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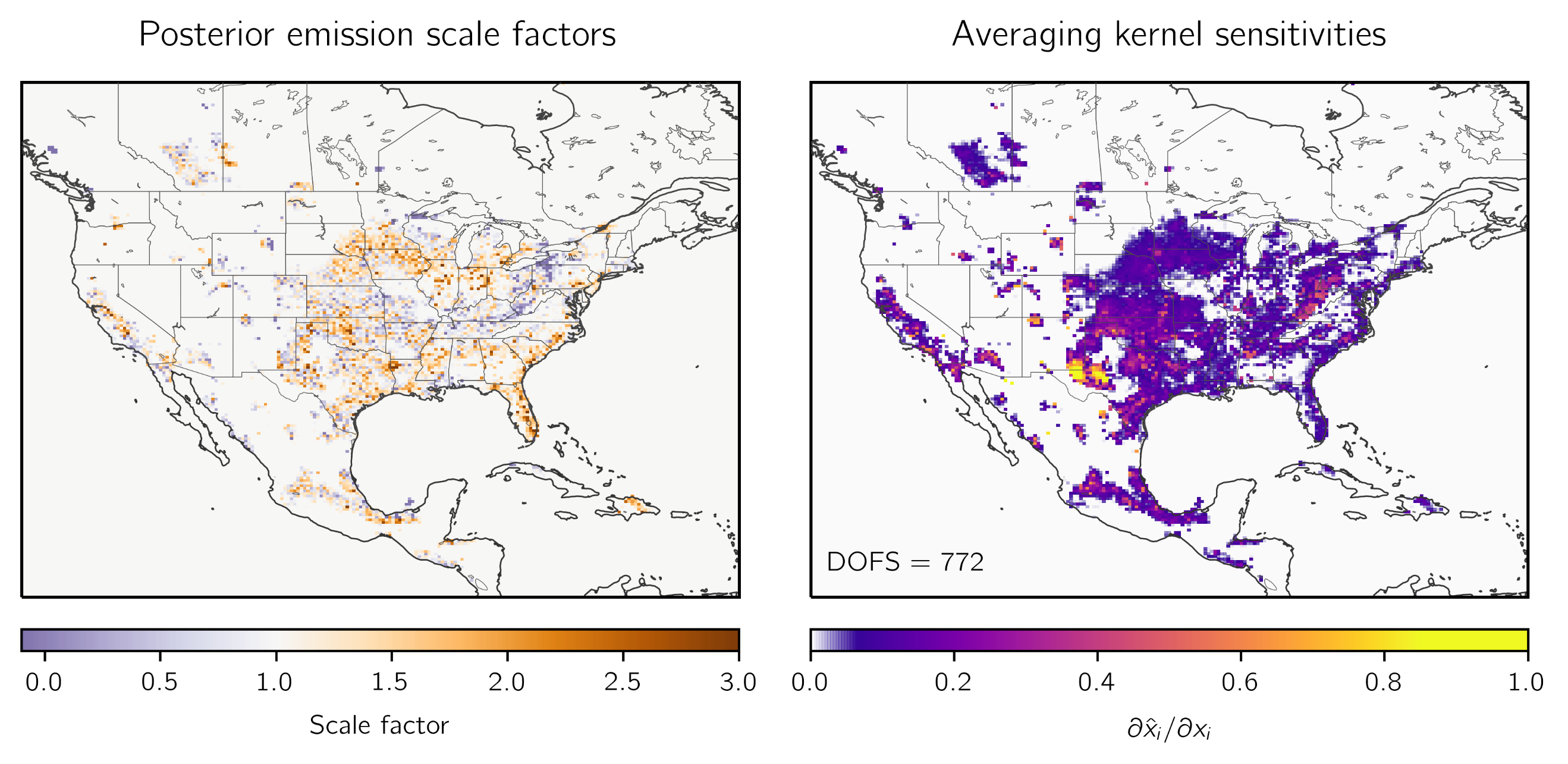
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**Figure 1:** Bottom-up methane emission inventories used as prior estimates for the inversion. Panels show annual mean methane emissions for different anthropogenic sectors from the gridded versions of the inventories of Canada (ECCC), the U.S. (EPA GHGI), and Mexico (INECC). Wetland emissions are given by a high-performance subset of the WetCHARTs version 1.3.1 wetlands inventory ensemble. Emissions are shown on the 0.25° ⨉ 0.3125° GEOS-Chem grid used for the inversion.

