas.info>) at the coordinates of the corresponding facility.

**Table S1. Anemometer heights and parameters of the logarithmic wind profile for each campaign**

| Dates           | Organizer     | Location        | Anemometer height (m) | Roughness length $z_0$ (m) | Displacement $d$ (m) |
|-----------------|---------------|-----------------|-----------------------|----------------------------|----------------------|
| 2021-03–2025-07 | GHGSat/CMC    | Brooks, AB      | 2.25                  | 0.03                       | 0.02                 |
| 2021-10–2021-11 | Stanford      | Ehrenberg, AZ   | 10.0                  | 0.01                       | 0.007                |
| 2022-10–2022-11 | Stanford      | Casa Grande, AZ | 10.0                  | 0.01                       | 0.007                |
| 2024-08–2024-10 | Stanford      | Evanston, WY    | 10.0                  | 0.03                       | 0.02                 |
| 2024-11–2024-12 | Stanford      | Casa Grande, AZ | 10.0                  | 0.01                       | 0.007                |
| 2025-01–2025-12 | Stanford      | Casa Grande, AZ | 10.0                  | 0.01                       | 0.007                |
| 2025-05–2025-06 | FluxLab       | Petrolia, ON    | 7.0*                  | 0.03                       | 0.02                 |
| 2025-06–2025-06 | TADI/Stanford | Lacq, France    | 10.0                  | 0.5                        | 0.333                |

\* Data from the 7m anemometer is not available on 2025-05-12, and a 2m anemometer was used instead for this specific date.

### S3 Retrieval algorithm and noise model

The analysis toolchain that processes raw data (L0) to infer methane concentration (L2) uses a variant of differential optical absorption spectroscopy (DOAS) inversion methods, where measurement data is fit to a mathematical forward model that combines the solar irradiance, atmospheric absorption, ground reflectivity, and instrument components. The model and retrieval procedure was described in details in Ref. (Jervis et al., 2021) and we hereby recapitulate the key concepts and introduce a residuals-based error model as a useful metric of retrieval quality which is used in assessing detection probability.

Following the notation from Rodgers (2000), we express the problem of inferring a state vector of atmospheric and instrument parameters  $\mathbf{x}$  which we seek to estimate from a vector of measurements  $\mathbf{y}$  (light irradiance within the passband of the Fabry-Pérot) via the equation

$$\mathbf{y} = \mathbf{F}(\mathbf{x}) + \hat{\mathbf{e}}, \quad (\text{S2})$$

where  $\mathbf{F}(\mathbf{x})$  is a nonlinear forward model which describes light propagation in a given state, and  $\hat{\mathbf{e}}$  is the vector of errors (residuals) which is the sum of measurement noise  $\boldsymbol{\epsilon}_y$  and imperfections of the forward model  $\boldsymbol{\epsilon}_F$ . More precisely, the forward model expresses the amount of photocurrent sensed by a sensor pixel  $(i, j)$  by integrating the fraction of the top-of-atmosphere spectral radiance  $L_{\text{TOASR}}$  which is transmitted by the instrument (order sorting filter transmission  $T_{\text{OSF}}$  and Fabry-Perot transmission  $T_{\text{FP}}$ ), and converted to electrical signal given the camera spectral responsivity  $R_C$ :

$$F_{i,j}(\mathbf{x}) = \int L_{\text{TOASR}}(\lambda) \cdot T_{\text{OSF}}(\lambda) \cdot T_{\text{FP}}(\theta_{i,j}, \lambda) \cdot R_C(\lambda) d\lambda. \quad (\text{S3})$$

The TOASR is obtained by integrating Beer-Lambert’s law through the downwelling and upwelling limbs of the light path, using known solar irradiance as the boundary condition (Chandrasekhar, 2011; Jervis et al., 2021). Evaluating Eq. (S3) is too expensive computationally to be solved at every ground cell of a spatially resolved retrieval. Instead, we use a two-step procedure: (1) a scene-wide average retrieval using the full forward model to estimate the scene-wide average state vector  $\hat{\mathbf{x}}_{\text{SR}}$ , and (2) a per-cell retrieval done using a linearized version of Eq. (S3) around  $\hat{\mathbf{x}}_{\text{SR}}$ .

