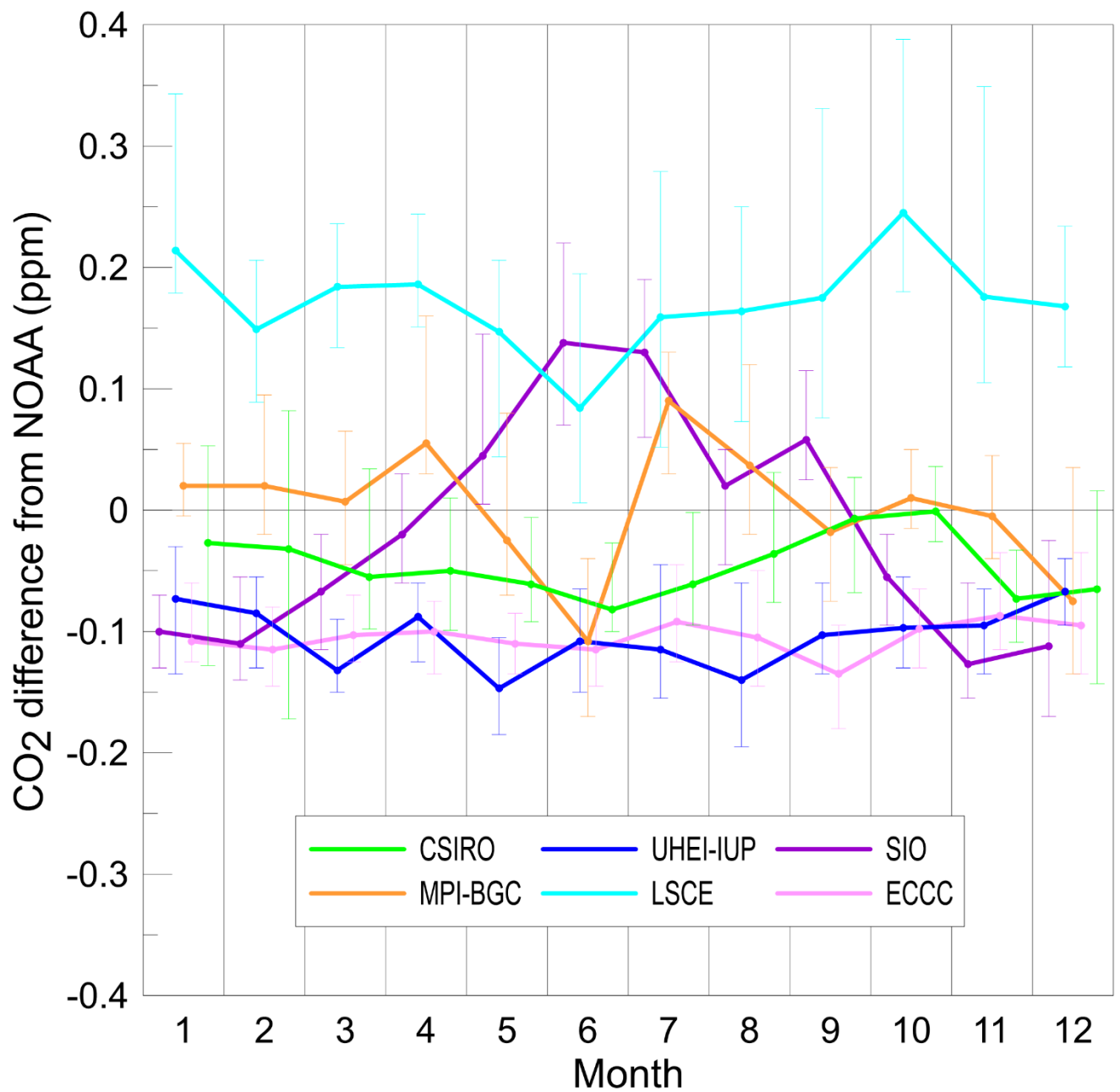