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**Figure 2:** TROPOMI methane observations in 2019. The left panel shows the average column dry methane mixing ratios (XCH4) for 2019 averaged on the 0.25° ⨉ 0.3125° GEOS-Chem grid. The right panel shows the observational density on the same grid. The observations have been filtered as described in section 2.4.

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**Figure 3:** Optimization of 2019 methane emissions by inversion of TROPOMI observations. The left panel shows the scale factors relative to the gridded versions of the national anthropogenic emission inventories for the U.S. (EPA GHGI), Canada (ECCC), and Mexico (INECC), and the WetCHARTs wetland emissions, used as prior estimates for the inversion (top left panel of Figure 1). The right panel shows the observing system information content as measured by the averaging kernel sensitivities (the diagonal elements of the averaging kernel matrix). Values of 1 indicate that TROPOMI quantifies emissions independent of the prior estimate, while values of 0 indicate that emissions are not optimized by the inversion. The sum of the averaging kernel sensitivities gives the degrees of freedom for signal (DOFS), shown inset, which defines the number of pieces of information independently quantified by the observing system. Grid cells with averaging kernel sensitivities lower than 0.05 are left blank.

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**Figure 4:** Optimization of 2019 sectoral methane emissions in the contiguous United States (CONUS) by inversion of TROPOMI observations. EPA GHGI (top bars) and posterior (bottom bars) emissions are shown for different sectors. For wetland emissions we show the WetCHARTs estimate (top bar). The dark shading corresponds to emissions actually optimized by the inversion (0.25o×0.3125o grid cells with averaging kernel sensitivities greater than 0.05), while the light shading represents emissions not optimized by the inversion (defaulting to the prior estimate). Error bars on the posterior emissions are given by the spread of the eight-member inversion ensemble. Also shown are previous inversion results from Lu et al. (2022) and Shen et al. (2022) with their reported error bars.