We infer the state vector’s best estimate  $\hat{\mathbf{x}}$  via nonlinear optimization, minimizing a cost function that is the sum of squared residuals, weighted by the measurement uncertainty

$$\chi^2(\mathbf{x}) = \hat{\mathbf{e}}^T \mathbf{W} \hat{\mathbf{e}}, \quad (S4)$$

where  $\mathbf{W} = \mathbf{S}_{y,m}^{-1}$  is the inverse of the measurement covariance matrix, estimated from known variances of photocurrent shot noise  $\sigma_{psn}^2$ , dark current shot noise  $\sigma_{dsn}^2$ , and camera read noise  $\sigma_{rn}^2$

$$\mathbf{S}_{y,m} = \text{diag}(\sigma_{psn}^2 + \sigma_{dsn}^2 + \sigma_{rn}^2). \quad (S5)$$

The subscript  $m$  in  $\mathbf{S}_{y,m}$  stands for “model”, indicating that it is derived from physical modelling prior to performing the inversion. An alternative, posterior estimate of the parameter covariance matrix can be formulated based on the residuals of the regression (identified by the subscript  $r$ ):

$$\mathbf{S}_{y,r} = \frac{1}{M - N} \frac{\hat{\mathbf{e}}^T \mathbf{W} \hat{\mathbf{e}}}{\text{Tr}(\mathbf{W})/M} \mathbf{I}, \quad (S6)$$

where  $M$  is the number of measurements and  $N$  is the number of retrieved parameters (length of the  $\mathbf{y}$  and  $\mathbf{x}$  vectors respectively). Error estimates in measurement space can be converted to state space via the equation

$$\hat{\mathbf{S}}_{x,r|m} = (\mathbf{K}^T \mathbf{W} \mathbf{K})^{-1} \mathbf{K}^T \mathbf{W} \mathbf{S}_{y,r|m} \mathbf{W}^T \mathbf{K} (\mathbf{K}^T \mathbf{W} \mathbf{K})^{-1}, \quad (S7)$$

where  $\mathbf{K} = \frac{\partial \mathbf{F}(\hat{\mathbf{x}})}{\partial \mathbf{x}}$  is the Jacobian matrix of the forward model around the solution  $\hat{\mathbf{x}}$ . This equation applies to both the model and residuals-based errors, and covariance matrices  $\hat{\mathbf{S}}_x$  and  $\mathbf{S}_y$  can take either of the subscripts  $r$  or  $m$  as noted by the vertical bar.

The covariance matrix  $\hat{\mathbf{S}}_{x,r}$ , specifically, the square root of its diagonal element corresponding to the methane column density, is the noise estimate  $n$  used in the PoD model throughout this article. This metric provides a more accurate and conservative estimate of the local noise than its “modeled” counterpart derived from  $\mathbf{S}_{x,m}$ . The latter accounts only for the noise sources that can be predicted before the retrieval is performed, namely shot noise and readout noise. Because the forward model is imperfect, the true retrieval noise is generally higher, making the modelled value a lower bound and the residual-based value the more reliable of the two for our purpose.

A third approach to estimating local noise, also considered here, is the local standard deviation of the column density, computed within a radius of the origin (Ramier et al., 2023). While this approach is in principle more robust to correlated noise, it requires that all variation within the region of interest be attributable to noise alone, and is therefore not applicable in the presence of an actual methane plume, such as in the context of controlled releases. In summary, of the three candidate noise estimates considered here, the residual-based one is the only estimate that both reflects the noise actually encountered in a retrieval and remains valid when a plume is present. We therefore select it for the purpose of modeling the detection performance.

## S4 Extended binary regression results

Table S2 and Table S3 report the full set of fitted models: four predictor functions, each combined with four inverse-link functions, using either the complete set of driving variables  $\{q, u, n, a\}$  or a subset of it. For each combination, Table S2 lists the fitted parameters (offset  $\mu$  or  $m$ , scale  $\sigma$  or  $s$ , and optional wind offset  $u_0$ ) and the goodness of fit metrics (Akaike information criterion AICc and Bayesian information criterion BIC). Table S3 lists the inferred detection limits ( $Z_{50}$ ,  $Q_{50}$ ,  $Q_{90}$ ) and transition width ( $\Delta Q_{80}$ ).