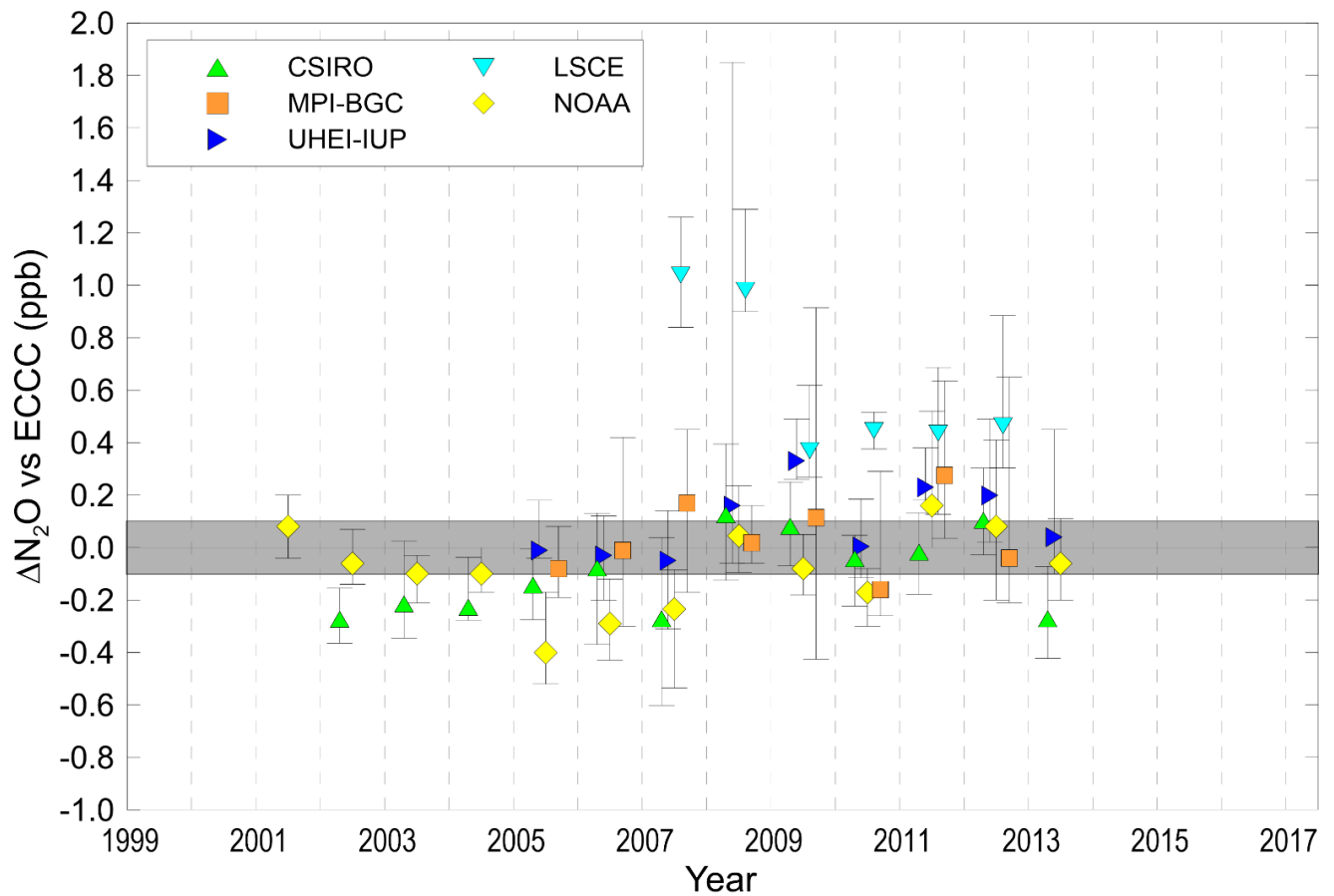