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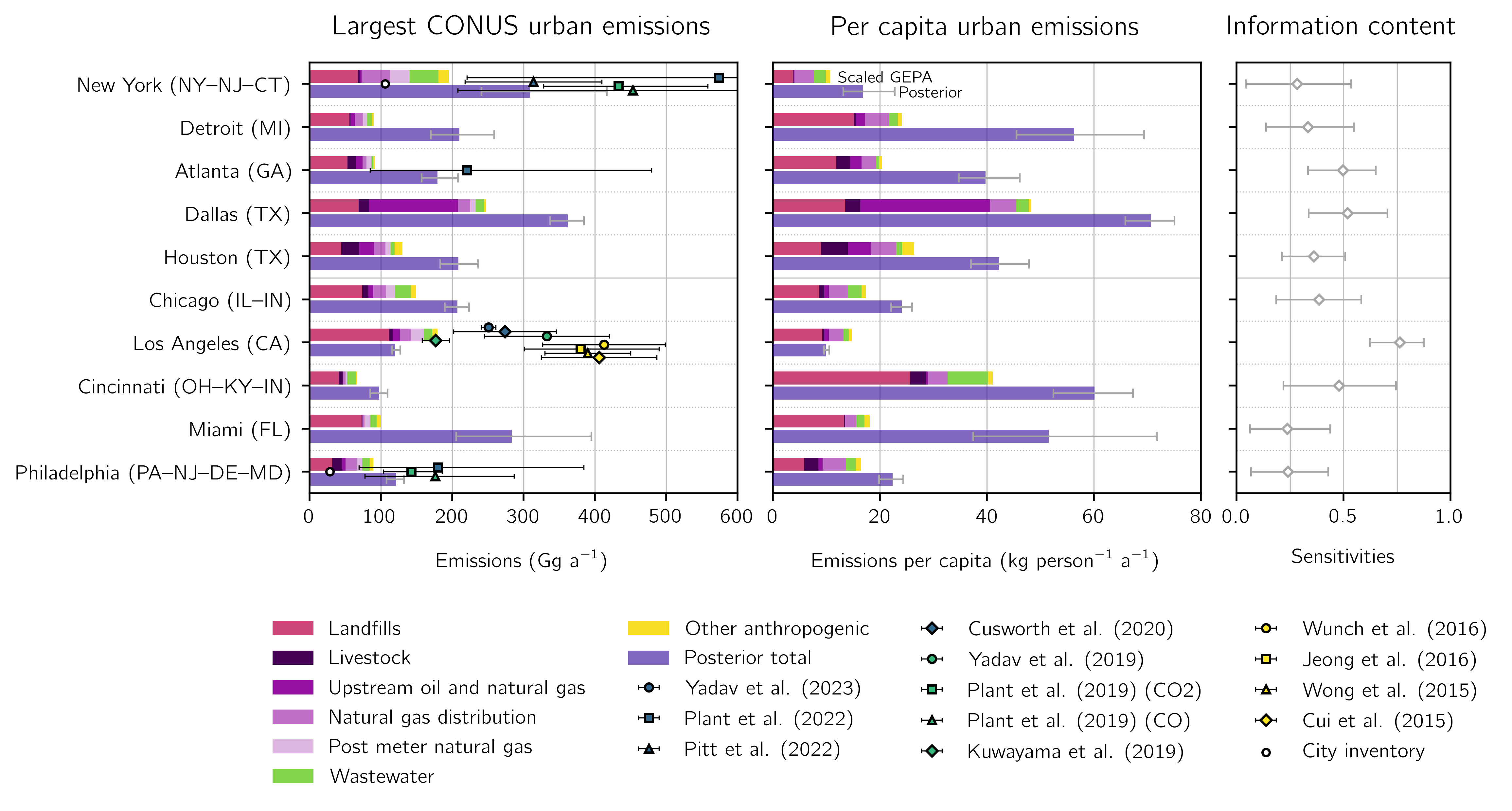
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**Figure 6:** Anthropogenic methane emissions in 2019 for the 29 states in the contiguous United States (CONUS) responsible for 90% of national anthropogenic posterior emissions. The bottom panel shows EPA GHGI state estimates (left bar) and our posterior estimates from inversion of TROPOMI data (right bar) divided by sector. States are listed from largest to smallest posterior emissions. The information content as defined by the reduced-form averaging kernel sensitivities (the diagonal elements of the reduced-form averaging kernel matrix; section 2.8) is shown in the top panel. The error bars give the spread from an eight-member inversion ensemble. Also shown are emissions estimates from independent state inventories cited in EPA (2022).

****

**Figure 7:** Anthropogenic methane emissions for the largest 10 methane-producing urban areas in the contiguous United States (CONUS). Urban area extents are given by the U.S. Census Bureau TIGER/Line files (U.S. Census, 2010). The top bars show prior anthropogenic sectoral emissions from the gridded EPA inventory scaled to match the 2019 EPA GHGI with post-meter emissions allocated by population. The bottom bar shows posterior emissions from the TROPOMI inversion for 2019. We do not resolve posterior sectoral emissions estimates due to source colocation within urban areas. Total emissions (left panel), per capita emissions (center panel), and averaging kernel sensitivities (right panel) are shown for each urban area. Error bars represent the spread of the eight-member inversion ensemble. Also shown are independent urban emissions estimates.

