



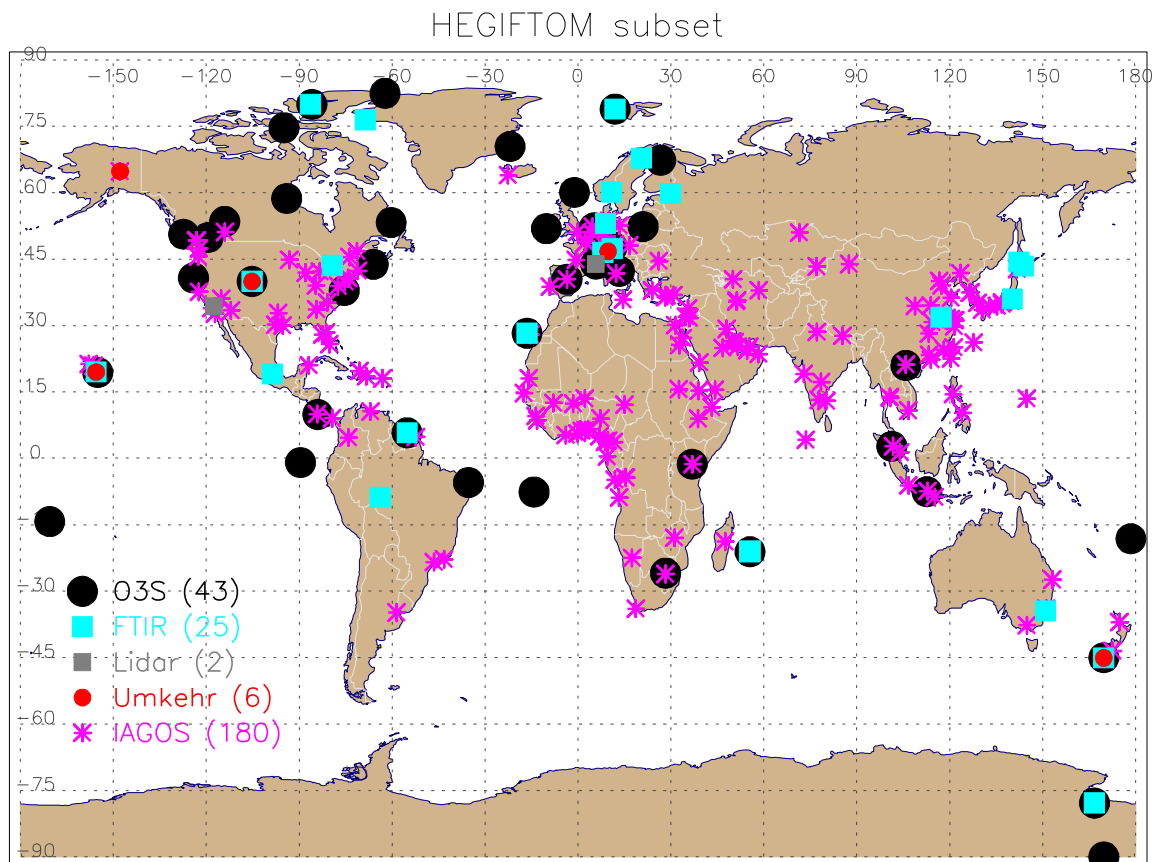
*Supplement of*

**Global ground-based tropospheric ozone measurements:  
reference data and individual site trends (2000–2022)  
from the TOAR-II/HEGIFTOM project**

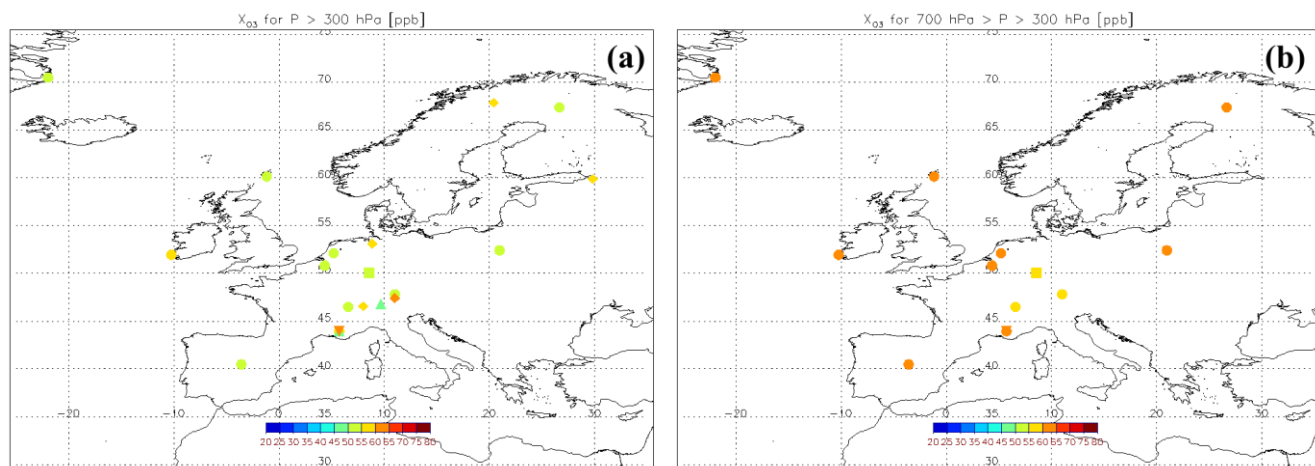
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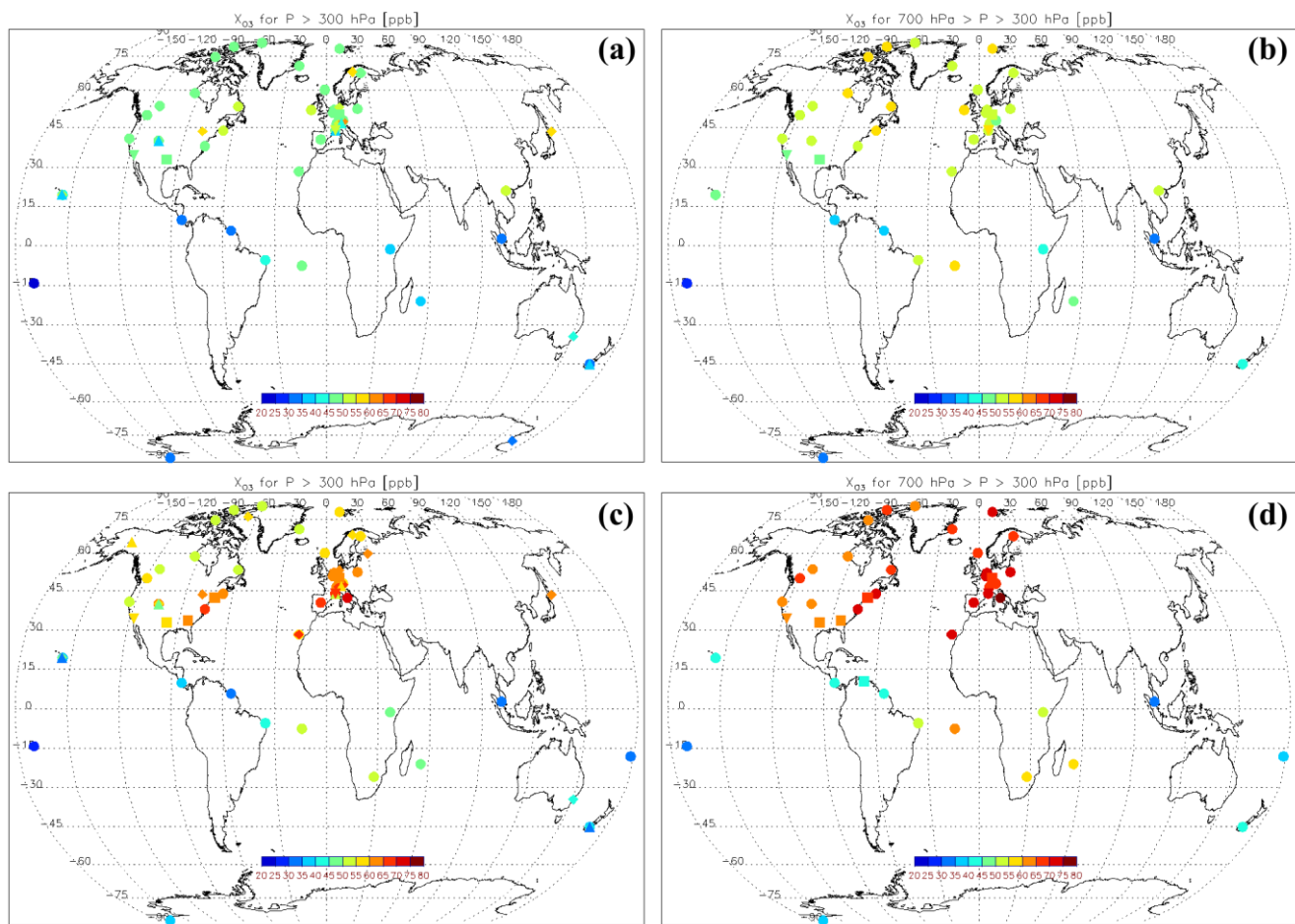
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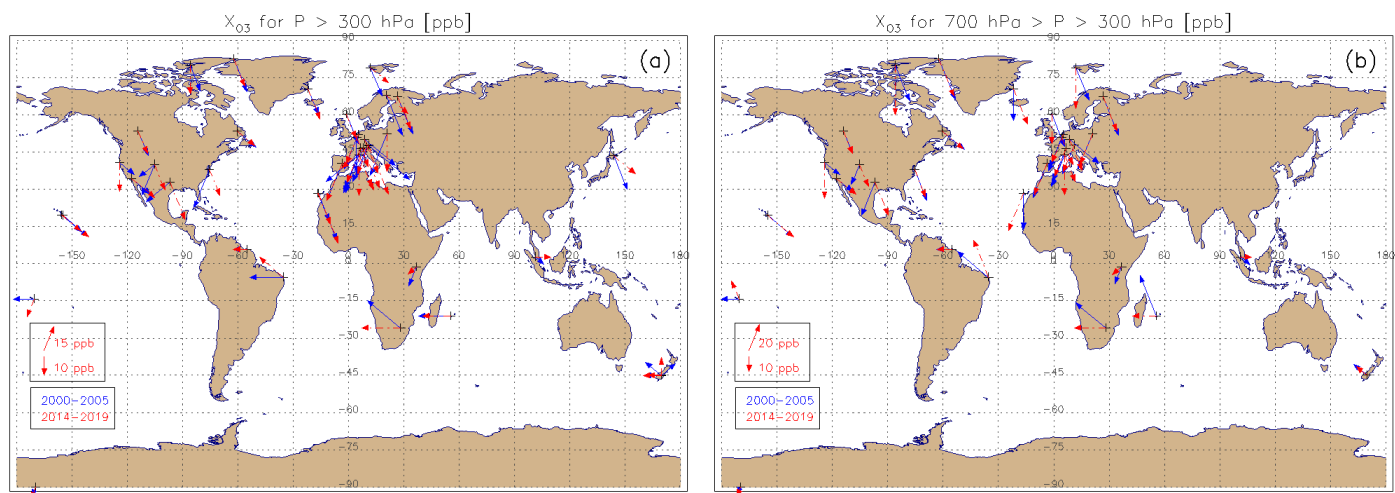
**Figure S1.** Map showing all HEGIFTOM sites with TrOC data since 2000. A list of those stations, together with latitude, longitude, beginning and end year, and amount of observations in the 2000-2022 period is provided in Table S1.