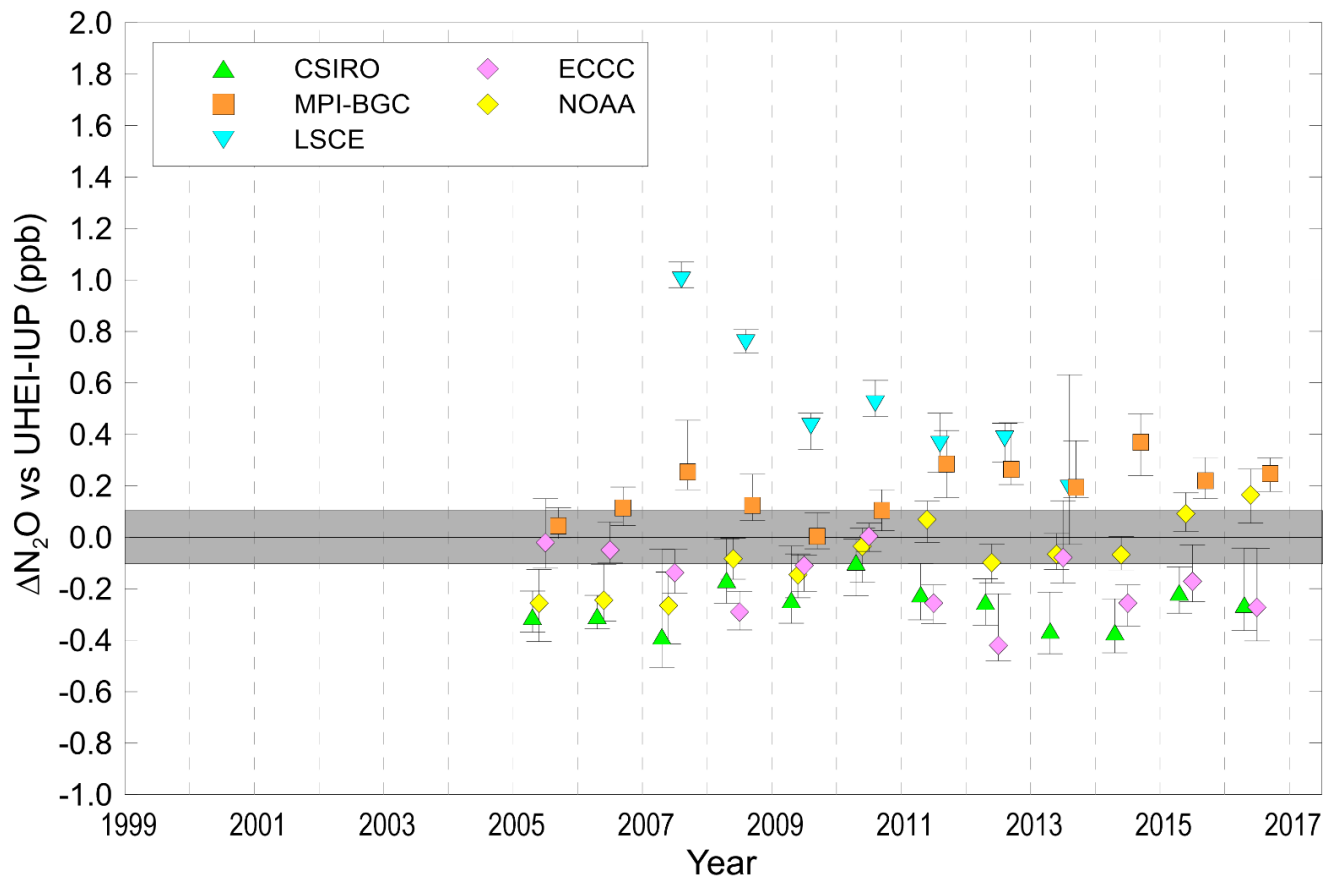
3  
4 **Figure S1.** Annual median CO<sub>2</sub> values and 95% confidence limits, in ppm, for each  
5 laboratory's same-flask difference distribution relative to ECCC as a function of time. The  
6 same-flask comparison program terminated in 2013. The shaded grey band around the zero  
7 line indicates the WMO/GAW recommended measurement agreement goal of ±0.1 ppm for  
8 CO<sub>2</sub>.