Both tables report two independent estimates of the uncertainty on each quantity. The first,  $\sigma_{bs}$ , is obtained by bootstrap resampling of the release dataset, following the procedure described in the main text, and therefore reflects the sampling variability of the underlying observations. The second,  $\sigma_{hess}$ , follows from standard error propagation: the covariance matrix of the fitted parameters is taken as the inverse of the Hessian of the negative log likelihood evaluated at the optimum, and uncertainties on the derived detection metrics are obtained by linear propagation from that matrix. Both metrics produce uncertainty estimates that are highly compatible. In particular, the detection limit  $Q_{50}$  for the selected model (linear wind predictor, log-normal link function, all 4 variables) is  $99.0 \pm 5.2$  using the Hessian, and  $99.0 \pm 5.3$  using bootstrapping.

**Table S2. Probability of detection models: model parameters at the maximum likelihood (opt) and associated uncertainties computed using the Hessian of the likelihood ( $\sigma_{\text{hess}}$ ) and bootstrap ( $\sigma_{\text{bs}}$ ). Goodness of fit metrics AICc and BIC are also listed.**

| Predictor  | Link         | Variables | Offset<br>(opt) | Offset<br>( $\sigma_{\text{hess}}$ ) | Offset<br>( $\sigma_{\text{bs}}$ ) | Scale<br>(opt) | Scale<br>( $\sigma_{\text{hess}}$ ) | Scale<br>( $\sigma_{\text{bs}}$ ) | $u_0$ (m s <sup>-1</sup> )<br>(opt) | $u_0$ (m s <sup>-1</sup> )<br>( $\sigma_{\text{hess}}$ ) | $u_0$ (m s <sup>-1</sup> )<br>( $\sigma_{\text{bs}}$ ) | AICc | BIC  |
|------------|--------------|-----------|-----------------|--------------------------------------|------------------------------------|----------------|-------------------------------------|-----------------------------------|-------------------------------------|----------------------------------------------------------|--------------------------------------------------------|------|------|
| Heuristic  | Log-normal   | {q}       | 1.167           | 0.072                                | 0.076                              | 0.426          | 0.091                               | 0.077                             |                                     |                                                          |                                                        | 85.6 | 91.1 |
| Heuristic  | Log-normal   | {q,a}     | 1.151           | 0.077                                | 0.081                              | 0.472          | 0.098                               | 0.095                             |                                     |                                                          |                                                        | 88.3 | 93.9 |
| Heuristic  | Log-normal   | {q,n}     | 1.162           | 0.071                                | 0.076                              | 0.397          | 0.087                               | 0.079                             |                                     |                                                          |                                                        | 80.7 | 86.3 |
| Heuristic  | Log-normal   | {q,u}     | 1.083           | 0.086                                | 0.087                              | 0.489          | 0.095                               | 0.091                             |                                     |                                                          |                                                        | 79.0 | 84.6 |
| Heuristic  | Log-normal   | {q,u,n}   | 1.069           | 0.076                                | 0.085                              | 0.424          | 0.081                               | 0.078                             |                                     |                                                          |                                                        | 71.1 | 76.7 |
| Heuristic  | Log-normal   | {q,u,a}   | 1.062           | 0.091                                | 0.091                              | 0.525          | 0.103                               | 0.098                             |                                     |                                                          |                                                        | 81.0 | 86.6 |
| Heuristic  | Log-normal   | {q,n,a}   | 1.151           | 0.068                                | 0.071                              | 0.390          | 0.082                               | 0.084                             |                                     |                                                          |                                                        | 78.9 | 84.5 |
| Heuristic  | Log-normal   | {q,u,n,a} | 1.061           | 0.069                                | 0.071                              | 0.373          | 0.073                               | 0.068                             |                                     |                                                          |                                                        | 66.9 | 72.5 |
| Heuristic  | Log-logistic | {q,u,n,a} | 1.060           | 0.069                                | 0.082                              | 0.214          | 0.045                               | 0.044                             |                                     |                                                          |                                                        | 67.2 | 72.8 |