**Table 1:** The 8 members of the inversion ensemble.

|  |  |  |  |
| --- | --- | --- | --- |
| **Optimized boundary conditions**1 | **Latitude correction**2 | **Prior error standard deviation**3 | **Regularization factor**3 |
| Yes | Yes | 50% | 0.2 |
| 75% | 0.45 |
| Yes | No | 50% | 0.175 |
| 75% | 0.3 |
| 100% | 0.5 |
| No | Yes | 50% | 0.175 |
| 75% | 0.35 |
| No | No | 75% | 0.175 |

We conduct inversions that do and do not optimize the boundary conditions. In inversions with optimized boundary conditions, we include in the inversion state vector four boundary condition elements corresponding to the northern, eastern, southern, and western borders of the North American domain.

2 We conduct inversions that do and do not correct the latitudinal bias in the (model – observation) difference with a first order polynomial. In inversions without a latitudinal correction, we still remove the mean (model – observation) difference as might be driven by boundary condition biases.

3 We balance the prior and observing system errors to avoid overfitting the emissions to the observations. The regularization factor is applied to the inverse observational error covariance matrix so that values less than one increase the observing system errors. We choose the value of the regularization factor for a given inversion so that the prior term of the posterior cost function is approximately unity as required by chi-square statistics (section 2.7).

**Table 2:** 2019 methane emissions for the contiguous United States (CONUS).



|  |  |  |  |
| --- | --- | --- | --- |
|  | **US EPA GHGI**1 | **Posterior**2 | **Sensitivity**3 |
| **Total sources [Tg a**-1**]** | 34.6 | 39.3 (38.2 – 40.3) |  |
| **Anthropogenic sources** | 26.8 | 30.9 (30.0 – 31.8) |  |
| Livestock | 9.4 (8.6 – 10.2) | 10.4 (10.0 – 10.7) | 0.66 (0.55 – 0.76) |
| Oil and natural gas | 8.8 (7.4 – 10.2) | 10.4 (10.1 – 10.7) | 0.91 (0.88 – 0.95) |
| Coal | 2.1 (2.0 – 2.5) | 1.5 (1.2 – 1.9) | 0.60 (0.45 – 0.80) |
| Landfills | 4.5 (3.5 – 5.5) | 6.9 (6.4 – 7.5) | 0.47 (0.34 – 0.64) |
| Wastewater | 0.7 (0.5 – 0.9) | 0.6 (0.5 – 0.7) | 0.33 (0.16 – 0.60) |
| Other anthropogenic | 0.6 (0.4 – 1.0) | 1.1 (1.0 – 1.2) | 0.59 (0.44 – 0.76) |
| **Natural sources** | 7.8 | 8.4 (8.1 – 8.6) |  |
| Wetlands | 6.6 | 7.2 (7.0 – 7.4) | 0.35 (0.16 – 0.55) |
| Other biogenic | 1.1 | 1.2 (1.2 - 1.2) | 0.25 (0.19 - 0.32) |

1 as reported in the mos recent version of the U.S. Environmental Protection Agency (EPA) 2022 Inventory of Greenhouse Gas Emissions and Sinks (GHGI) for 2019. Error ranges are given by the sum in quadrature of bottom-up subsector errors. Wetland emissions are from a subset of the high performance WetCHARTs ensemble version 1.3.1. See section 2.2 for details.

2Optimized emissions from the inversion of TROPOMI data, with the range from the eight members of the inversion ensemble shown in parentheses.

3The sensitivity of the posterior emissions to the observing system as given by the diagonal elements of the sectoral averaging kernel matrix calculated as described in section 2.8. The parenthetical values give the range of the inversion ensemble. Values range from 0 (no sensitivity) to 1 (full sensitivity)

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Description automatically generated**Figure S1:** I will write this caption once we decide whether to use this figure.

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**Figure S2:** I will write this caption once we decide whether to use this figure.

**A picture containing night, light, dark

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**Figure S3:** I will write this caption once we decide whether to use this figure.

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**Figure S4:** I will write this caption once we decide whether to use this figure.