**Figure S2.** CO<sub>2</sub> median differences and 95% confidence limits (laboratory minus NOAA) by month, for the entire 17-year period. The SIO measurements relative to NOAA during the May-September period relative to the October-March period possibly show a seasonal bias on the order of 0.25 ppm.



**Figure S3.** Annual median  $N_2O$  values and 95% confidence limits, in ppb, for each laboratory's same-flask difference distribution relative to ECCC as a function of time. The same-flask comparison program terminated in 2013. The shaded grey band around the zero line indicates the WMO/GAW recommended measurement agreement goal of  $\pm 0.1$  ppb for  $N_2O$ .



**Figure S4.** Annual median N<sub>2</sub>O values and 95% confidence limits, in ppb, for each laboratory's co-located difference distribution relative to UHEI-IUP as a function of time. The shaded grey band around the zero line indicates the WMO/GAW recommended measurement agreement goal of  $\pm 0.1$  ppb for N<sub>2</sub>O.

**Table S1.** WMO/ IAEA Round Robin experiments results (laboratory minus NOAA or INSTAAR) conducted over the time period, that are on the same scale as the data used in the analysis. All RR data is from the WMORR website ([Global Monitoring Laboratory - Carbon Cycle Greenhouse Gases \(noaa.gov\)](http://Global Monitoring Laboratory - Carbon Cycle Greenhouse Gases (noaa.gov))) except for ECCC's SF<sub>6</sub> data, which was updated to the SF<sub>6</sub> X2014 scale, and CSIRO's  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  RR#5 data, which was provided to us by CSIRO. INSTARR's  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  data was provided to us by INSTAAR, with updates for scale via JRAS-06 realization, to be consistent with flask data scale realization. All other laboratory isotope differences were updated according to the new INSTAAR data.

| Species                   | RR # | Date      | CSIRO                  | MPI-BGC                | UHEI-IUP               | LSCE             | SIO                 | ECCC                 |
|---------------------------|------|-----------|------------------------|------------------------|------------------------|------------------|---------------------|----------------------|
| CO <sub>2</sub> (ppm)     | 5    | 2009-2012 | 0.00, 0.00, -0.10      | -0.02, 0.00, 0.00      | -0.04, -0.04, -0.08    | 0.06, 0.01, 0.12 | -0.20, -0.14, -0.11 | -0.02, -0.02, -0.03  |
|                           | 6    | 2014-2015 | 0.04, 0.00             | -0.01, -0.02           | -0.03, -0.06           | -0.05, 0.00      |                     | 0.09, 0.06           |
| $\delta^{13}\text{C}$ (‰) | 5    | 2009-2012 | -0.055, -0.061, -0.082 | -0.056, -0.044, -0.048 | -0.053, -0.050, -0.056 |                  |                     | 0.006, 0.001, -0.006 |
|                           | 6    | 2014-2015 | -0.042, -0.029         | -0.033, -0.039         | -0.082, -0.086         |                  |                     | 0.013, 0.003         |
| $\delta^{18}\text{O}$ (‰) | 5    | 2009-2012 | 0.111, 0.022, -0.024   | -0.452, -0.066, -0.656 | 0.081, 0.112, 0.035    |                  |                     | 0.187, 0.170, 0.107  |
|                           | 6    | 2014-2015 | 0.177, 0.210           | 0.066, 0.066           | 0.035, -0.016          |                  |                     | 0.200, 0.165         |
| CH <sub>4</sub> (ppb)     | 6    | 2014-2015 | 0.50, 0.60             | 0.30, -0.40            | 0.10, -0.70            |                  |                     | 0.30, 0.40           |
| N <sub>2</sub> O (ppb)    | 5    | 2009-2012 | -0.11, -0.05, -0.15    |                        |                        |                  |                     |                      |
|                           | 6    | 2014-2015 | -0.10, -0.14           | 0.16, -0.07            | 0.14, 0.01             | 0.24, 0.37       |                     | -0.45, -0.43         |
| SF <sub>6</sub> (ppt)     | 6    | 2014-2015 |                        | 0.00, 0.02             |                        |                  |                     | 0.05, 0.05           |