| Heuristic  | Weibull      | {q,u,n,a} | 1.238           | 0.069                                | 0.071                              | 0.362          | 0.076                               | 0.074                             |                                     |                                                          |                                                        | 67.6 | 73.1 |
| Heuristic  | Fréchet      | {q,u,n,a} | 0.896           | 0.067                                | 0.073                              | 0.301          | 0.057                               | 0.053                             |                                     |                                                          |                                                        | 68.0 | 73.6 |
| Linear     | Log-normal   | {q,u,n,a} | 0.540           | 0.225                                | 0.248                              | 0.267          | 0.055                               | 0.050                             | 2.289                               | 1.215                                                    | 1.694                                                  | 60.4 | 68.7 |
| Linear     | Log-logistic | {q,u,n,a} | 0.551           | 0.223                                | 0.266                              | 0.155          | 0.034                               | 0.031                             | 2.197                               | 1.157                                                    | 1.858                                                  | 60.9 | 69.2 |
| Linear     | Weibull      | {q,u,n,a} | 0.669           | 0.240                                | 0.273                              | 0.258          | 0.053                               | 0.055                             | 2.304                               | 1.265                                                    | 1.910                                                  | 61.5 | 69.8 |
| Linear     | Fréchet      | {q,u,n,a} | 0.396           | 0.193                                | 0.232                              | 0.206          | 0.042                               | 0.037                             | 2.437                               | 1.076                                                    | 1.662                                                  | 60.4 | 68.7 |
| Quadrature | Log-normal   | {q,u,n,a} | 0.819           | 0.130                                | 0.152                              | 0.275          | 0.056                               | 0.052                             | 2.499                               | 0.782                                                    | 1.099                                                  | 61.7 | 70.0 |
| Quadrature | Log-logistic | {q,u,n,a} | 0.826           | 0.135                                | 0.165                              | 0.160          | 0.038                               | 0.033                             | 2.423                               | 0.812                                                    | 1.142                                                  | 62.2 | 70.5 |
| Quadrature | Weibull      | {q,u,n,a} | 0.941           | 0.144                                | 0.167                              | 0.263          | 0.057                               | 0.058                             | 2.584                               | 0.831                                                    | 1.190                                                  | 62.5 | 70.8 |
| Quadrature | Fréchet      | {q,u,n,a} | 0.691           | 0.107                                | 0.140                              | 0.215          | 0.043                               | 0.039                             | 2.538                               | 0.651                                                    | 1.429                                                  | 62.1 | 70.4 |
| Rice       | Log-normal   | {q,u,n,a} | 0.851           | 0.120                                | 0.183                              | 0.278          | 0.056                               | 0.095                             | 2.003                               | 0.573                                                    | 0.957                                                  | 62.2 | 70.4 |
| Rice       | Log-logistic | {q,u,n,a} | 0.858           | 0.124                                | 0.169                              | 0.162          | 0.035                               | 0.036                             | 1.948                               | 0.602                                                    | 0.873                                                  | 62.7 | 70.9 |
| Rice       | Weibull      | {q,u,n,a} | 0.973           | 0.131                                | 0.174                              | 0.264          | 0.059                               | 0.066                             | 2.077                               | 0.620                                                    | 0.880                                                  | 62.8 | 71.1 |
| Rice       | Fréchet      | {q,u,n,a} | 0.725           | 0.098                                | 0.220                              | 0.218          | 0.043                               | 0.077                             | 2.018                               | 0.475                                                    | 1.339                                                  | 62.7 | 71.0 |