**Table S1:** 2019 methane emissions from the 48 states in the contiguous United States.

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Emissions**  **(Gg a-1)**1 | **Livestock** | | **Oil & natural gas** | | **Coal** | | **Landfills** | | **Wastewater** | | **Other anthropogenic** | | **Total** | | |
| **State** | **GHGI**2 | **x̂**3 | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂**4 | **DOFS**5 |
| 1. Texas | 1023 | 1165 | 2096 | 4299 | 9 | 24 | 509 | 627 | 60 | 48 | 42 | 110 | 3739 | 6274 (6101, 6454) | 0.94 (0.89, 0.97) |
| 2. California | 760 | 1104 | 309 | 231 | 0 | 0 | 348 | 514 | 65 | 58 | 63 | 148 | 1544 | 2055 (1970, 2122) | 0.86 (0.75, 0.93) |
| 3. Oklahoma | 380 | 399 | 643 | 894 | 3 | 20 | 86 | 121 | 10 | 3 | 7 | 6 | 1128 | 1444 (1384, 1511) | 0.86 (0.75, 0.92) |
| 4. Pennsylvania | 199 | 196 | 703 | 238 | 498 | 524 | 109 | 196 | 25 | 20 | 22 | 20 | 1555 | 1194 (1061, 1384) | 0.57 (0.35, 0.77) |
| 5. New Mexico | 170 | 211 | 406 | 925 | 28 | 32 | 34 | -34 | 3 | 2 | 6 | 3 | 647 | 1139 (1100, 1180) | 0.96 (0.93, 0.98) |
| 6. Louisiana | 62 | 79 | 443 | 731 | 1 | 2 | 131 | 126 | 10 | 9 | 13 | 174 | 660 | 1121 (1010, 1258) | 0.55 (0.28, 0.76) |
| 7. Iowa | 555 | 793 | 57 | 59 | 0 | 0 | 63 | 116 | 23 | 13 | 6 | 7 | 705 | 989 (952, 1010) | 0.75 (0.54, 0.88) |
| 8. Illinois | 160 | 191 | 143 | 121 | 126 | 170 | 157 | 368 | 25 | 37 | 17 | 21 | 627 | 907 (862, 944) | 0.55 (0.29, 0.79) |
| 9. Florida | 155 | 250 | 56 | 26 | 0 | 0 | 311 | 540 | 38 | 15 | 22 | 47 | 582 | 878 (699, 1106) | 0.32 (0.04, 0.58) |
| 10. Kansas | 490 | 448 | 373 | 358 | 0 | 0 | 54 | 41 | 18 | 9 | 5 | 3 | 940 | 860 (839, 888) | 0.80 (0.66, 0.89) |
| 11. Colorado | 263 | 232 | 392 | 351 | 65 | 102 | 72 | 110 | 14 | 4 | 10 | 5 | 816 | 804 (740, 861) | 0.59 (0.44, 0.72) |