**Table S2.** Time stamps of data sets (mm/dd/yyyy)

**ALERT**

| Laboratory | CO <sub>2</sub> | CH <sub>4</sub> | N <sub>2</sub> O | SF <sub>6</sub> | $\delta^{13}\text{C-CO}_2$ | $\delta^{18}\text{O-CO}_2$ |
|------------|-----------------|-----------------|------------------|-----------------|----------------------------|----------------------------|
| CSIRO      | 10/09/2018      | 10/09/2018      | 10/09/2018       |                 | 10/09/2018                 | 10/09/2018                 |
| MPI-BGC    | 02/02/2018      | 02/02/2018      | 02/02/2018       | 02/02/2018      | 02/02/2018                 | 02/02/2018                 |
| UHEI-IUP   | 03/06/2018      | 03/06/2018      | 03/06/2018       |                 | 03/06/2018                 | 03/06/2018                 |
| LSCE       | 03/23/2017      | 03/20/2017      | 03/23/2017       | 03/23/2017      | 09/25/2017                 | 10/03/2017                 |
| SIO        | 01/17/2019      |                 |                  |                 | 01/17/2019                 | 01/17/2019                 |
| ECCC       | 11/06/2018      | 01/24/2018      | 03/02/2018       | 01/24/2018      | 09/04/2018                 | 08/20/2018                 |
| NOAA       | 10/10/2018      | 11/14/2018      | 11/14/2018       | 02/02/2018      | 01/23/2020                 | 01/23/2020                 |

**CGO**

| Laboratory | CO <sub>2</sub> | CH <sub>4</sub> | N <sub>2</sub> O | $\delta^{13}\text{C-CO}_2$ | $\delta^{18}\text{O-CO}_2$ |
|------------|-----------------|-----------------|------------------|----------------------------|----------------------------|
| CSIRO      | 10/18/2018      | 07/05/2018      | 07/05/2018       | 10/18/2018                 | 10/09/2018                 |
| NOAA       | 10/18/2018      | 07/03/2018      | 06/28/2018       | 01/23/2020                 | 01/23/2020                 |

**MLO**

| Laboratory | CO <sub>2</sub> | CH <sub>4</sub> | N <sub>2</sub> O | $\delta^{13}\text{C-CO}_2$ | $\delta^{18}\text{O-CO}_2$ |
|------------|-----------------|-----------------|------------------|----------------------------|----------------------------|
| CSIRO      | 10/18/2018      | 07/05/2018      | 07/05/2018       | 10/18/2018                 | 10/10/2018                 |
| SIO        | 01/17/2019      |                 |                  | 01/17/2019                 | 01/17/2019                 |
| NOAA       | 10/18/2018      | 06/28/2018      | 06/28/2018       | 01/23/2020                 | 01/23/2020                 |

**Table S3.** Compilation of average flask pair differences for CO<sub>2</sub>, in ppm, for each laboratory

|          | AVERAGE FLASK<br>PAIR DIFFERENCE | STD DEV. OF PAIR<br>DIFF | NUMBER OF<br>DATA PTS |
|----------|----------------------------------|--------------------------|-----------------------|
| CSIRO    | 0.15                             | 0.21                     | 346                   |
| MPI-BGC  | 0.12                             | 0.25                     | 302                   |
| UHEI-IUP | 0.12                             | 0.07                     | 546                   |
| LSCE     | 0.11                             | 0.11                     | 239                   |
| SIO      | 0.10                             | 0.08                     | 640                   |
| ECCC     | 0.10                             | 0.12                     | 825                   |
| NOAA     | 0.10                             | 0.17                     | 879                   |

**Table S4.** Compilation of average flask pair differences for  $\delta^{13}\text{C}\text{-CO}_2$  (‰) for each laboratory

|                 | AVERAGE FLASK<br>PAIR DIFFERENCE | STD DEV. OF PAIR<br>DIFF | NUMBER OF<br>DATA PTS |
|-----------------|----------------------------------|--------------------------|-----------------------|
| <b>CSIRO</b>    | 0.015                            | 0.015                    | 309                   |
| <b>MPI-BGC</b>  | 0.039                            | 0.093                    | 286                   |
| <b>UHEI-IUP</b> | 0.013                            | 0.009                    | 409                   |
| <b>LSCE</b>     | 0.057                            | 0.102                    | 189                   |
| <b>SIO</b>      | 0.028                            | 0.018                    | 11                    |
| <b>ECCC</b>     | 0.013                            | 0.018                    | 439                   |
| <b>NOAA</b>     | 0.018                            | 0.021                    | 795                   |