**Table S3. Probability of detection models: derived detection performance metrics at the maximum likelihood (opt) and associated uncertainties computed using the Hessian of the likelihood ( $\sigma_{\text{hess}}$ , for  $Q_{50}$  and  $Q_{90}$  only) and bootstrap ( $\sigma_{\text{bs}}$ ).**

| Predictor  | Link         | Variables | $Z_{50}$<br>(opt) | $Z_{50}$<br>( $\sigma_{\text{bs}}$ ) | $Q_{50}$<br>(kg h <sup>-1</sup> )<br>(opt) | $Q_{50}$<br>(kg h <sup>-1</sup> )<br>( $\sigma_{\text{hess}}$ ) | $Q_{50}$<br>(kg h <sup>-1</sup> )<br>( $\sigma_{\text{bs}}$ ) | $Q_{90}$<br>(kg h <sup>-1</sup> )<br>(opt) | $Q_{90}$<br>(kg h <sup>-1</sup> )<br>( $\sigma_{\text{hess}}$ ) | $Q_{90}$<br>(kg h <sup>-1</sup> )<br>( $\sigma_{\text{bs}}$ ) | $\Delta Q_{80}$<br>(kg h <sup>-1</sup> )<br>(opt) | $\Delta Q_{80}$<br>(kg h <sup>-1</sup> )<br>( $\sigma_{\text{bs}}$ ) |
|------------|--------------|-----------|-------------------|--------------------------------------|--------------------------------------------|-----------------------------------------------------------------|---------------------------------------------------------------|--------------------------------------------|-----------------------------------------------------------------|---------------------------------------------------------------|---------------------------------------------------|----------------------------------------------------------------------|
| Heuristic  | Log-normal   | {q}       | 3.21              | 0.24                                 | 104.3                                      | 7.5                                                             | 7.8                                                           | 180.1                                      | 21.6                                                            | 16.7                                                          | 119.6                                             | 21.0                                                                 |
| Heuristic  | Log-normal   | {q,a}     | 3.16              | 0.25                                 | 102.6                                      | 7.9                                                             | 8.2                                                           | 188.0                                      | 22.9                                                            | 21.5                                                          | 131.9                                             | 26.8                                                                 |
| Heuristic  | Log-normal   | {q,n}     | 3.20              | 0.24                                 | 103.8                                      | 7.3                                                             | 7.7                                                           | 172.6                                      | 19.5                                                            | 14.8                                                          | 110.1                                             | 20.3                                                                 |
| Heuristic  | Log-normal   | {q,u}     | 2.95              | 0.26                                 | 96.7                                       | 8.3                                                             | 8.4                                                           | 181.1                                      | 22.0                                                            | 22.7                                                          | 129.4                                             | 26.3                                                                 |
| Heuristic  | Log-normal   | {q,u,n}   | 2.91              | 0.24                                 | 95.3                                       | 7.3                                                             | 8.0                                                           | 164.2                                      | 17.3                                                            | 15.7                                                          | 108.8                                             | 19.4                                                                 |
| Heuristic  | Log-normal   | {q,u,a}   | 2.89              | 0.26                                 | 94.7                                       | 8.6                                                             | 8.7                                                           | 185.7                                      | 24.6                                                            | 25.4                                                          | 137.4                                             | 29.0                                                                 |
| Heuristic  | Log-normal   | {q,n,a}   | 3.16              | 0.22                                 | 102.6                                      | 6.9                                                             | 7.2                                                           | 169.2                                      | 18.4                                                            | 16.0                                                          | 106.9                                             | 22.1                                                                 |
| Heuristic  | Log-normal   | {q,u,n,a} | 2.89              | 0.20                                 | 94.6                                       | 6.5                                                             | 6.7                                                           | 152.7                                      | 15.7                                                            | 15.0                                                          | 94.1                                              | 18.0                                                                 |
| Heuristic  | Log-logistic | {q,u,n,a} | 2.89              | 0.27                                 | 94.5                                       | 6.5                                                             | 8.7                                                           | 151.2                                      | 16.6                                                            | 15.7                                                          | 92.1                                              | 19.7                                                                 |
| Heuristic  | Weibull      | {q,u,n,a} | 3.02              | 0.23                                 | 98.9                                       | 7.7                                                             | 7.5                                                           | 152.6                                      | 13.3                                                            | 14.1                                                          | 102.6                                             | 19.0                                                                 |