| 12. Michigan | 182 | 187 | 160 | 121 | 0 | 0 | 196 | 392 | 18 | 19 | 27 | 22 | 582 | 742 (674, 813) | 0.49 (0.16, 0.74) |
| 13. Alabama | 102 | 109 | 122 | 120 | 183 | 154 | 168 | 259 | 21 | 25 | 10 | 11 | 605 | 677 (629, 717) | 0.75 (0.55, 0.89) |
| 14. North Carolina | 266 | 375 | 41 | 23 | 0 | 0 | 185 | 225 | 28 | 17 | 12 | 13 | 531 | 654 (547, 744) | 0.48 (0.24, 0.71) |
| 15. Ohio | 165 | 146 | 348 | 160 | 30 | 32 | 214 | 244 | 19 | 24 | 18 | 16 | 793 | 622 (578, 673) | 0.63 (0.38, 0.82) |
| 16. Indiana | 140 | 170 | 74 | 60 | 132 | 79 | 115 | 274 | 15 | 17 | 13 | 16 | 489 | 616 (561, 676) | 0.54 (0.28, 0.74) |
| 17. Nebraska | 531 | 533 | 45 | 24 | 0 | 0 | 47 | 46 | 22 | 5 | 4 | 3 | 649 | 611 (604, 619) | 0.64 (0.48, 0.73) |
| 18. West Virginia | 28 | 26 | 386 | 182 | 582 | 360 | 30 | 32 | 3 | 2 | 5 | 4 | 1033 | 607 (485, 730) | 0.66 (0.46, 0.83) |
| 19. Arkansas | 124 | 122 | 136 | 134 | 0 | 13 | 61 | 106 | 15 | 10 | 8 | 218 | 343 | 605 (569, 636) | 0.74 (0.48, 0.86) |
| 20. Georgia | 114 | 127 | 51 | 47 | 0 | 0 | 256 | 374 | 28 | 9 | 14 | 18 | 462 | 575 (509, 655) | 0.58 (0.35, 0.73) |
| 21. Wisconsin | 424 | 407 | 46 | 16 | 0 | 0 | 83 | 114 | 15 | 8 | 17 | 14 | 584 | 559 (518, 595) | 0.47 (0.07, 0.70) |
| 22. Idaho | 316 | 317 | 13 | 11 | 0 | 0 | 20 | 219 | 5 | 2 | 8 | 3 | 362 | 551 (498, 596) | 0.63 (0.49, 0.76) |
| 23. Minnesota | 295 | 381 | 48 | 26 | 0 | 0 | 52 | 83 | 16 | 4 | 15 | 10 | 425 | 504 (475, 534) | 0.53 (0.13, 0.69) |
| 24. Mississippi | 77 | 104 | 87 | 132 | 3 | 6 | 73 | 134 | 11 | 24 | 6 | 23 | 256 | 423 (380, 478) | 0.53 (0.22, 0.75) |
| 25. New York | 230 | 139 | 131 | 47 | 0 | 0 | 107 | 154 | 29 | 43 | 27 | 23 | 524 | 405 (352, 445) | 0.30 (0.06, 0.50) |