**Table S5.** Compilation of average flask pair differences for  $\delta^{18}\text{O}\text{-CO}_2$  (‰) for each laboratory

|                 | AVERAGE FLASK<br>PAIR DIFFERENCE | STD DEV. OF PAIR<br>DIFF | NUMBER OF<br>DATA PTS |
|-----------------|----------------------------------|--------------------------|-----------------------|
| <b>CSIRO</b>    | 0.119                            | 0.132                    | 307                   |
| <b>MPI-BGC</b>  | 0.104                            | 0.179                    | 286                   |
| <b>UHEI-IUP</b> | 0.029                            | 0.018                    | 280                   |
| <b>LSCE</b>     | 0.215                            | 0.396                    | 189                   |
| <b>SIO</b>      | 0.080                            | 0.089                    | 11                    |
| <b>ECCC</b>     | 0.060                            | 0.050                    | 391                   |
| <b>NOAA</b>     | 0.064                            | 0.103                    | 635                   |

**Table S6.** Compilation of average flask pair differences for CH<sub>4</sub>, in ppb, for each laboratory

|                 | AVERAGE FLASK<br>PAIR DIFFERENCE | STD DEV. OF PAIR<br>DIFF | NUMBER OF<br>DATA PTS |
|-----------------|----------------------------------|--------------------------|-----------------------|
| <b>CSIRO</b>    | 1.86                             | 1.74                     | 346                   |
| <b>MPI-BGC</b>  | 1.42                             | 1.44                     | 303                   |
| <b>UHEI-IUP</b> | 1.61                             | 2.34                     | 504                   |
| <b>LSCE</b>     | 1.23                             | 1.22                     | 247                   |
| <b>ECCC</b>     | 0.74                             | 0.76                     | 841                   |
| <b>NOAA</b>     | 1.24                             | 1.23                     | 886                   |

**Table S7.** Compilation of average flask pair differences for N<sub>2</sub>O, in ppb, for each laboratory

|                 | <b>AVERAGE FLASK<br/>PAIR DIFFERENCE</b> | <b>STD DEV. OF PAIR<br/>DIFF</b> | <b>NUMBER OF<br/>DATA PTS</b> |
|-----------------|--|----------------------------------|-------------------------------|
| <b>CSIRO</b>    | 0.29                                     | 0.22                             | 338                           |
| <b>MPI-BCG</b>  | 0.16                                     | 0.13                             | 302                           |
| <b>UHEI-IUP</b> | 0.13                                     | 0.08                             | 532                           |
| <b>LSCE</b>     | 0.16                                     | 0.13                             | 245                           |
| <b>ECCC</b>     | 0.19                                     | 0.14                             | 690                           |
| <b>NOAA</b>     | 0.30                                     | 0.26                             | 861                           |

**Table S8.** Compilation of average flask pair differences for SF<sub>6</sub>, in ppt, for each laboratory

|                | AVERAGE FLASK<br>PAIR DIFFERENCE | STD DEV. OF PAIR<br>DIFF | NUMBER OF<br>DATA PTS |
|----------------|----------------------------------|--------------------------|-----------------------|
| <b>MPI-BGC</b> | 0.03                             | 0.04                     | 299                   |
| <b>LSCE</b>    | 0.04                             | 0.04                     | 257                   |
| <b>ECCC</b>    | 0.03                             | 0.03                     | 691                   |
| <b>NOAA</b>    | 0.04                             | 0.04                     | 864                   |

**Table S9.** Summary of same-flask annual median CO<sub>2</sub> values, in ppm, for each of the five laboratory difference distributions (laboratory minus ECCC). The 95 % confidence limits of the computed annual median value are shown in parentheses followed by the number of individual measurement differences included in the computation.