| Heuristic  | Fréchet      | {q,u,n,a} | 2.74              | 0.19                                 | 89.5                                       | 5.8                                                             | 6.1                                                           | 157.7                                      | 20.4                                                            | 18.0                                                          | 95.3                                              | 20.1                                                                 |
| Linear     | Log-normal   | {q,u,n,a} | 1.72              | 0.39                                 | 99.0                                       | 5.2                                                             | 5.3                                                           | 139.3                                      | 11.4                                                            | 11.1                                                          | 68.9                                              | 13.5                                                                 |
| Linear     | Log-logistic | {q,u,n,a} | 1.73              | 0.42                                 | 98.4                                       | 5.1                                                             | 5.5                                                           | 138.3                                      | 12.0                                                            | 11.5                                                          | 68.3                                              | 14.4                                                                 |
| Linear     | Weibull      | {q,u,n,a} | 1.78              | 0.43                                 | 102.8                                      | 5.9                                                             | 6.0                                                           | 140.2                                      | 9.4                                                             | 10.7                                                          | 76.9                                              | 15.4                                                                 |
| Linear     | Fréchet      | {q,u,n,a} | 1.60              | 0.35                                 | 95.0                                       | 4.6                                                             | 4.8                                                           | 140.0                                      | 14.0                                                            | 12.3                                                          | 65.8                                              | 13.6                                                                 |
| Quadrature | Log-normal   | {q,u,n,a} | 2.27              | 0.32                                 | 96.6                                       | 5.2                                                             | 5.4                                                           | 137.6                                      | 11.2                                                            | 11.0                                                          | 69.7                                              | 13.7                                                                 |
| Quadrature | Log-logistic | {q,u,n,a} | 2.28              | 0.35                                 | 96.1                                       | 5.4                                                             | 6.2                                                           | 136.8                                      | 12.6                                                            | 11.3                                                          | 69.2                                              | 14.8                                                                 |
| Quadrature | Weibull      | {q,u,n,a} | 2.33              | 0.36                                 | 100.6                                      | 6.0                                                             | 6.0                                                           | 137.9                                      | 9.3                                                             | 10.7                                                          | 76.6                                              | 15.7                                                                 |
| Quadrature | Fréchet      | {q,u,n,a} | 2.16              | 0.29                                 | 92.6                                       | 4.5                                                             | 4.9                                                           | 138.7                                      | 13.8                                                            | 12.3                                                          | 67.2                                              | 13.9                                                                 |
| Rice       | Log-normal   | {q,u,n,a} | 2.34              | 0.39                                 | 95.9                                       | 5.3                                                             | 5.5                                                           | 137.0                                      | 11.3                                                            | 10.8                                                          | 69.8                                              | 13.6                                                                 |
| Rice       | Log-logistic | {q,u,n,a} | 2.36              | 0.41                                 | 95.5                                       | 5.4                                                             | 6.4                                                           | 136.2                                      | 12.1                                                            | 11.3                                                          | 69.4                                              | 14.8                                                                 |
| Rice       | Weibull      | {q,u,n,a} | 2.40              | 0.43                                 | 99.9                                       | 6.2                                                             | 6.1                                                           | 137.1                                      | 9.1                                                             | 11.6                                                          | 76.4                                              | 16.5                                                                 |
| Rice       | Fréchet      | {q,u,n,a} | 2.24              | 0.59                                 | 91.8                                       | 4.4                                                             | 5.1                                                           | 138.4                                      | 13.8                                                            | 12.2                                                          | 67.7                                              | 13.9                                                                 |