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Emissions**  **(Gg a-1)**1 | **Livestock** | | **Oil & natural gas** | | **Coal** | | **Landfills** | | **Wastewater** | | **Other anthropogenic** | | **Total** | | |
| **State** | **GHGI**2 | **x̂**3 | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂** | **GHGI** | **x̂**4 | **DOFS**5 |
| 26. Kentucky | 154 | 143 | 148 | 68 | 61 | 69 | 152 | 105 | 11 | 4 | 9 | 7 | 536 | 395 (347, 449) | 0.64 (0.40, 0.82) |
| 27. South Dakota | 332 | 347 | 13 | 12 | 0 | 0 | 11 | 18 | 5 | 12 | 2 | 2 | 362 | 392 (376, 401) | 0.38 (0.11, 0.53) |
| 28. Missouri | 331 | 266 | 42 | 14 | 0 | 0 | 64 | 54 | 16 | 9 | 13 | 24 | 467 | 367 (339, 394) | 0.55 (0.29, 0.69) |
| 29. Virginia | 112 | 109 | 88 | 31 | 153 | 20 | 119 | 169 | 20 | 22 | 14 | 11 | 507 | 362 (299, 428) | 0.56 (0.35, 0.75) |
| 30. Tennessee | 132 | 122 | 54 | 40 | 2 | 2 | 114 | 132 | 13 | 20 | 9 | 7 | 324 | 322 (301, 349) | 0.60 (0.33, 0.77) |
| 31. Montana | 215 | 211 | 87 | 63 | 20 | 10 | 13 | 19 | 2 | 1 | 8 | 3 | 344 | 306 (292, 322) | 0.31 (0.22, 0.40) |
| 32. North Dakota | 136 | 124 | 139 | 141 | 5 | 6 | 18 | 26 | 2 | 2 | 3 | 2 | 302 | 300 (286, 317) | 0.59 (0.41, 0.70) |
| 33. Washington | 147 | 149 | 25 | 20 | 0 | 0 | 70 | 98 | 16 | 14 | 21 | 13 | 280 | 293 (269, 337) | 0.10 (0.04, 0.14) |
| 34. Utah | 92 | 105 | 103 | 49 | 28 | 79 | 30 | 49 | 6 | 0 | 5 | 3 | 265 | 285 (248, 336) | 0.74 (0.57, 0.87) |
| 35. Oregon | 115 | 132 | 24 | 23 | 0 | 0 | 55 | 111 | 7 | 3 | 14 | 8 | 215 | 276 (256, 304) | 0.08 (0.05, 0.11) |
| 36. Arizona | 121 | 141 | 50 | 41 | 1 | 2 | 70 | 72 | 11 | 4 | 6 | 3 | 259 | 263 (261, 266) | 0.80 (0.74, 0.84) |
| 37. South Carolina | 37 | 53 | 26 | 11 | 0 | 0 | 68 | 145 | 12 | 21 | 8 | 8 | 151 | 237 (220, 249) | 0.51 (0.20, 0.70) |
| 38. New Jersey | 4 | 4 | 44 | 51 | 0 | 0 | 56 | 116 | 13 | 35 | 11 | 27 | 128 | 233 (186, 294) | 0.28 (0.06, 0.52) |
| 39. Maryland | 23 | 28 | 19 | 20 | 2 | 4 | 44 | 57 | 12 | 4 | 8 | 7 | 109 | 120 (112, 126) | 0.26 (0.04, 0.45) |
| 40. Nevada | 45 | 49 | 20 | 9 | 0 | 0 | 17 | 30 | 4 | 2 | 3 | 2 | 90 | 93 (93, 93) | 0.00 (0.00, 0.00) |
| 41. Massachusetts | 4 | 4 | 29 | 17 | 0 | 0 | 24 | 48 | 10 | 4 | 9 | 7 | 76 | 80 (66, 93) | 0.15 (0.00, 0.35) |
| 42. Wyoming | 109 | 113 | 281 | 142 | 200 | -186 | 6 | 10 | 1 | 0 | 3 | 1 | 601 | 80 (-194, 279) | 0.68 (0.48, 0.86) |
| 43. Vermont | 38 | 29 | 1 | 0 | 0 | 0 | 6 | 13 | 1 | 1 | 6 | 3 | 52 | 46 (45, 49) | 0.02 (0.00, 0.07) |
| 44. Connecticut | 8 | 5 | 12 | 8 | 0 | 0 | 8 | 15 | 5 | 12 | 5 | 4 | 38 | 45 (35, 51) | 0.26 (0.01, 0.50) |
| 45. Maine | 11 | 10 | 4 | 2 | 0 | 0 | 13 | 20 | 3 | 1 | 10 | 6 | 40 | 38 (37, 39) | 0.00 (0.00, 0.00) |
| 46. New Hampshire | 4 | 5 | 3 | 1 | 0 | 0 | 21 | 16 | 3 | 1 | 6 | 3 | 36 | 25 (23, 27) | 0.03 (0.00, 0.08) |
| 47. Delaware | 3 | 4 | 5 | 2 | 0 | 0 | 17 | 8 | 5 | 5 | 1 | 2 | 31 | 20 (19, 22) | 0.12 (0.04, 0.23) |
| 48. Rhode Island | 0 | 1 | 5 | 4 | 0 | 0 | 6 | 11 | 2 | 2 | 2 | 1 | 15 | 18 (14, 21) | 0.19 (0.07, 0.34) |