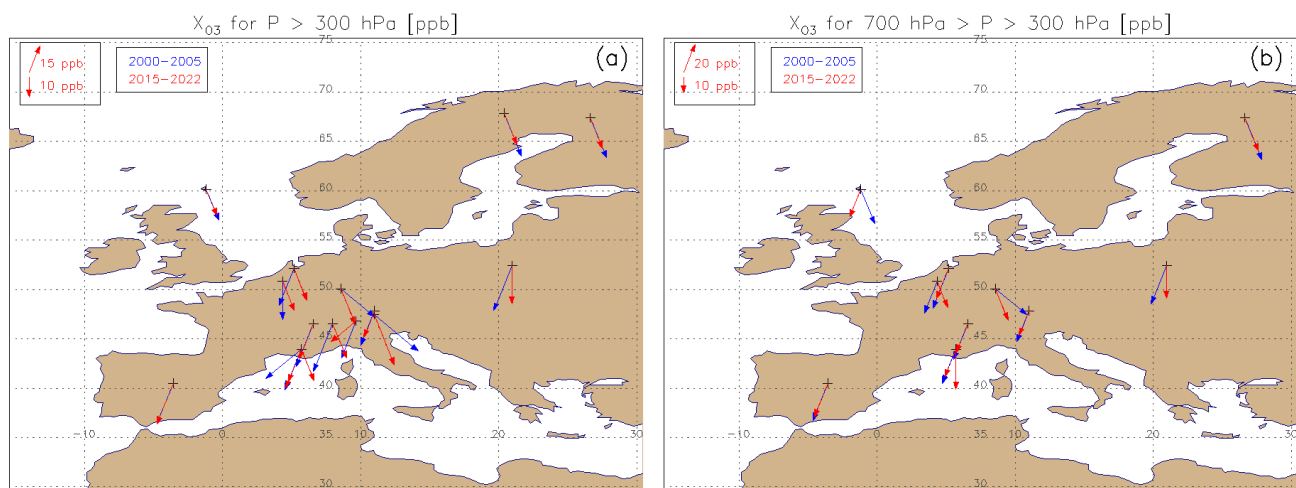
**Figure S2.** Close-up of Fig. 4: mean (a) TrOC (ppb, surface to 300 hPa), (b) FTOC (ppb, 700 > p > 300 hPa) at European HEGIFTOM sites with at least 120 monthly values in the 2000-2022 period. Circles denote ozonesondes, squares denote IAGOS airports, diamonds denote FTIR, upward triangles denote Umkehr, and downward triangles denote lidar.



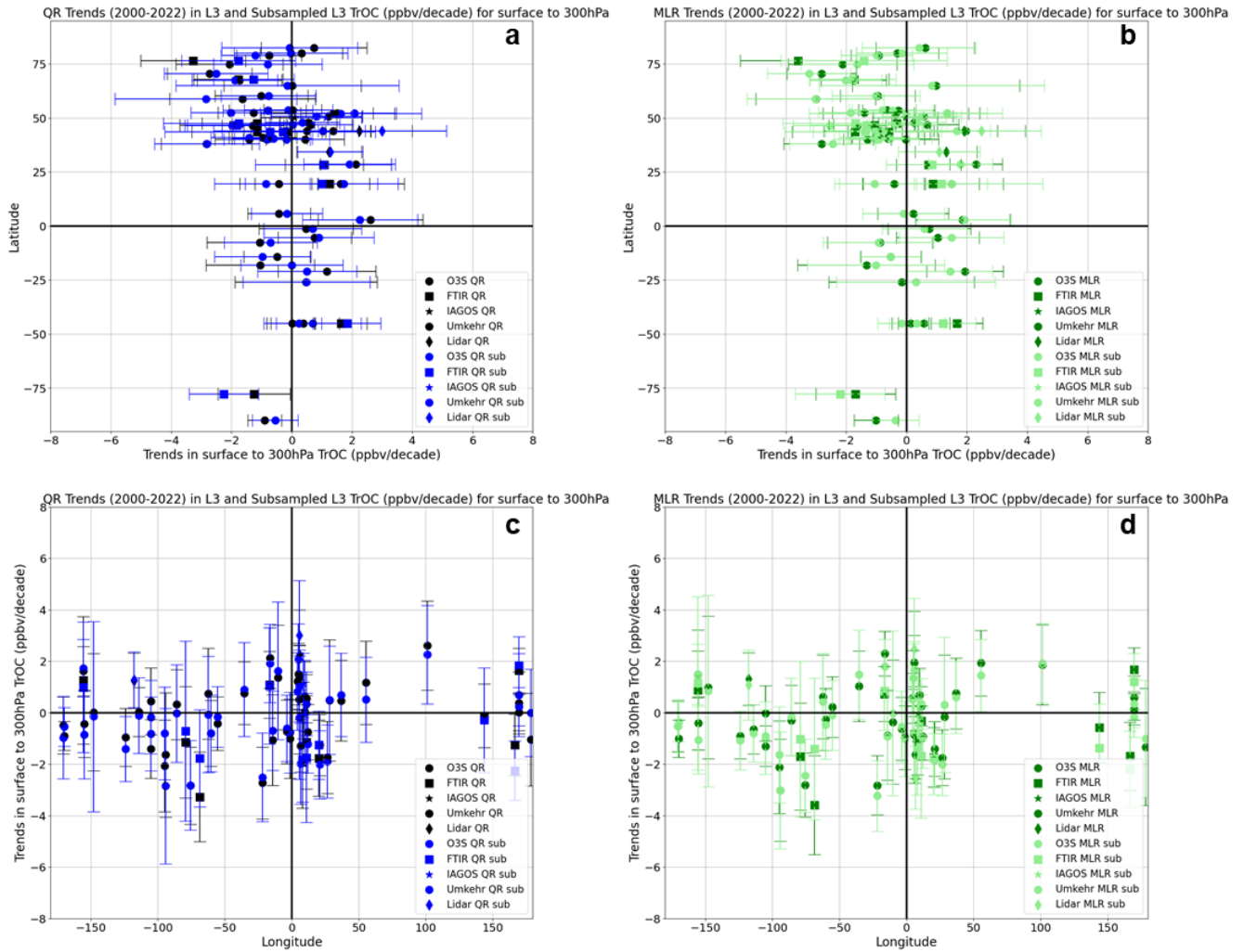
**Figure S3.** Same as Fig. 4, but now for TrOC DJF (a) and JJA (c), and for FTOC DJF (b) and JJA (d).



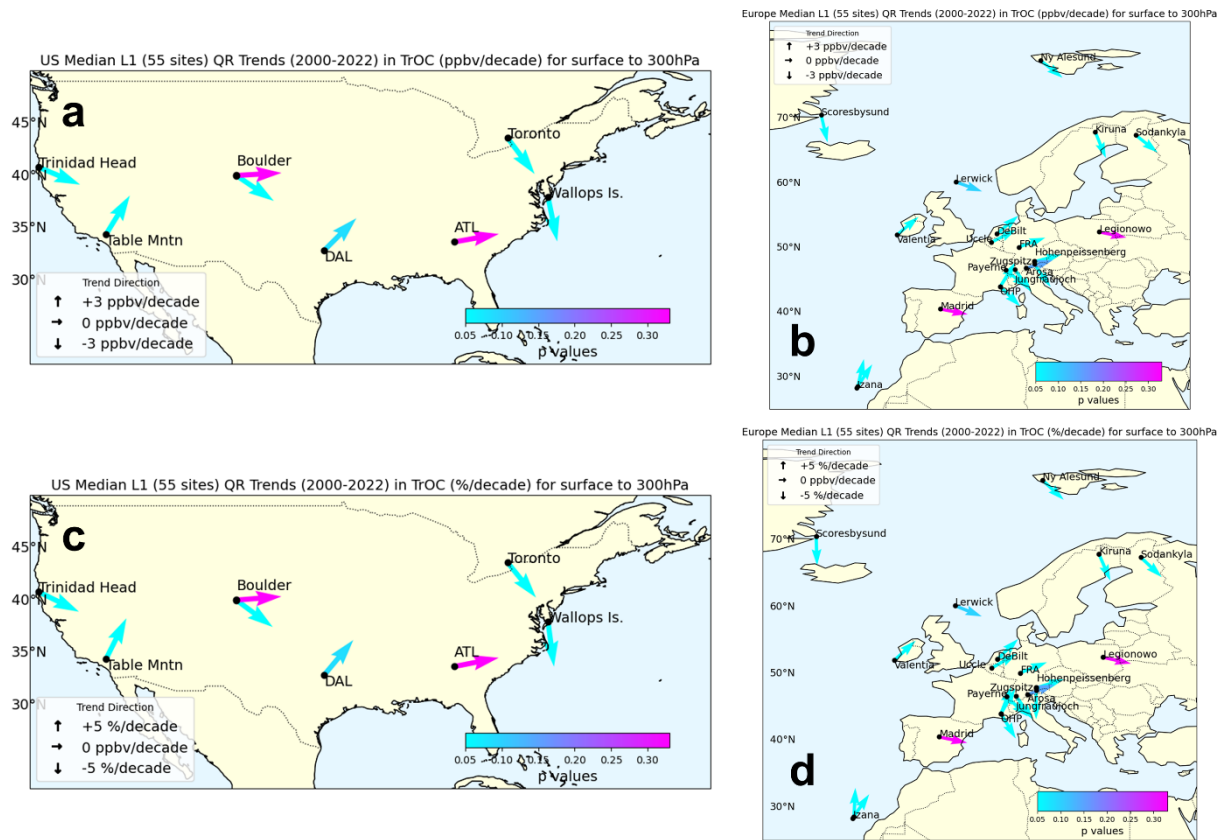
**Figure S4.** Same as Fig. 6, but now for the 2014-2019 period (red) instead of 2015-2022.



