S1 Arc length and distance from the center line

For the mass-balance approach, a two-dimensional curve \( p(r) \) is fitted that consists of two parabolic polynomials:

\[
x(r) = a_0 r^2 + a_1 r + a_2
\]
\[
y(r) = b_0 r^2 + b_1 r + b_2
\]

with coefficients \( a_k \) and \( b_k \) and radial distance \( r \). The parameter is calculated as the distance from the origin

\[
r = \sqrt{(x-x_o)^2 + (y-y_o)^2}
\]

where \( x \) and \( y \) are easting and northing in the "DHDN / Soldner Berlin" spatial reference system (EPSG: 3068).

To convert pixel coordinates in easting and northing to plume coordinates, we need to compute the arc length of the curve from the origin of the source \( x_p \) and the distance of the pixel center from the curve \( y_p \). The minimum distance can be found by minimizing the following equation

\[
\min_r \left[ (x(r) - p_x)^2 + (y(r) - p_y)^2 \right]
\]

which can be done by substituting \( x(r) \) and \( y(r) \), taking the first derivative and finding the values for \( r \) where the first derivative is equal to zero. The steps were executed using the Sympy Library (www.sympy.org), which is a Python library for symbolic computing. The result is a cubic equation for which the roots can be computed using the general cubic formula (e.g. https://en.wikipedia.org/wiki/Cubic_equation#General_cubic_formula). The shortest distance to the curve was used in case of multiple (real) solutions. The sign for the distance was assigned a negative value if the pixel was on the right side of the curve when viewed from the source location and positive otherwise.

The along-plume distance is the arc length \( x_p \) from the source origin to the point where the satellite pixel is perpendicular to the curve. The arc length for a parametric curve (computed from the source origin to the points computed above) is

\[
s = \int_a^b \sqrt{\left( \frac{\partial x(r)}{\partial r} \right)^2 + \left( \frac{\partial y(r)}{\partial r} \right)^2}
\]

which with Eq. (1) results in the integral over the square root of a quadratic polynomial whose solution can be found in most integral tables (e.g. Eq. (37) on http://integral-table.com/):

\[
\int \sqrt{ax^2 + bx + c} \, dx = \frac{b + 2ax}{4a} \sqrt{ax^2 + bx + c} + \frac{4ac - b^2}{8a^{3/2}} \ln \left| 2ax + b + 2\sqrt{a(ax^2 + bx + c)} \right|.
\]

S2 Hermite spline

The seasonal cycle is fitted by a cubic Hermite spline with periodic boundary conditions:

\[
s(\xi) = p_k \cdot (1 + 2\xi)(1 - \xi)^2 + m_k \cdot \xi(1 - \xi)^2 + p_{k+1} \cdot \xi^2(1 - 2\xi) + m_{k+1} \cdot \xi^2(\xi - 1)
\]

with control points \( p_k \) and three-point difference \( m_k \).
Figure S1. Diurnal cycle of Berlin’s CO$_2$ emissions for winter (JFM: January, February and March) and summer (JAS: July, August and September).

Figure S2. Time series of CO$_2$ emissions of Berlin estimated with the analytical inversion using six satellites with $\sigma_{\text{VEG50}}$ of 0.7 ppm for (a) constant and (b) time-varying emissions. Emission estimates with uncertainties larger than 10.0 Mt yr$^{-1}$ (50% of mean emissions at 11:30 local time) are not shown.
**Figure S3.** Time series of estimated CO₂ emissions of Berlin using a constellation of six satellites with medium noise instruments ($\sigma_{\text{VEG50}} = 0.7$ ppm). The plumes were detected from (a) the CO₂ and (b) the NO₂ observations. The error bars show constant errors of 10.0 Mt yr\(^{-1}\) corresponding to the standard deviation of the differences between estimated and real emissions.

**Figure S4.** Histograms of (a) method, (b) retrieval, (c) background, (d) wind and (e) total errors for the emissions estimated by the mass-balance approach. The values are for plumes detected with the medium noise CO₂ observations ($\sigma_{\text{VEG50}} = 0.7$ ppm) and line densities computed from the sub-polygon means.
Figure S5. Same as Fig. S4 but for plumes detected from NO₂ observations.

S4 Additional tables

Table S1. Relative mean bias (MB in %) and standard deviation (SD in %) for method, retrieval, background, wind and total error using the CO₂ observations for detecting the plume.

<table>
<thead>
<tr>
<th>σVEG50 (ppm)</th>
<th>Method error</th>
<th>Retrieval error</th>
<th>Background error</th>
<th>Wind error</th>
<th>Total error</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.5 0.7 1.0</td>
<td>0.5 0.7 1.0</td>
<td>0.5 0.7 1.0</td>
<td>0.5 0.7 1.0</td>
<td>0.5 0.7 1.0</td>
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<tr>
<td>MB (sub-polys)</td>
<td>4 -0 -1</td>
<td>7 10 13</td>
<td>-13 -19 -27</td>
<td>14 19 22</td>
<td>8 5 3</td>
</tr>
<tr>
<td>MB (Gauss)</td>
<td>7 4 3</td>
<td>13 17 14</td>
<td>-13 -12 -23</td>
<td>15 19 21</td>
<td>17 21 12</td>
</tr>
<tr>
<td>SD (sub-polys)</td>
<td>32 30 33</td>
<td>17 18 22</td>
<td>57 53 49</td>
<td>39 35 35</td>
<td>47 40 45</td>
</tr>
<tr>
<td>SD (Gauss)</td>
<td>32 32 33</td>
<td>20 31 29</td>
<td>49 52 47</td>
<td>39 35 35</td>
<td>50 47 47</td>
</tr>
</tbody>
</table>

Table S2. Relative mean bias (MB in %) and standard deviation (SD in %) for method, retrieval, background, wind and total error using the NO₂ observations for detecting the plume.

<table>
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<tr>
<th>σVEG50 (ppm)</th>
<th>Method error</th>
<th>Retrieval error</th>
<th>Background error</th>
<th>Wind error</th>
<th>Total error</th>
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</thead>
<tbody>
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<td>0.5 0.7 1.0</td>
</tr>
<tr>
<td>MB (sub-polys)</td>
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<td>4 5 7</td>
<td>7 7 7</td>
<td>5 5 5</td>
<td>13 15 17</td>
</tr>
<tr>
<td>MB (Gauss CO₂)</td>
<td>-3 -3 -3</td>
<td>5 8 17</td>
<td>13 14 16</td>
<td>6 6 6</td>
<td>22 26 31</td>
</tr>
<tr>
<td>MB (Gauss CO₂+NO₂)</td>
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<td>5 9 14</td>
<td>13 15 14</td>
<td>6 6 3</td>
<td>18 22 25</td>
</tr>
<tr>
<td>SD (sub-polys)</td>
<td>32 32 32</td>
<td>15 21 30</td>
<td>39 40 42</td>
<td>31 31 31</td>
<td>51 51 53</td>
</tr>
<tr>
<td>SD (Gauss CO₂)</td>
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<td>17 21 27</td>
<td>34 35 37</td>
<td>31 31 31</td>
<td>46 48 55</td>
</tr>
<tr>
<td>SD (Gauss CO₂+NO₂)</td>
<td>33 33 32</td>
<td>13 16 25</td>
<td>34 36 38</td>
<td>31 31 28</td>
<td>41 42 45</td>
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