## S5 Background region parameter sensitivity

The local background correction defined in Sect. 4.1 of the main text estimates the column density bias (to be subtracted out) using an annular region around each plume. This region is defined by 2 parameters: a buffer distance ( $b = 3$  pixels in the main analysis) which is neither included in the plume or background mask, and the actual width of the background region ( $w = 10$  pixels in the main analysis). To verify that the reported results do not depend on this particular choice of values, we conducted a sensitivity analysis and tested alternative corrections with combinations of  $w \in \{5, 7, 10, 15, 20, 30\}$  and  $b \in \{1, 2, 3, 5, 10\}$  pixels. For each combination, we plot the corrected source rate as a function of the original uncorrected rate (Figure S2). A linear regression is fitted (in log scale) for each case. We note that the linear regression is highly stable to the change of parameters, with the slope varying between 0.83 and 0.85, and the intercept between 0.08 and 0.11. We also compile the

ensemble of fitted slopes, normalized by the reference configuration ( $w = 10$ ,  $b = 3$  highlighted in red), and represent the distribution as a histogram in Figure S3. The slope varies within around  $\pm 2\%$  of the reference configuration, which is about the amount of variability to be expected on the global source rate calibration if alternative parameterizations were used.

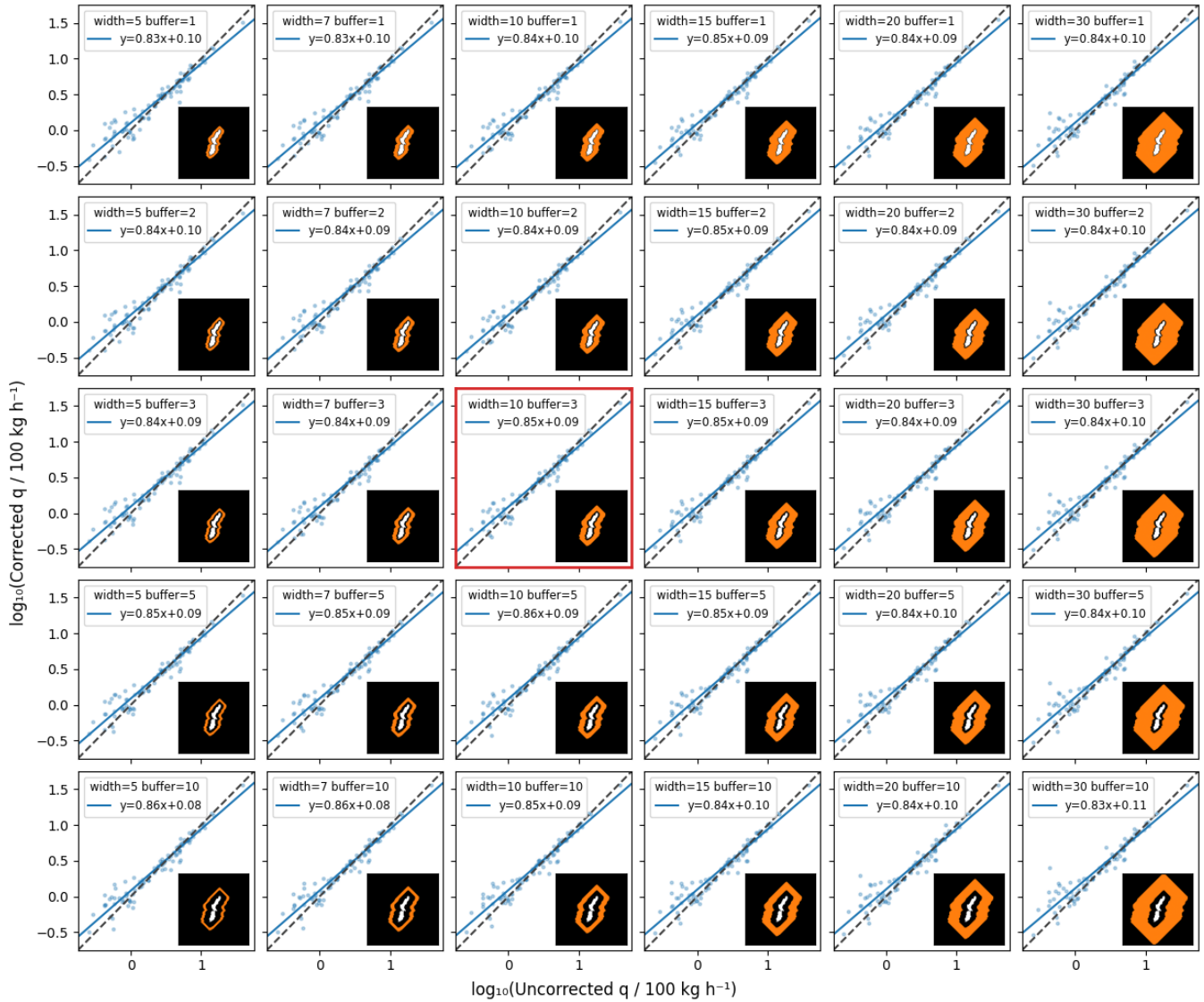


**Figure S2.** Sensitivity of the local background correction to the choice of background region parameters. Each panel shows the corrected source rate against the uncorrected rate ( $\log_{10}$  scale, in units of  $100 \text{ kg h}^{-1}$ ) for one combination of background region width  $w$  and buffer distance  $b$ , with the fitted linear regression (blue line) and the 1:1 line (dashed). The lower right inset in each panel illustrates the plume mask (white) and annular background region (orange) corresponding to that parameter combination. The reference configuration used in the main analysis ( $w = 10$ ,  $b = 3$ ) is outlined in red.

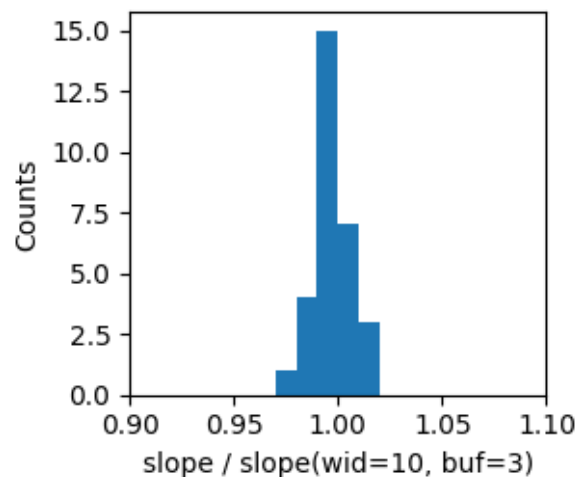


**Figure S3. Distribution of the fitted regression slopes of Figure S2, normalized by the slope of the reference configuration ( $w = 10$ ,  $b = 3$ ). Most parameter combinations fall within approximately  $\pm 2\%$  of the reference slope.**

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