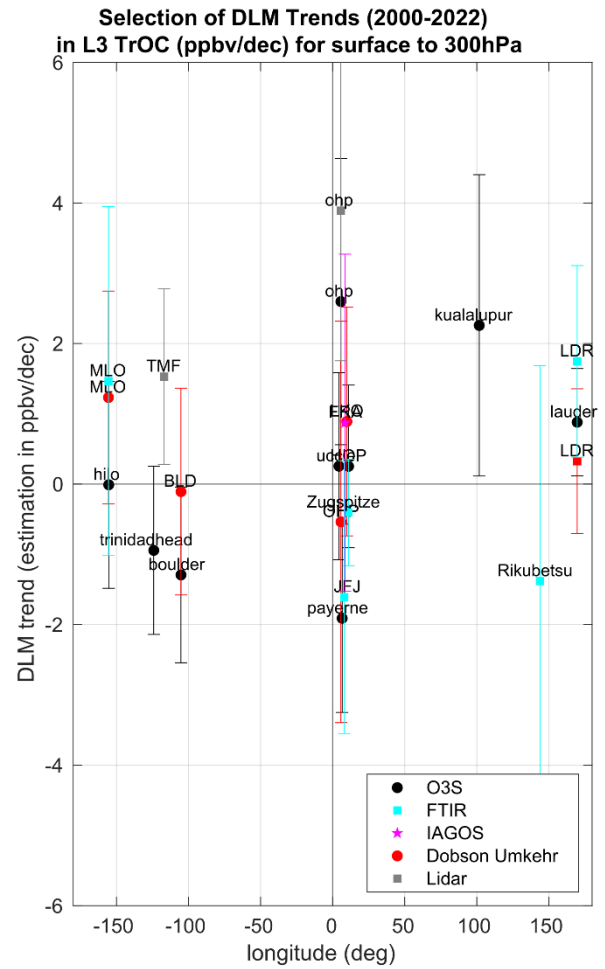
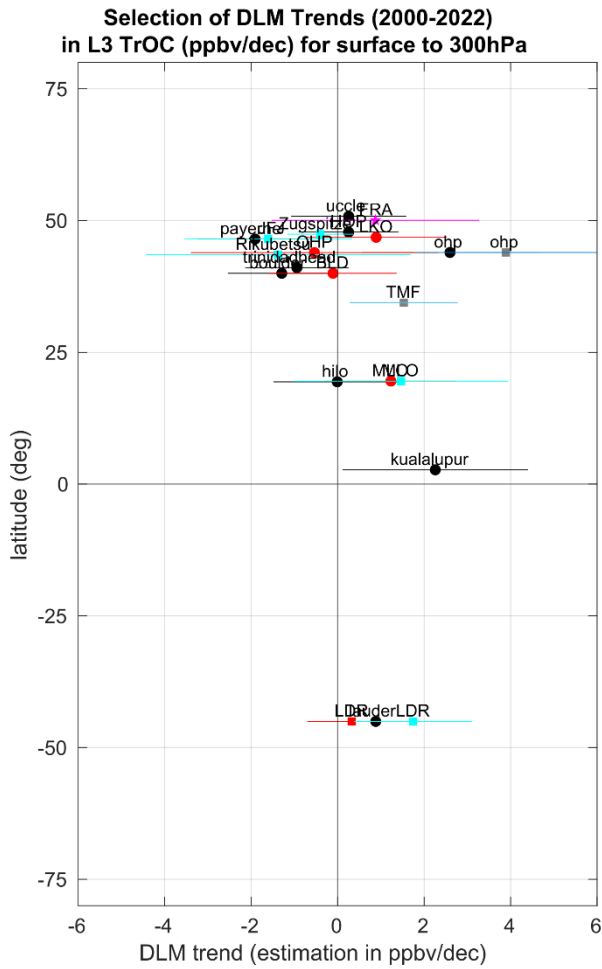
**Figure S5.** Close-up of Fig. 6 for Europe.



**Figure S6.** QR (left) and MLR (right) TrOC trend (2000-2022) estimates as a function of latitude (upper panels) and longitude (lower panels) for the original L3 datasets and for the datasets subsampled to exactly two random daily values for each month (“sub” in legend).



**Figure S7.** Close-up of Fig. 10 trends at stations within highly-sampled regions: (a) US; (b) Europe. Multiple arrows correspond to multi-instrument sites, Boulder in (a), OHP and Izaña in (b). (c) Trend same as (a) except in % per decade with  $p$  values; (d) same as (b) except in % per decade with  $p$  values.



**Figure S8.** DLM trend estimates (ppbv per decade), based on the monthly mean time series for a subset of sites from our sample. DLM trend values are output in ppbv per year. In order to compare DLM trend values with QR and MLR decadal trend values, we need to give an estimate of the trend in ppbv per decade. DLM outputs a distribution of ozone change between the beginning and the end of the considered time period and the trend values in ppbv per decade are determined in ratio. The uncertainty on these values is determined by Monte-Carlo simulations. Assuming the distribution to be normal, we estimate the standard deviation of the trends values from the FWHM of the distribution.



**Table S1.** Overview of all HEGIFTOM sites with TrOC data since 2000. The instruments, site names, latitude, longitude, begin and end period of the observations, the total amount of (L1) TrOC observations, and the number of months with (L3) observations are shown.

| <b>Instrument</b> | <b>Site/Airport</b> | <b>lat</b> | <b>lon</b> | <b>begin</b> | <b>end</b> | <b>N<sub>obs</sub></b> | <b>N<sub>months</sub></b> |
|-------------------|---------------------|------------|------------|--------------|------------|------------------------|---------------------------|
| O3S               | Alert               | 82.49      | -62.34     | 2000         | 2020       | 931                    | 227                       |
| FTIR              | Eureka              | 80.05      | -86.42     | 2006         | 2020       | 6261                   | 97                        |
| O3S               | Eureka              | 79.98      | -85.94     | 2000         | 2021       | 1345                   | 248                       |
| O3S               | Ny Alesund          | 78.92      | 11.93      | 2000         | 2022       | 1794                   | 276                       |
| FTIR              | Ny Alesund          | 78.92      | 11.92      | 2000         | 2022       | 2052                   | 119                       |
| FTIR              | Thule               | 76.53      | -68.74     | 2000         | 2022       | 6204                   | 163                       |
| O3S               | Resolute            | 74.70      | -94.96     | 2000         | 2021       | 771                    | 199                       |
| O3S               | Scoresbysund        | 70.48      | -21.97     | 2000         | 2022       | 1127                   | 264                       |
| FTIR              | Kiruna              | 67.84      | 20.40      | 2000         | 2022       | 4853                   | 230                       |
| O3S               | Sodankylä           | 67.37      | 26.65      | 2000         | 2022       | 1074                   | 254                       |
| Umk               | 105 (Fairbanks)     | 64.86      | -147.85    | 2000         | 2021       | 1652                   | 148                       |
| IAGOS             | FAI                 | 64.82      | -147.87    | 2015         | 2016       | 7                      | 3                         |
| IAGOS             | KEF                 | 64.00      | -22.62     | 2003         | 2013       | 14                     | 5                         |
| FTIR              | Harestua            | 60.20      | 10.80      | 2009         | 2020       | 2366                   | 112                       |
| O3S               | Lerwick             | 60.13      | -1.18      | 2000         | 2022       | 1203                   | 243                       |
| FTIR              | Peterhof            | 59.88      | 29.82      | 2009         | 2023       | 5764                   | 133                       |
| O3S               | Churchill           | 58.74      | -94.07     | 2000         | 2021       | 690                    | 183                       |
| IAGOS             | HAM                 | 53.63      | 10.01      | 2000         | 2011       | 5                      | 5                         |
| O3S               | Edmonton            | 53.54      | -114.10    | 2000         | 2021       | 969                    | 244                       |
| O3S               | Goose Bay           | 53.31      | -60.36     | 2000         | 2021       | 953                    | 230                       |
| FTIR              | Bremen              | 53.10      | 8.85       | 2004         | 2023       | 1994                   | 164                       |
| IAGOS             | TXL                 | 52.55      | 13.29      | 2001         | 2021       | 38                     | 8                         |
| O3S               | Legionowo           | 52.40      | 20.97      | 2000         | 2022       | 1340                   | 276                       |
| IAGOS             | AMS                 | 52.31      | 4.76       | 2014         | 2016       | 151                    | 21                        |
| O3S               | De Bilt             | 52.10      | 5.18       | 2000         | 2020       | 1085                   | 252                       |
| O3S               | Valentia            | 51.94      | -10.25     | 2000         | 2022       | 600                    | 127                       |
| IAGOS             | DUS                 | 51.28      | 6.77       | 2000         | 2015       | 716                    | 26                        |
| IAGOS             | LGW                 | 51.16      | -0.16      | 2005         | 2009       | 378                    | 40                        |
| IAGOS             | YYC                 | 51.13      | -114.01    | 2009         | 2011       | 168                    | 17                        |
| IAGOS             | TSE                 | 51.03      | 71.46      | 2006         | 2022       | 214                    | 55                        |
| IAGOS             | BRU                 | 50.90      | 4.48       | 2000         | 2009       | 866                    | 24                        |
| O3S               | Uccle               | 50.80      | 4.35       | 2000         | 2022       | 3265                   | 276                       |
| O3S               | Port Hardy          | 50.68      | -127.38    | 2018         | 2021       | 109                    | 29                        |