Sectoral emissions in gigagrams per year (Gg a-1) for anthropogenic sources, including livestock, oil and natural gas, landfills, wastewater, and other categories, including rice.

2Bottom-up emissions for each state from the Environmental Protection Agency (EPA) Inventory of U.S. Greenhouse Gas Emissions and Sinks (GHGI) state estimates for 2019.

3Optimized sectoral anthropogenic emissions from an inversion of TROPOMI data.

4The total anthropogenic optimized emissions. Values in parentheses give the minimum and maximum of the ensemble of 8 inversions.

5The sensitivity of the total state posterior emissions to the observing system, given by the diagonal elements of the state averaging kernel matrix calculated. Values in parentheses give the ensemble range. Sensitivities range from 0 (unresponsive to the observing system) to 1 (fully responsive).

**Table S2:** Top 10 methane-producing urban areas in the contiguous U.S. (CONUS) for 2019.

I will update this table when we decide what to do with urban areas.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Urban area**1 | **Prior emissions (Gg a-1)**2 | | | | | **Posterior emissions** | | |
| **Landfills** | **Wastewater** | **Oil and natural gas** | **Livestock** | **Other anthropogenic** | **Total (Gg a-1)** | **Per capita**  **(kg person-1 a-1)**4 | **Sensitivity**5 |
| New York-Newark, NY-NJ-CT | 57.24 | 24.33 | 35.90 | 0.68 | 15.72 | 222 (169, 302) | 12.10 | 0.28 (0.04, 0.54) |
| Dallas-Fort Worth-Arlington, TX | 53.78 | 8.99 | 59.82 | 3.37 | 2.78 | 157 (138, 172) | 30.80 | 0.52 (0.34, 0.70) |
| Detroit, MI | 36.26 | 4.04 | 8.93 | 0.53 | 3.63 | 119 (95, 150) | 31.80 | 0.33 (0.14, 0.55) |
| Chicago, IL--IN | 47.31 | 11.97 | 15.25 | 2.12 | 6.54 | 110 (101, 118) | 12.80 | 0.38 (0.18, 0.58) |
| Houston, TX | 36.86 | 3.51 | 20.72 | 3.55 | 5.37 | 106 (86, 124) | 21.30 | 0.36 (0.21, 0.51) |
| Atlanta, GA | 33.69 | 2.22 | 7.36 | 3.06 | 2.68 | 100 (86, 117) | 22.20 | 0.50 (0.33, 0.65) |
| Miami, FL | 29.75 | 2.04 | 1.55 | 0.41 | 3.54 | 93 (68, 128) | 16.90 | 0.24 (0.06, 0.44) |
| Los Angeles-Long Beach-Anaheim, CA | 97.40 | 5.68 | 19.18 | 3.24 | 7.57 | 89 (83, 103) | 7.30 | 0.76 (0.62, 0.88) |
| Philadelphia, PA-NJ-DE-MD | 17.54 | 6.39 | 16.25 | 4.01 | 5.73 | 69 (58, 78) | 12.70 | 0.24 (0.07, 0.43) |
| Indianapolis, IN | 15.54 | 0.86 | 7.09 | 0.91 | 1.49 | 60 (49, 78) | 40.60 | 0.34 (0.13, 0.60) |

The 10 urban areas with the largest optimized methane emissions. Urban area extents are given by the U.S. Census Bureau TIGER/Line files.

2The prior anthropogenic emissions for landfills, wastewater, oil and natural gas, livestock, and other sources for each urban area in gigagrams per year (Gg a-1) from the gridded version of the Environmental Protection Agency (EPA) Inventory of U.S. Greenhouse Gas Emissions and Sinks (GHGI) for 2012. Oil and natural gas emissions are updated to match 2018 infrastructure and 2018 emissions as reported by the 2020 GHGI for 2018. There are no coal emissions in these urban areas.

3Optimized emissions from inversion of TROPOMI observations in gigagrams per year. Values in parentheses represent the range from an eight-member inversion ensemble.

4Mean per capita methane emissions assuming 2010 Census populations in kilograms per person per year (kg-1 person-1 a-1).

5The sensitivity of an urban area to the satellite-model observing system as given by the diagonal elements of the urban averaging kernel matrix calculated as described in section 2.8. Values close to 1 indicate that the posterior emissions are fully sensitive to the observing system, while values close to 0 rely almost entirely on the prior estimate. Values in parentheses give the ensemble range.