| Year      | CSIRO                 | MPI-BGC                | UHEI-IUP               | LSCE                 | NOAA                     |
|-----------|-----------------------|------------------------|------------------------|----------------------|--------------------------|
| 1999      |                       |                        |                        |                      | -0.17 (-0.20,-0.12) 13   |
| 2000      |                       |                        |                        |                      | -0.02 (-0.03,0.01) 60    |
| 2001      |                       |                        |                        |                      | 0.02 (-0.01,0.04) 46     |
| 2002      | 0.25 (0.18,0.33) 32   |                        |                        |                      | 0.02 (0.00,0.03) 89      |
| 2003      | 0.21 (0.15,0.26) 40   |                        |                        |                      | 0.00 (-0.01,0.03) 88     |
| 2004      | 0.20 (0.16,0.27) 34   |                        |                        |                      | -0.02 (-0.03,0.00) 73    |
| 2005      | 0.10 (0.05,0.15) 37   | 0.00 (-0.05,0.05) 34   | 0.13 (0.10,0.15) 46    |                      | -0.03 (-0.05,0.00) 76    |
| 2006      | 0.10 (0.06,0.14) 32   | 0.01 (-0.05,0.14) 18   | 0.16 (0.10,0.22) 33    |                      | 0.03 (0.01,0.05) 95      |
| 2007      | 0.10 (0.04,0.20) 23   | 0.06 (0.01,0.10) 20    | 0.10 (0.05,0.17) 31    | 0.49 (0.44,0.56) 23  | 0.04 (0.02,0.05) 92      |
| 2008      | 0.05 (0.02,0.09) 35   | -0.10 (-0.21,0.04) 18  | -0.13 (-0.15,0.00) 14  | 0.16 (0.12,0.24) 32  | -0.16 (-0.20,-0.09) 69   |
| 2009      | 0.10 (0.08,0.14) 57   | -0.08 (-0.16,0.16) 17  | 0.00 (-0.04,0.08) 30   | 0.17 (0.15,0.23) 37  | -0.04 (-0.07,-0.01) 88   |
| 2010      | 0.02 (0.00,0.06) 74   | -0.03 (-0.10,0.03) 21  | -0.05 (-0.05,-0.01) 40 | 0.07 (0.04,0.10) 37  | -0.08 (-0.09,-0.06) 92   |
| 2011      | -0.01 (-0.06,0.03) 62 | 0.00 (-0.04,0.05) 19   | -0.04 (-0.06,0.00) 33  | 0.06 (-0.02,0.14) 37 | -0.04 (-0.06,0.00) 46    |
| 2012      | -0.01 (-0.03,0.03) 67 | -0.04 (-0.10,0.00) 22  | -0.03 (-0.06,0.01) 41  | 0.11 (0.08,0.13) 43  | -0.06 (-0.08,-0.04) 49   |
| 2013      | 0.01 (-0.02,0.04) 66  | -0.09 (-0.12,0.29) 17  | -0.07 (-0.09,-0.04) 32 | 0.07 (0.02,0.20) 13  | -0.09 (-0.10,-0.06) 48   |
| 1999-2013 | 0.06 (0.05,0.07) 559  | -0.02 (-0.04,0.00) 186 | 0.00 (-0.01,0.02) 300  | 0.12 (0.11,0.15) 222 | -0.02 (-0.03,-0.01) 1024 |

**Table S10.** 11-12 year increases of CO<sub>2</sub> (ppm) calculated from individual datasets from six laboratories (2005-2016), using the de-seasoned data from Nakazawa's curve-fitting routine. (Nakazawa et al., 1997)

| 12 yr trend | 2016   | 2005   | 2005-2016 | 11 yr trend | 2015   | 2005   | 2005-2015 | 11 yr trend | 2016   | 2006   | 2006-2016 |
|-------------|--------|--------|-----------|-------------|--------|--------|-----------|-------------|--------|--------|-----------|
| CSIRO       | 404.22 | 380.64 | 23.59     |             | 401.63 | 380.64 | 20.99     |             | 404.22 | 383.52 | 20.70     |
| MPI-BGC     | 404.25 | 380.82 | 23.43     |             | 401.88 | 380.82 | 21.06     |             | 404.25 | 383.45 | 20.81     |
| UHEI-IUP    | 404.15 | 380.68 | 23.47     |             | 401.69 | 380.68 | 21.01     |             | 404.15 | 383.32 | 20.83     |
| SIO         | 404.44 | 380.46 | 23.98     |             | 401.95 | 380.46 | 21.50     |             | 404.44 | 383.52 | 20.92     |
| ECCC        | 404.17 | 380.57 | 23.60     |             | 401.61 | 380.57 | 21.04     |             | 404.17 | 383.17 | 21.01     |
| NOAA        | 404.32 | 380.68 | 23.64     |             | 401.72 | 380.68 | 21.04     |             | 404.32 | 383.38 | 20.94     |
|             |        |        |           |             |        |        |           |             |        |        |           |
| Mean        | 404.26 | 380.64 | 23.62     | Mean        | 401.75 | 380.64 | 21.11     | Mean        | 404.26 | 383.39 | 20.87     |
| s.d.        | 0.11   | 0.12   | 0.20      | s.d.        | 0.14   | 0.12   | 0.19      | s.d.        | 0.11   | 0.13   | 0.11      |
| (Max - min) | 0.29   | 0.36   | 0.55      | (Max - min) | 0.35   | 0.36   | 0.50      | (Max - min) | 0.29   | 0.35   | 0.30      |
| Rel. Diff % |        |        | 2.33      | Rel. Diff % |        |        | 2.39      | Rel. Diff % |        |        | 1.45      |