|       |                  |       |         |      |      |       |     |
|-------|------------------|-------|---------|------|------|-------|-----|
| IAGOS | FRA              | 50.05 | 8.57    | 2000 | 2022 | 14358 | 246 |
| O3S   | Kelowna          | 49.93 | -119.40 | 2003 | 2017 | 699   | 161 |
| IAGOS | CXH              | 49.27 | -123.12 | 2001 | 2011 | 2     | 2   |
| IAGOS | YVR              | 49.19 | -123.18 | 2000 | 2021 | 556   | 73  |
| IAGOS | CDG              | 49.00 | 2.57    | 2000 | 2022 | 2460  | 82  |
| IAGOS | ORY              | 48.73 | 2.36    | 2002 | 2018 | 149   | 8   |
| IAGOS | MUC              | 48.35 | 11.79   | 2000 | 2011 | 1983  | 51  |
| IAGOS | VIE              | 48.12 | 16.56   | 2000 | 2021 | 2367  | 91  |
| O3S   | Hohenpeissenberg | 47.80 | 11.01   | 2000 | 2023 | 3014  | 285 |
| IAGOS | SEA              | 47.44 | -122.30 | 2013 | 2022 | 113   | 28  |
| FTIR  | Zugspitze        | 47.42 | 10.98   | 2000 | 2022 | 19529 | 264 |
| IAGOS | YQB              | 46.79 | -71.38  | 2015 | 2022 | 5     | 2   |
| Umk   | 35 (Arosa)       | 46.78 | 9.68    | 2000 | 2022 | 2936  | 268 |
| FTIR  | Jungfraujoch     | 46.55 | 7.98    | 2000 | 2023 | 8649  | 261 |
| O3S   | Payerne          | 46.49 | 6.57    | 2002 | 2022 | 3112  | 244 |
| IAGOS | PDX              | 45.59 | -122.59 | 2003 | 2018 | 380   | 54  |
| IAGOS | YUL              | 45.46 | -73.75  | 2000 | 2021 | 272   | 51  |
| IAGOS | MSP              | 44.88 | -93.21  | 2013 | 2018 | 28    | 11  |
| IAGOS | BOD              | 44.83 | -0.70   | 2000 | 2018 | 11    | 7   |
| IAGOS | OTP              | 44.57 | 26.08   | 2004 | 2006 | 4     | 2   |
| FTIR  | Moshiri          | 44.40 | 142.30  | 2000 | 2007 | 1042  | 83  |
| Lidar | OHPO3T           | 43.94 | 5.71    | 2000 | 2022 | 1592  | 237 |
| O3S   | OHP              | 43.94 | 5.71    | 2000 | 2023 | 1075  | 279 |
| Umk   | 40 (OHP)         | 43.94 | 5.71    | 2000 | 2022 | 3596  | 238 |
| IAGOS | URC              | 43.90 | 87.48   | 2016 | 2016 | 2     | 2   |
| O3S   | Yarmouth         | 43.87 | -66.11  | 2003 | 2021 | 793   | 189 |
| IAGOS | YYZ              | 43.68 | -79.61  | 2000 | 2021 | 774   | 95  |
| FTIR  | Toronto          | 43.60 | -79.36  | 2002 | 2023 | 5497  | 210 |
| FTIR  | Rikubetsu        | 43.46 | 143.77  | 2000 | 2022 | 1745  | 191 |
| IAGOS | ALA              | 43.35 | 77.01   | 2001 | 2022 | 257   | 59  |
| IAGOS | CTS              | 42.79 | 141.68  | 2016 | 2018 | 23    | 8   |
| IAGOS | BOS              | 42.37 | -71.02  | 2000 | 2022 | 632   | 102 |
| O3S   | L'Aquila         | 42.30 | 13.31   | 2000 | 2023 | 342   | 116 |
| IAGOS | DTW              | 42.21 | -83.36  | 2000 | 2022 | 404   | 86  |
| IAGOS | ORD              | 41.98 | -87.90  | 2000 | 2021 | 1089  | 91  |
| IAGOS | SHE              | 41.86 | 123.43  | 2012 | 2021 | 219   | 43  |
| IAGOS | FCO              | 41.79 | 12.25   | 2016 | 2021 | 7     | 4   |
| O3S   | Trinidad Head    | 40.80 | -124.16 | 2000 | 2023 | 1239  | 273 |
| IAGOS | EWR              | 40.69 | -74.18  | 2000 | 2021 | 804   | 70  |

|       |                |       |         |      |      |      |     |
|-------|----------------|-------|---------|------|------|------|-----|
| IAGOS | JFK            | 40.64 | -73.79  | 2000 | 2022 | 761  | 88  |
| IAGOS | MAD            | 40.47 | -3.57   | 2014 | 2014 | 15   | 2   |
| O3S   | Madrid         | 40.47 | -3.58   | 2000 | 2022 | 935  | 234 |
| IAGOS | GYD            | 40.46 | 50.05   | 2001 | 2013 | 69   | 14  |
| IAGOS | PEK            | 40.08 | 116.59  | 2000 | 2021 | 236  | 50  |
| FTIR  | Boulder        | 40.04 | -105.24 | 2010 | 2022 | 3613 | 122 |
| O3S   | Boulder        | 40.00 | -105.25 | 2000 | 2023 | 1276 | 283 |
| Umk   | 67 (Boulder)   | 39.99 | -105.26 | 2000 | 2022 | 4721 | 272 |
| IAGOS | PHL            | 39.88 | -75.24  | 2000 | 2022 | 686  | 115 |
| IAGOS | DEN            | 39.85 | -104.67 | 2001 | 2022 | 48   | 18  |
| IAGOS | TSN            | 39.12 | 117.34  | 2020 | 2020 | 2    | 2   |
| IAGOS | CVG            | 39.06 | -84.66  | 2000 | 2004 | 6    | 2   |
| IAGOS | IAD            | 38.95 | -77.45  | 2000 | 2022 | 497  | 81  |
| IAGOS | LIS            | 38.77 | -9.13   | 2002 | 2021 | 5    | 3   |
| IAGOS | ASB            | 37.98 | 58.37   | 2001 | 2013 | 34   | 14  |
| IAGOS | ATH            | 37.94 | 23.95   | 2003 | 2017 | 8    | 5   |
| O3S   | Wallops Island | 37.93 | -75.48  | 2000 | 2020 | 1143 | 245 |
| IAGOS | SFO            | 37.62 | -122.39 | 2000 | 2018 | 153  | 39  |
| IAGOS | GMP            | 37.56 | 126.80  | 2000 | 2000 | 28   | 8   |
| IAGOS | ICN            | 37.45 | 126.45  | 2001 | 2022 | 171  | 35  |
| IAGOS | AYT            | 36.90 | 30.80   | 2000 | 2006 | 23   | 6   |
| IAGOS | DLM            | 36.72 | 28.78   | 2005 | 2005 | 10   | 3   |
| IAGOS | RHO            | 36.40 | 28.09   | 2002 | 2002 | 11   | 4   |
| IAGOS | TAO            | 36.27 | 120.38  | 2012 | 2022 | 127  | 48  |
| IAGOS | LAS            | 36.09 | -115.15 | 2017 | 2018 | 74   | 12  |
| FTIR  | Tsukuba        | 36.05 | 140.12  | 2014 | 2020 | 780  | 74  |
| IAGOS | MLA            | 35.86 | 14.48   | 2011 | 2019 | 13   | 9   |
| IAGOS | NRT            | 35.77 | 140.39  | 2000 | 2018 | 855  | 96  |
| IAGOS | THR            | 35.69 | 51.32   | 2000 | 2005 | 202  | 46  |
| IAGOS | HND            | 35.55 | 139.78  | 2017 | 2022 | 66   | 17  |
| IAGOS | IKA            | 35.41 | 51.15   | 2009 | 2022 | 150  | 35  |
| IAGOS | NKM            | 35.25 | 136.92  | 2000 | 2004 | 26   | 14  |
| IAGOS | CLT            | 35.22 | -80.94  | 2004 | 2011 | 36   | 9   |
| IAGOS | PUS            | 35.18 | 128.95  | 2016 | 2018 | 9    | 5   |
| IAGOS | NGO            | 34.86 | 136.81  | 2006 | 2020 | 413  | 76  |
| IAGOS | CGO            | 34.53 | 113.84  | 2016 | 2016 | 2    | 2   |
| IAGOS | KIX            | 34.44 | 135.24  | 2000 | 2018 | 634  | 83  |
| IAGOS | HIJ            | 34.44 | 132.92  | 2016 | 2017 | 4    | 2   |
| IAGOS | XIY            | 34.44 | 108.76  | 2016 | 2017 | 7    | 3   |

|       |       |       |         |      |      |      |     |
|-------|-------|-------|---------|------|------|------|-----|
| Lidar | TMF   | 34.38 | -117.68 | 2000 | 2023 | 2918 | 276 |
| IAGOS | LAX   | 33.94 | -118.41 | 2000 | 2018 | 255  | 34  |
| IAGOS | BEY   | 33.83 | 35.49   | 2013 | 2022 | 25   | 8   |
| IAGOS | ATL   | 33.64 | -84.44  | 2000 | 2022 | 1465 | 139 |
| IAGOS | FUK   | 33.58 | 130.44  | 2015 | 2018 | 75   | 14  |
| IAGOS | PHX   | 33.44 | -112.00 | 2001 | 2018 | 17   | 6   |
| IAGOS | DAL   | 32.84 | -96.85  | 2000 | 2022 | 734  | 131 |
| IAGOS | SAN   | 32.73 | -117.20 | 2017 | 2020 | 89   | 21  |
| IAGOS | TLV   | 32.00 | 34.87   | 2000 | 2022 | 305  | 59  |
| FTIR  | Hefei | 31.91 | 117.17  | 2015 | 2020 | 3375 | 55  |
| IAGOS | NKG   | 31.74 | 118.87  | 2009 | 2020 | 69   | 41  |
| IAGOS | AMM   | 31.72 | 35.99   | 2006 | 2020 | 3    | 2   |
| IAGOS | WUX   | 31.49 | 120.42  | 2016 | 2018 | 8    | 4   |
| IAGOS | SHA   | 31.20 | 121.34  | 2000 | 2000 | 11   | 3   |
| IAGOS | PVG   | 31.15 | 121.80  | 2000 | 2021 | 182  | 41  |
| IAGOS | WUH   | 30.78 | 114.21  | 2017 | 2018 | 17   | 8   |
| IAGOS | AUS   | 30.20 | -97.67  | 2019 | 2022 | 23   | 9   |
| IAGOS | CAI   | 30.12 | 31.41   | 2000 | 2019 | 320  | 44  |
| IAGOS | IAH   | 29.98 | -95.34  | 2000 | 2022 | 266  | 62  |
| IAGOS | NGB   | 29.82 | 121.46  | 2012 | 2018 | 11   | 8   |
| IAGOS | KWI   | 29.24 | 47.97   | 2001 | 2022 | 417  | 80  |
| IAGOS | DEL   | 28.56 | 77.10   | 2000 | 2021 | 607  | 77  |
| IAGOS | MCO   | 28.43 | -81.31  | 2008 | 2022 | 27   | 10  |
| FTIR  | Izaña | 28.30 | -16.48  | 2000 | 2022 | 7665 | 259 |
| O3S   | Izaña | 28.30 | -16.50  | 2000 | 2022 | 1086 | 270 |
| IAGOS | CSX   | 28.19 | 113.21  | 2015 | 2018 | 6    | 3   |
| IAGOS | TPA   | 27.98 | -82.54  | 2018 | 2022 | 11   | 5   |
| IAGOS | LPA   | 27.94 | -15.39  | 2002 | 2005 | 2    | 2   |
| IAGOS | KTM   | 27.70 | 85.36   | 2003 | 2006 | 10   | 5   |
| IAGOS | HRG   | 27.19 | 33.81   | 2000 | 2001 | 13   | 3   |
| IAGOS | DMM   | 26.47 | 49.80   | 2003 | 2022 | 68   | 31  |
| IAGOS | BAH   | 26.27 | 50.63   | 2010 | 2021 | 14   | 10  |
| IAGOS | OKA   | 26.20 | 127.65  | 2015 | 2018 | 31   | 10  |
| IAGOS | MIA   | 25.80 | -80.28  | 2001 | 2021 | 51   | 16  |
| IAGOS | LXR   | 25.67 | 32.70   | 2001 | 2001 | 4    | 4   |
| IAGOS | DOH   | 25.27 | 51.56   | 2012 | 2017 | 131  | 28  |
| IAGOS | DXB   | 25.25 | 55.35   | 2000 | 2022 | 448  | 88  |
| IAGOS | TPE   | 25.08 | 121.22  | 2012 | 2018 | 1422 | 40  |
| IAGOS | RUH   | 24.96 | 46.70   | 2000 | 2022 | 287  | 81  |

|       |                |       |         |      |      |      |     |
|-------|----------------|-------|---------|------|------|------|-----|
| IAGOS | XMN            | 24.54 | 118.13  | 2012 | 2018 | 31   | 14  |
| IAGOS | AUH            | 24.43 | 54.65   | 2003 | 2017 | 65   | 20  |
| IAGOS | MCT            | 23.59 | 58.29   | 2003 | 2015 | 38   | 12  |
| IAGOS | CAN            | 23.39 | 113.30  | 2009 | 2018 | 88   | 17  |
| IAGOS | KHH            | 22.57 | 120.35  | 2018 | 2018 | 6    | 2   |
| IAGOS | HKG            | 22.32 | 113.94  | 2000 | 2021 | 719  | 68  |
| IAGOS | JED            | 21.67 | 39.15   | 2001 | 2019 | 238  | 50  |
| IAGOS | HNL            | 21.33 | -157.92 | 2016 | 2018 | 239  | 24  |
| IAGOS | HAN            | 21.21 | 105.80  | 2016 | 2018 | 20   | 8   |
| IAGOS | CUN            | 21.04 | -86.87  | 2001 | 2021 | 31   | 10  |
| O3S   | Hanoi          | 21.01 | 105.80  | 2004 | 2021 | 350  | 121 |
| IAGOS | OGG            | 20.89 | -156.44 | 2017 | 2018 | 23   | 8   |
| IAGOS | POP            | 19.75 | -70.56  | 2001 | 2002 | 2    | 2   |
| IAGOS | KOA            | 19.74 | -156.04 | 2017 | 2018 | 4    | 3   |
| FTIR  | Mauna Loa      | 19.54 | -155.57 | 2000 | 2022 | 9025 | 165 |
| Umk   | 31 (Mauna Loa) | 19.54 | -155.58 | 2000 | 2022 | 7822 | 266 |
| O3S   | Hilo           | 19.43 | -155.04 | 2000 | 2023 | 1174 | 284 |
| FTIR  | Altzomoni      | 19.12 | -98.68  | 2012 | 2022 | 4307 | 100 |
| IAGOS | BOM            | 19.10 | 72.87   | 2000 | 2022 | 95   | 34  |
| IAGOS | PUJ            | 18.56 | -68.36  | 2021 | 2021 | 23   | 4   |
| IAGOS | NKC            | 18.10 | -15.95  | 2003 | 2022 | 64   | 15  |
| IAGOS | SXM            | 18.04 | -63.11  | 2013 | 2022 | 44   | 17  |
| IAGOS | HYD            | 17.24 | 78.43   | 2005 | 2018 | 485  | 62  |
| IAGOS | KRT            | 15.59 | 32.55   | 2000 | 2014 | 260  | 31  |
| IAGOS | SAH            | 15.47 | 44.23   | 2000 | 2004 | 10   | 4   |
| IAGOS | ASM            | 15.29 | 38.91   | 2002 | 2012 | 18   | 5   |
| IAGOS | DKR            | 14.74 | -17.49  | 2000 | 2009 | 50   | 12  |
| IAGOS | MNL            | 14.51 | 121.01  | 2005 | 2018 | 73   | 19  |
| IAGOS | DMK            | 13.91 | 100.61  | 2005 | 2006 | 211  | 13  |
| IAGOS | BKK            | 13.69 | 100.75  | 2006 | 2022 | 272  | 43  |
| IAGOS | GUM            | 13.49 | 144.80  | 2016 | 2016 | 2    | 2   |
| IAGOS | NIM            | 13.48 | 2.18    | 2002 | 2022 | 73   | 22  |
| IAGOS | BLR            | 13.20 | 77.71   | 2017 | 2022 | 50   | 10  |
| IAGOS | MAA            | 12.98 | 80.16   | 2000 | 2018 | 315  | 59  |
| IAGOS | BKO            | 12.54 | -7.95   | 2013 | 2017 | 29   | 15  |
| IAGOS | OUA            | 12.36 | -1.51   | 2002 | 2022 | 61   | 27  |
| IAGOS | NDJ            | 12.13 | 15.03   | 2013 | 2022 | 26   | 10  |
| IAGOS | JIB            | 11.55 | 43.15   | 2013 | 2018 | 9    | 6   |
| IAGOS | SGN            | 10.81 | 106.66  | 2005 | 2018 | 130  | 26  |

|       |                  |       |        |      |      |     |     |
|-------|------------------|-------|--------|------|------|-----|-----|
| IAGOS | CCS              | 10.60 | -67.01 | 2000 | 2018 | 722 | 104 |
| IAGOS | CEB              | 10.31 | 123.98 | 2018 | 2018 | 10  | 2   |
| IAGOS | SJO              | 10.00 | -84.20 | 2014 | 2022 | 42  | 20  |
| O3S   | Costa Rica       | 9.94  | -84.04 | 2005 | 2023 | 638 | 176 |
| IAGOS | CKY              | 9.58  | -13.62 | 2003 | 2022 | 58  | 25  |
| IAGOS | PTY              | 9.07  | -79.39 | 2017 | 2020 | 24  | 8   |
| IAGOS | ABV              | 9.00  | 7.27   | 2003 | 2022 | 247 | 54  |
| IAGOS | ADD              | 8.98  | 38.80  | 2001 | 2019 | 242 | 58  |
| IAGOS | FNA              | 8.62  | -13.20 | 2009 | 2022 | 9   | 5   |
| IAGOS | LOS              | 6.58  | 3.32   | 2000 | 2022 | 476 | 93  |
| IAGOS | COO              | 6.35  | 2.39   | 2001 | 2022 | 68  | 32  |
| IAGOS | LFW              | 6.17  | 1.25   | 2003 | 2022 | 60  | 26  |
| FTIR  | Paramaribo       | 5.81  | -55.20 | 2004 | 2016 | 234 | 17  |
| O3S   | Paramaribo       | 5.80  | -55.21 | 2000 | 2022 | 855 | 247 |
| IAGOS | ACC              | 5.61  | -0.17  | 2003 | 2022 | 45  | 19  |
| IAGOS | ABJ              | 5.25  | -3.93  | 2000 | 2022 | 118 | 26  |
| IAGOS | PHC              | 5.01  | 6.95   | 2002 | 2022 | 79  | 39  |
| IAGOS | CAY              | 4.82  | -52.37 | 2002 | 2022 | 112 | 8   |
| IAGOS | BOG              | 4.70  | -74.14 | 2000 | 2021 | 292 | 30  |
| IAGOS | MLE              | 4.20  | 73.52  | 2000 | 2022 | 27  | 12  |
| IAGOS | DLA              | 4.01  | 9.72   | 2000 | 2022 | 125 | 30  |
| IAGOS | SSG              | 3.76  | 8.72   | 2014 | 2022 | 126 | 41  |
| IAGOS | NSI              | 3.70  | 11.55  | 2017 | 2022 | 18  | 5   |
| IAGOS | KUL              | 2.76  | 101.71 | 2005 | 2016 | 72  | 15  |
| O3S   | Kuala Lumpur     | 2.73  | 101.27 | 2000 | 2022 | 456 | 203 |
| IAGOS | SIN              | 1.36  | 103.99 | 2000 | 2022 | 78  | 19  |
| IAGOS | LBV              | 0.46  | 9.41   | 2000 | 2022 | 12  | 7   |
| O3S   | San Cristobal    | -0.92 | -89.62 | 2000 | 2022 | 350 | 82  |
| O3S   | Nairobi          | -1.27 | 36.80  | 2000 | 2022 | 872 | 223 |
| IAGOS | NBO              | -1.32 | 36.93  | 2018 | 2022 | 89  | 28  |
| IAGOS | BZV              | -4.26 | 15.25  | 2002 | 2022 | 31  | 9   |
| IAGOS | FIH              | -4.39 | 15.45  | 2000 | 2022 | 73  | 21  |
| IAGOS | PNR              | -4.81 | 11.88  | 2014 | 2022 | 29  | 11  |
| O3S   | Natal            | -5.42 | -35.38 | 2000 | 2022 | 676 | 175 |
| IAGOS | CGK              | -6.13 | 106.66 | 2005 | 2018 | 54  | 11  |
| IAGOS | SUB              | -7.38 | 112.78 | 2016 | 2018 | 14  | 5   |
| O3S   | Watukosek        | -7.50 | 112.60 | 2000 | 2022 | 326 | 75  |
| O3S   | Ascension Island | -7.58 | -14.24 | 2000 | 2022 | 676 | 174 |
| IAGOS | DPS              | -8.74 | 115.16 | 2016 | 2018 | 20  | 7   |