Ref. Nakazawa, T., Ishizawa, M., Higuchi, K. and Trivett, N.B.: Two curve fitting methods applied to CO<sub>2</sub> flask data, Environmetrics, Vol 8, 197-218, 1997.

**Table S11.** Summary of same-flask annual median N<sub>2</sub>O values, in ppb, for each of the five laboratory difference distributions (laboratory minus ECCC). The 95 % confidence limits of the computed annual median value are shown in parentheses followed by the number of individual measurement differences included in the computation.

| Year      | CSIRO                   | MPI-BGC               | UHEI-IUP              | LSCE                 | NOAA                    |
|-----------|-------------------------|-----------------------|-----------------------|----------------------|-------------------------|
| 2001      |                         |                       |                       |                      | 0.08 (-0.04,0.20) 73    |
| 2002      | -0.27 (-0.36,-0.15) 30  |                       |                       |                      | -0.06 (-0.14,0.07) 97   |
| 2003      | -0.22 (-0.34,0.02) 39   |                       |                       |                      | -0.10 (-0.21,-0.03) 89  |
| 2004      | -0.23 (-0.28,-0.04) 31  |                       |                       |                      | -0.10 (-0.17,0.00) 77   |
| 2005      | -0.14 (-0.28,-0.01) 36  | -0.08 (-0.19,0.08) 32 | -0.01 (-0.04,0.18) 46 |                      | -0.40 (-0.52,-0.17) 73  |
| 2006      | -0.08 (-0.37,0.13) 33   | -0.01 (-0.30,0.42) 17 | -0.03 (-0.20,0.12) 34 |                      | -0.29 (-0.43,-0.12) 99  |
| 2007      | -0.27 (-0.60,0.04) 23   | 0.17 (-0.17,0.45) 21  | -0.05 (-0.31,0.14) 32 | 1.04 (0.84,1.26) 26  | -0.24 (-0.53,-0.09) 94  |
| 2008      | 0.13 (-0.13,0.40) 36    | 0.02 (-0.06,0.16) 19  | 0.16 (-0.06,1.85) 14  | 0.98 (0.90,1.29) 30  | 0.05 (-0.09,0.23) 68    |
| 2009      | 0.08 (-0.07,0.25) 55    | 0.12 (-0.42,0.91) 16  | 0.33 (0.26,0.49) 29   | 0.37 (0.27,0.62) 38  | -0.08 (-0.18,0.05) 85   |
| 2010      | -0.04 (-0.22,0.04) 73   | -0.16 (-0.26,0.29) 21 | 0.01 (-0.11,0.18) 38  | 0.45 (0.37,0.51) 37  | -0.17 (-0.30,-0.08) 89  |
| 2011      | -0.02 (-0.18,0.14) 62   | 0.28 (0.04,0.63) 18   | 0.23 (0.18,0.38) 32   | 0.44 (0.13,0.69) 35  | 0.16 (0.00,0.52) 47     |
| 2012      | 0.10 (-0.02,0.30) 37    | -0.04 (-0.21,0.65) 15 | 0.20 (0.02,0.49) 21   | 0.46 (0.30,0.89) 27  | 0.08 (-0.20,0.41) 31    |
| 2013      | -0.27 (-0.42,-0.08) 33  |                       | 0.04 (-0.10,0.45) 14  |                      | -0.06 (-0.20,0.11) 21   |
| 2001-2013 | -0.11 (-0.17,-0.05) 488 | 0.03 (-0.06,0.09) 163 | 0.09 (0.03,0.14) 260  | 0.55 (0.48,0.66) 201 | -0.10 (-0.14,-0.08) 943 |