|       |                 |        |         |      |      |       |     |
|-------|-----------------|--------|---------|------|------|-------|-----|
| FTIR  | Porto Velho     | -8.77  | -63.87  | 2019 | 2019 | 491   | 5   |
| IAGOS | LAD             | -8.85  | 13.23   | 2000 | 2022 | 175   | 68  |
| O3S   | Samoa           | -14.23 | -170.56 | 2000 | 2023 | 821   | 241 |
| IAGOS | HRE             | -17.92 | 31.10   | 2000 | 2000 | 20    | 8   |
| O3S   | Fiji            | -18.13 | 178.40  | 2000 | 2023 | 397   | 125 |
| IAGOS | TNR             | -18.80 | 47.48   | 2016 | 2017 | 9     | 3   |
| FTIR  | St Denis        | -20.90 | 55.48   | 2004 | 2011 | 1778  | 37  |
| O3S   | Reunion Island  | -21.06 | 55.48   | 2000 | 2022 | 735   | 215 |
| FTIR  | Maido           | -21.08 | 55.38   | 2013 | 2023 | 4460  | 76  |
| IAGOS | WDH             | -22.49 | 17.46   | 2000 | 2013 | 1618  | 69  |
| IAGOS | GIG             | -22.81 | -43.25  | 2000 | 2014 | 58    | 25  |
| IAGOS | GRU             | -23.43 | -46.48  | 2000 | 2021 | 23    | 11  |
| O3S   | Irene           | -25.90 | 28.22   | 2000 | 2023 | 391   | 140 |
| IAGOS | JNB             | -26.13 | 28.23   | 2000 | 2021 | 77    | 19  |
| IAGOS | BNE             | -27.40 | 153.11  | 2016 | 2018 | 61    | 11  |
| IAGOS | SYD             | -33.93 | 151.18  | 2013 | 2018 | 85    | 18  |
| IAGOS | CPT             | -33.97 | 18.60   | 2005 | 2021 | 103   | 34  |
| FTIR  | Wollongong      | -34.41 | 150.88  | 2007 | 2023 | 8481  | 150 |
| IAGOS | EZE             | -34.81 | -58.54  | 2014 | 2014 | 4     | 2   |
| IAGOS | AKL             | -37.00 | 174.78  | 2016 | 2018 | 59    | 13  |
| IAGOS | MEL             | -37.67 | 144.85  | 2016 | 2018 | 25    | 8   |
| IAGOS | CHC             | -43.49 | 172.54  | 2016 | 2018 | 20    | 6   |
| O3S   | Lauder          | -45.00 | 169.68  | 2000 | 2021 | 923   | 237 |
| FTIR  | Lauder          | -45.04 | 169.68  | 2001 | 2023 | 10389 | 252 |
| Umk   | 256 (Lauder)    | -45.04 | 169.68  | 2000 | 2022 | 2957  | 262 |
| FTIR  | Arrival Heights | -77.82 | 166.65  | 2000 | 2022 | 2563  | 176 |
| O3S   | Mc Murdo        | -77.85 | 166.67  | 2000 | 2010 | 295   | 36  |
| O3S   | South Pole      | -90.00 | 169.68  | 2000 | 2023 | 1371  | 279 |

**Table S2.** Results of the intercomparison analysis between all coincident and collocated measurements (L1) for column-averaged tropospheric ozone column amounts (in ppbv, from surface to 300 hPa). The different columns show the sites (filenames of the time series at the HEGIFTOM ftp-server have been used), the number of coincident observations, the mean bias, the standard deviation of differences, the linear Pearson correlation coefficient, the slope and offset of the linear regression line. The rows are ordered in blocks according to the instruments compared with each other. The intercomparison selection criteria are sites within  $\pm 4^\circ$  in latitude and longitude, observations coincident within 12h (closest measurements) and requiring at least 15 coincident measurements.

| site1                | site2            | N    | bias<br>(2 - 1) | stdev | r    | slope | offset |
|----------------------|------------------|------|-----------------|-------|------|-------|--------|
| mcmurdo_O3S          | Ahts_FTIR        | 39   | 3.80            | 3.42  | 0.56 | 0.57  | 18.42  |
| lauder_O3S           | Lauder_FTIR      | 309  | 2.54            | 5.47  | 0.61 | 0.76  | 11.22  |
| hilo_O3S             | Mauna_Loa_FTIR   | 389  | 10.57           | 8.32  | 0.80 | 1.16  | 4.31   |
| izana_O3S            | Izana_FTIR       | 431  | 5.37            | 7.68  | 0.82 | 1.07  | 1.80   |
| boulder_O3S          | Boulder_FTIR     | 211  | 1.36            | 6.41  | 0.79 | 0.90  | 7.25   |
| payerne_O3S          | Jungfrauoch_FTIR | 825  | 2.70            | 9.53  | 0.70 | 0.89  | 8.99   |
| hohenpeissenberg_O3S | Zugspitze_FTIR   | 1324 | 11.41           | 14.25 | 0.49 | 0.88  | 17.83  |
| sodankyla_O3S        | Kiruna_FTIR      | 344  | 6.07            | 9.92  | 0.53 | 0.73  | 20.95  |
| nyalesund_O3S        | Ny_Alesund_FTIR  | 118  | 3.80            | 9.81  | 0.53 | 0.51  | 33.57  |
| eureka_O3S           | Eureka_FTIR      | 239  | 9.90            | 6.82  | 0.73 | 0.75  | 23.12  |
| yarmouth_O3S         | BOS_IAGOS        | 20   | -1.84           | 7.17  | 0.72 | 0.95  | 1.33   |
| kelowna_O3S          | PDX_IAGOS        | 37   | 0.06            | 9.29  | 0.75 | 0.86  | 7.89   |
| kelowna_O3S          | YVR_IAGOS        | 19   | 1.81            | 9.28  | 0.66 | 1.43  | -19.51 |
| payerne_O3S          | FRA_IAGOS        | 2115 | -0.69           | 8.85  | 0.66 | 0.68  | 16.90  |
| hohenpeissenberg_O3S | MUC_IAGOS        | 303  | 2.85            | 7.45  | 0.74 | 0.80  | 12.84  |
| uccle_O3S            | ORY_IAGOS        | 35   | -2.82           | 8.94  | 0.29 | 0.28  | 37.02  |
| uccle_O3S            | CDG_IAGOS        | 1028 | -2.83           | 9.40  | 0.61 | 0.62  | 17.83  |
| uccle_O3S            | BRU_IAGOS        | 481  | 0.40            | 10.00 | 0.64 | 0.77  | 13.40  |
| debilt_O3S           | DUS_IAGOS        | 84   | -1.36           | 6.61  | 0.84 | 0.79  | 10.94  |
| legionowo_O3S        | VIE_IAGOS        | 458  | -1.95           | 10.01 | 0.66 | 0.70  | 15.62  |
| lauder_O3S           | 256_Umk          | 445  | -2.07           | 9.57  | 0.04 | 0.04  | 33.33  |
| hilo_O3S             | 031_Umk          | 43   | -9.26           | 7.72  | 0.57 | 0.51  | 9.15   |
| boulder_O3S          | 067_Umk          | 24   | -10.36          | 10.93 | 0.13 | 0.14  | 37.39  |
| ohp_O3S              | 040_Umk          | 456  | -10.05          | 12.72 | 0.44 | 0.60  | 13.00  |
| payerne_O3S          | 035_Umk          | 760  | -5.30           | 10.24 | 0.59 | 0.67  | 12.97  |
| sodankyla_O3S        | 105_Umk          | 224  | 0.97            | 12.36 | 0.29 | 0.46  | 31.91  |
| Mauna_Loa_FTIR       | HNL_IAGOS        | 55   | -6.12           | 8.54  | 0.76 | 0.61  | 13.91  |
| Tsukuba_FTIR         | KIX_IAGOS        | 17   | -5.17           | 6.62  | 0.12 | 0.07  | 45.55  |
| Tsukuba_FTIR         | NRT_IAGOS        | 17   | -3.81           | 8.88  | 0.35 | 0.26  | 38.52  |
| Toronto_FTIR         | DTW_IAGOS        | 54   | -5.54           | 11.44 | 0.54 | 0.47  | 27.74  |
| Toronto_FTIR         | YYZ_IAGOS        | 63   | -5.38           | 16.41 | 0.58 | 0.32  | 38.04  |



|                  |           |      |       |       |      |      |        |
|------------------|-----------|------|-------|-------|------|------|--------|
| Jungfrauoch_FTIR | FRA_IAGOS | 2330 | -5.92 | 11.24 | 0.57 | 0.46 | 26.94  |
| Zugspitze_FTIR   | MUC_IAGOS | 363  | -9.83 | 13.56 | 0.55 | 0.33 | 33.26  |
| Lauder_FTIR      | 256_Umk   | 950  | -5.09 | 8.12  | 0.29 | 0.31 | 21.67  |
| Jungfrauoch_FTIR | 035_Umk   | 1080 | -7.76 | 9.92  | 0.66 | 0.64 | 13.01  |
| Kiruna_FTIR      | 105_Umk   | 373  | -6.50 | 14.90 | 0.30 | 0.32 | 36.88  |
| 031_Umk          | HNL_IAGOS | 46   | 8.46  | 7.46  | 0.59 | 0.80 | 15.83  |
| 035_Umk          | FRA_IAGOS | 2209 | 2.46  | 11.18 | 0.52 | 0.44 | 30.00  |
| KIX_IAGOS        | NKM_IAGOS | 15   | 0.68  | 5.64  | 0.94 | 1.17 | -10.00 |
| KIX_IAGOS        | NRT_IAGOS | 44   | 0.76  | 11.66 | 0.57 | 0.46 | 29.19  |
| CDG_IAGOS        | BRU_IAGOS | 743  | 1.55  | 10.57 | 0.58 | 0.67 | 19.60  |
| DUS_IAGOS        | AMS_IAGOS | 22   | 2.13  | 7.63  | 0.58 | 0.53 | 27.49  |
| TMF_Lidar        | LAX_IAGOS | 33   | -2.80 | 6.55  | 0.84 | 0.93 | 1.10   |
| TMF_Lidar        | LAS_IAGOS | 17   | -4.97 | 8.67  | 0.74 | 0.57 | 20.64  |
| OHPO3T_Lidar     | ohp_O3S   | 205  | -5.43 | 11.90 | 0.52 | 0.41 | 31.03  |

**Table S3.** Same as Table S2, but now for the column-averaged tropospheric ozone column monthly means (in ppbv). The coincidence criterion is now that monthly means from the same month are compared.

| site1                | site2            | N   | Bias<br>(2 - 1) | stdev | r     | slope | offset |
|----------------------|------------------|-----|-----------------|-------|-------|-------|--------|
| mcmurdo_O3S          | Ahts_FTIR        | 26  | 3.50            | 2.13  | 0.54  | 0.58  | 18.08  |
| lauder_O3S           | Lauder_FTIR      | 210 | 2.31            | 3.55  | 0.76  | 0.94  | 4.33   |
| paramaribo_O3S       | Paramaribo_FTIR  | 16  | 2.70            | 6.79  | 0.44  | 0.45  | 21.19  |
| hilo_O3S             | Mauna_Loa_FTIR   | 200 | 10.49           | 6.05  | 0.78  | 0.99  | 10.97  |
| izana_O3S            | Izana_FTIR       | 257 | 4.96            | 5.83  | 0.84  | 1.13  | -2.03  |
| boulder_O3S          | Boulder_FTIR     | 120 | 2.27            | 6.38  | 0.67  | 0.83  | 11.51  |
| payerne_O3S          | Jungfrauoch_FTIR | 226 | 4.51            | 6.12  | 0.78  | 0.88  | 10.86  |
| hohenpeissenberg_O3S | Zugspitze_FTIR   | 305 | 11.10           | 10.39 | 0.51  | 0.85  | 19.02  |
| sodankyla_O3S        | Kiruna_FTIR      | 252 | 5.05            | 6.84  | 0.70  | 0.90  | 10.78  |
| nyalesund_O3S        | Ny_Alesund_FTIR  | 146 | 3.45            | 7.20  | 0.61  | 0.87  | 11.17  |
| eureka_O3S           | Eureka_FTIR      | 94  | 10.18           | 6.02  | 0.76  | 0.96  | 12.53  |
| irene_O3S            | JNB_IAGOS        | 19  | -6.68           | 8.42  | 0.66  | 0.58  | 16.63  |
| nairobi_O3S          | NBO_IAGOS        | 20  | -1.46           | 6.50  | 0.63  | 0.62  | 13.82  |
| kualalumpur_O3S      | SIN_IAGOS        | 22  | -2.36           | 8.70  | 0.66  | 1.15  | -7.09  |
| hilo_O3S             | HNL_IAGOS        | 23  | 5.63            | 6.27  | 0.50  | 0.62  | 20.09  |
| boulder_O3S          | DEN_IAGOS        | 17  | -0.85           | 6.60  | 0.66  | 0.91  | 4.40   |
| yarmouth_O3S         | BOS_IAGOS        | 53  | -3.02           | 6.20  | 0.63  | 0.73  | 13.19  |
| kelowna_O3S          | PDX_IAGOS        | 43  | -1.38           | 7.75  | 0.43  | 0.59  | 20.16  |
| kelowna_O3S          | YVR_IAGOS        | 56  | 0.07            | 6.01  | 0.63  | 0.82  | 9.07   |
| payerne_O3S          | FRA_IAGOS        | 214 | -0.11           | 4.36  | 0.86  | 0.84  | 8.35   |
| hohenpeissenberg_O3S | MUC_IAGOS        | 55  | 2.55            | 4.06  | 0.88  | 0.94  | 5.71   |
| uccle_O3S            | CDG_IAGOS        | 119 | -1.85           | 4.70  | 0.83  | 0.86  | 5.94   |
| uccle_O3S            | BRU_IAGOS        | 54  | 0.01            | 3.26  | 0.93  | 1.07  | -3.76  |
| debilt_O3S           | DUS_IAGOS        | 61  | 2.63            | 5.85  | 0.84  | 1.01  | 2.35   |
| debilt_O3S           | AMS_IAGOS        | 22  | 1.03            | 4.84  | 0.83  | 0.92  | 5.44   |
| edmonton_O3S         | YYC_IAGOS        | 16  | -0.17           | 5.29  | 0.64  | 0.71  | 14.90  |
| legionowo_O3S        | VIE_IAGOS        | 146 | -1.78           | 5.22  | 0.86  | 0.84  | 7.44   |
| lauder_O3S           | 256_Umk          | 342 | -1.59           | 6.94  | -0.11 | -0.10 | 38.43  |
| hilo_O3S             | 031_Umk          | 398 | -5.20           | 5.37  | 0.73  | 0.58  | 11.72  |
| boulder_O3S          | 067_Umk          | 400 | -12.14          | 6.43  | 0.63  | 0.46  | 17.95  |
| ohp_O3S              | 040_Umk          | 303 | -10.14          | 7.49  | 0.70  | 0.81  | 0.34   |
| payerne_O3S          | 035_Umk          | 235 | -4.69           | 4.95  | 0.83  | 0.81  | 5.65   |
| sodankyla_O3S        | 105_Umk          | 172 | -0.73           | 7.66  | 0.46  | 0.56  | 24.21  |
| Mauna_Loa_FTIR       | HNL_IAGOS        | 22  | -5.65           | 5.61  | 0.71  | 0.63  | 12.92  |
| Tsukuba_FTIR         | KIX_IAGOS        | 19  | -4.88           | 7.51  | 0.71  | 0.53  | 23.36  |

|                  |           |     |        |       |      |      |       |
|------------------|-----------|-----|--------|-------|------|------|-------|
| Tsukuba_FTIR     | NGO_IAGOS | 34  | -5.00  | 8.33  | 0.76 | 0.72 | 13.09 |
| Tsukuba_FTIR     | NRT_IAGOS | 18  | -8.09  | 6.99  | 0.86 | 0.51 | 22.43 |
| Toronto_FTIR     | DTW_IAGOS | 58  | -9.54  | 8.33  | 0.55 | 0.54 | 19.22 |
| Toronto_FTIR     | YYZ_IAGOS | 60  | -6.31  | 7.83  | 0.62 | 0.52 | 22.63 |
| Jungfrauoch_FTIR | FRA_IAGOS | 230 | -5.26  | 6.23  | 0.78 | 0.68 | 13.76 |
| Zugspitze_FTIR   | MUC_IAGOS | 50  | -11.45 | 10.36 | 0.54 | 0.38 | 28.77 |
| Lauder_FTIR      | 256_Umk   | 235 | -4.34  | 6.65  | 0.10 | 0.07 | 31.48 |
| Boulder_FTIR     | 067_Umk   | 121 | -12.95 | 6.37  | 0.66 | 0.49 | 16.45 |
| Jungfrauoch_FTIR | 035_Umk   | 250 | -9.90  | 7.30  | 0.68 | 0.57 | 15.55 |
| Kiruna_FTIR      | 105_Umk   | 171 | -5.13  | 8.72  | 0.51 | 0.46 | 27.58 |
| 031_Umk          | HNL_IAGOS | 21  | 7.58   | 5.04  | 0.71 | 1.09 | 4.48  |
| 067_Umk          | DEN_IAGOS | 15  | 10.28  | 9.35  | 0.13 | 0.19 | 46.08 |
| 035_Umk          | FRA_IAGOS | 303 | 3.68   | 6.57  | 0.69 | 0.75 | 16.40 |
| KIX_IAGOS        | NGO_IAGOS | 16  | -0.72  | 6.53  | 0.81 | 0.97 | 0.85  |
| KIX_IAGOS        | NKM_IAGOS | 36  | 0.05   | 8.30  | 0.77 | 1.05 | -2.86 |
| KIX_IAGOS        | NRT_IAGOS | 111 | 2.59   | 8.69  | 0.62 | 0.62 | 22.85 |
| NGO_IAGOS        | NRT_IAGOS | 20  | 1.30   | 11.15 | 0.40 | 0.29 | 39.09 |
| NKM_IAGOS        | NRT_IAGOS | 23  | 3.26   | 8.56  | 0.76 | 0.58 | 26.26 |
| DTW_IAGOS        | YYZ_IAGOS | 36  | 0.22   | 7.02  | 0.65 | 0.58 | 22.21 |
| PDX_IAGOS        | YVR_IAGOS | 18  | 2.66   | 11.28 | 0.04 | 0.06 | 46.50 |
| CDG_IAGOS        | BRU_IAGOS | 45  | 0.89   | 3.14  | 0.93 | 1.10 | -4.30 |
| TMF_Lidar        | SAN_IAGOS | 20  | -4.81  | 6.56  | 0.70 | 0.59 | 18.77 |
| TMF_Lidar        | LAX_IAGOS | 33  | -3.13  | 5.38  | 0.80 | 0.83 | 6.53  |
| OHPO3T_Lidar     | ohp_O3S   | 235 | -5.98  | 7.83  | 0.64 | 0.57 | 20.65 |
| OHPO3T_Lidar     | 040_Umk   | 206 | -16.41 | 9.86  | 0.46 | 0.45 | 17.63 |

**Table S4.** Same as Table S2, but now for the tropospheric ozone column amounts from the surface to 300 hPa in Dobson unit (DU).

| site1                | site2            | N    | Bias<br>(2 - 1) | stdev | r    | slope | offset |
|----------------------|------------------|------|-----------------|-------|------|-------|--------|
| mcmurdo_O3S          | Ahts_FTIR        | 39   | 1.41            | 1.83  | 0.59 | 0.55  | 9.77   |
| lauder_O3S           | Lauder_FTIR      | 309  | 1.31            | 2.85  | 0.62 | 0.75  | 6.11   |
| hilo_O3S             | Mauna_Loa_FTIR   | 389  | -7.44           | 3.34  | 0.79 | 0.59  | 1.91   |
| izana_O3S            | Izana_FTIR       | 431  | -6.57           | 3.29  | 0.82 | 0.77  | 0.56   |
| boulder_O3S          | Boulder_FTIR     | 211  | 1.19            | 2.73  | 0.79 | 0.90  | 3.53   |
| payerne_O3S          | Jungfrauoch_FTIR | 825  | -12.01          | 3.66  | 0.73 | 0.53  | 1.97   |
| hohenpeissenberg_O3S | Zugspitze_FTIR   | 1324 | -4.40           | 4.73  | 0.51 | 0.59  | 5.80   |
| sodankyla_O3S        | Kiruna_FTIR      | 344  | 3.97            | 5.47  | 0.52 | 0.71  | 12.56  |
| nyalesund_O3S        | Ny_Alesund_FTIR  | 118  | 2.32            | 5.60  | 0.52 | 0.50  | 19.34  |
| eureka_O3S           | Eureka_FTIR      | 239  | 1.79            | 4.28  | 0.69 | 0.57  | 14.91  |
| yarmouth_O3S         | BOS_IAGOS        | 20   | -2.93           | 2.99  | 0.81 | 0.92  | -0.18  |
| kelowna_O3S          | PDX_IAGOS        | 37   | 0.76            | 5.44  | 0.68 | 0.56  | 13.27  |
| kelowna_O3S          | YVR_IAGOS        | 19   | 0.89            | 3.97  | 0.65 | 1.23  | -4.98  |
| payerne_O3S          | FRA_IAGOS        | 2115 | -0.14           | 4.19  | 0.70 | 0.68  | 8.96   |
| hohenpeissenberg_O3S | MUC_IAGOS        | 303  | 2.53            | 3.40  | 0.76 | 0.82  | 6.97   |
| uccle_O3S            | ORY_IAGOS        | 35   | -3.27           | 4.24  | 0.40 | 0.30  | 18.08  |
| uccle_O3S            | CDG_IAGOS        | 1028 | -2.98           | 4.73  | 0.64 | 0.59  | 9.52   |
| uccle_O3S            | BRU_IAGOS        | 481  | -1.28           | 4.38  | 0.72 | 0.73  | 7.09   |
| debilt_O3S           | DUS_IAGOS        | 84   | -2.52           | 3.59  | 0.85 | 0.77  | 4.88   |
| legionowo_O3S        | VIE_IAGOS        | 458  | -2.65           | 5.13  | 0.69 | 0.65  | 8.46   |
| lauder_O3S           | 256_Umk          | 445  | -1.23           | 5.10  | 0.02 | 0.02  | 17.94  |
| hilo_O3S             | 031_Umk          | 43   | -9.94           | 3.93  | 0.46 | 0.32  | 4.15   |
| boulder_O3S          | 067_Umk          | 24   | -4.03           | 4.52  | 0.14 | 0.16  | 15.29  |
| ohp_O3S              | 040_Umk          | 456  | -5.09           | 6.65  | 0.41 | 0.52  | 8.73   |
| payerne_O3S          | 035_Umk          | 760  | -8.72           | 4.72  | 0.59 | 0.51  | 5.21   |
| sodankyla_O3S        | 105_Umk          | 224  | 0.49            | 6.68  | 0.30 | 0.47  | 16.98  |
| Mauna_Loa_FTIR       | HNL_IAGOS        | 55   | 9.18            | 3.46  | 0.76 | 1.04  | 8.59   |
| Tsukuba_FTIR         | KIX_IAGOS        | 17   | -3.61           | 3.51  | 0.23 | 0.17  | 22.08  |
| Tsukuba_FTIR         | NRT_IAGOS        | 17   | -3.70           | 4.47  | 0.48 | 0.28  | 19.52  |
| Toronto_FTIR         | DTW_IAGOS        | 54   | -4.58           | 6.44  | 0.46 | 0.32  | 18.67  |
| Toronto_FTIR         | YYZ_IAGOS        | 63   | -4.57           | 9.52  | 0.51 | 0.24  | 22.01  |
| Jungfrauoch_FTIR     | FRA_IAGOS        | 2330 | 10.90           | 4.34  | 0.59 | 0.80  | 14.55  |
| Zugspitze_FTIR       | MUC_IAGOS        | 363  | 6.68            | 4.64  | 0.56 | 0.53  | 16.39  |
| Lauder_FTIR          | 256_Umk          | 950  | -2.80           | 4.30  | 0.28 | 0.30  | 11.66  |
| Jungfrauoch_FTIR     | 035_Umk          | 1080 | 2.92            | 3.61  | 0.66 | 0.87  | 5.19   |

|              |           |      |       |      |      |      |       |
|--------------|-----------|------|-------|------|------|------|-------|
| Kiruna_FTIR  | 105_Umk   | 373  | -4.44 | 8.07 | 0.30 | 0.33 | 19.59 |
| 031_Umk      | HNL_IAGOS | 46   | 10.60 | 3.84 | 0.60 | 1.13 | 8.75  |
| 035_Umk      | FRA_IAGOS | 2209 | 7.57  | 5.04 | 0.51 | 0.54 | 16.57 |
| KIX_IAGOS    | NKM_IAGOS | 15   | -0.05 | 2.11 | 0.96 | 1.07 | -2.52 |
| KIX_IAGOS    | NRT_IAGOS | 44   | 0.12  | 5.99 | 0.59 | 0.45 | 15.69 |
| CDG_IAGOS    | BRU_IAGOS | 743  | 0.98  | 4.51 | 0.66 | 0.76 | 7.83  |
| DUS_IAGOS    | AMS_IAGOS | 22   | 1.21  | 3.63 | 0.68 | 0.53 | 14.68 |
| TMF_Lidar    | LAX_IAGOS | 33   | 8.57  | 3.70 | 0.81 | 1.33 | 1.60  |
| TMF_Lidar    | LAS_IAGOS | 17   | 4.76  | 3.24 | 0.76 | 0.82 | 8.71  |
| OHPO3T_Lidar | ohp_O3S   | 205  | 5.51  | 4.79 | 0.53 | 0.66 | 13.25 |

**Table S5.** Same as Table S3, but now for the tropospheric ozone column amounts from the surface to 300 hPa in Dobson unit (DU).

| site1                | site2             | N   | Bias<br>(2 - 1) | stdev | r     | slope | offset |
|----------------------|-------------------|-----|-----------------|-------|-------|-------|--------|
| mcmurdo_O3S          | Ahts_FTIR         | 26  | 1.27            | 1.20  | 0.57  | 0.55  | 9.55   |
| lauder_O3S           | Lauder_FTIR       | 210 | 1.20            | 1.86  | 0.76  | 0.94  | 2.43   |
| paramaribo_O3S       | Paramaribo_FTIR   | 16  | 1.51            | 3.78  | 0.45  | 0.46  | 11.79  |
| hilo_O3S             | Mauna_Loa_FTIR    | 200 | -7.44           | 2.86  | 0.77  | 0.50  | 3.88   |
| izana_O3S            | Izana_FTIR        | 257 | -6.50           | 2.41  | 0.84  | 0.82  | -1.21  |
| boulder_O3S          | Boulder_FTIR      | 120 | 1.57            | 2.72  | 0.66  | 0.80  | 6.12   |
| payerne_O3S          | Jungfraujoch_FTIR | 226 | -10.89          | 2.65  | 0.82  | 0.53  | 2.33   |
| hohenpeissenberg_O3S | Zugspitze_FTIR    | 305 | -4.43           | 3.48  | 0.54  | 0.59  | 5.74   |
| sodankyla_O3S        | Kiruna_FTIR       | 252 | 3.59            | 3.78  | 0.71  | 0.90  | 6.57   |
| nyalesund_O3S        | Ny_Alesund_FTIR   | 146 | 2.34            | 4.29  | 0.57  | 0.79  | 9.31   |
| eureka_O3S           | Eureka_FTIR       | 94  | 2.03            | 3.03  | 0.77  | 0.86  | 6.17   |
| irene_O3S            | JNB_IAGOS         | 19  | -4.48           | 3.74  | 0.66  | 0.52  | 6.92   |
| nairobi_O3S          | NBO_IAGOS         | 20  | -1.01           | 2.53  | 0.66  | 0.62  | 5.22   |
| kualalumpur_O3S      | SIN_IAGOS         | 22  | -1.47           | 4.59  | 0.67  | 1.11  | -3.36  |
| hilo_O3S             | HNL_IAGOS         | 23  | 2.29            | 3.38  | 0.53  | 0.60  | 10.82  |
| boulder_O3S          | DEN_IAGOS         | 17  | -0.47           | 2.28  | 0.71  | 0.88  | 2.42   |
| yarmouth_O3S         | BOS_IAGOS         | 53  | -3.31           | 3.36  | 0.65  | 0.63  | 9.39   |
| kelowna_O3S          | PDX_IAGOS         | 43  | -0.10           | 3.64  | 0.46  | 0.51  | 13.31  |
| kelowna_O3S          | YVR_IAGOS         | 56  | 0.34            | 2.77  | 0.63  | 0.68  | 8.65   |
| payerne_O3S          | FRA_IAGOS         | 214 | 0.11            | 2.09  | 0.88  | 0.85  | 4.27   |
| hohenpeissenberg_O3S | MUC_IAGOS         | 55  | 2.31            | 2.00  | 0.89  | 1.00  | 2.38   |
| uccle_O3S            | CDG_IAGOS         | 119 | -2.53           | 2.54  | 0.83  | 0.79  | 3.93   |
| uccle_O3S            | BRU_IAGOS         | 54  | -1.44           | 1.57  | 0.93  | 1.00  | -1.36  |
| debilt_O3S           | DUS_IAGOS         | 61  | -0.18           | 3.23  | 0.84  | 0.96  | 1.13   |
| debilt_O3S           | AMS_IAGOS         | 22  | -1.30           | 2.58  | 0.83  | 0.83  | 4.16   |
| edmonton_O3S         | YYC_IAGOS         | 16  | -2.33           | 2.59  | 0.58  | 0.51  | 10.36  |
| legionowo_O3S        | VIE_IAGOS         | 146 | -2.71           | 2.69  | 0.88  | 0.81  | 3.21   |
| lauder_O3S           | 256_Umk           | 342 | -0.88           | 3.68  | -0.12 | -0.12 | 20.62  |
| hilo_O3S             | 031_Umk           | 398 | -9.22           | 3.15  | 0.73  | 0.39  | 4.59   |
| boulder_O3S          | 067_Umk           | 400 | -4.65           | 2.64  | 0.64  | 0.48  | 7.44   |
| ohp_O3S              | 040_Umk           | 303 | -4.98           | 4.00  | 0.66  | 0.76  | 1.78   |
| payerne_O3S          | 035_Umk           | 235 | -8.27           | 2.52  | 0.82  | 0.63  | 2.30   |
| sodankyla_O3S        | 105_Umk           | 172 | -0.46           | 4.22  | 0.46  | 0.54  | 13.86  |
| Mauna_Loa_FTIR       | HNL_IAGOS         | 22  | 8.95            | 2.54  | 0.72  | 1.15  | 6.78   |
| Tsukuba_FTIR         | KIX_IAGOS         | 19  | -4.03           | 4.35  | 0.68  | 0.47  | 13.91  |
| Tsukuba_FTIR         | NGO_IAGOS         | 34  | -4.29           | 4.42  | 0.77  | 0.68  | 7.20   |

|                  |           |     |       |      |      |      |       |
|------------------|-----------|-----|-------|------|------|------|-------|
| Tsukuba_FTIR     | NRT_IAGOS | 18  | -5.88 | 3.88 | 0.85 | 0.52 | 11.01 |
| Toronto_FTIR     | DTW_IAGOS | 58  | -6.77 | 4.46 | 0.54 | 0.51 | 10.19 |
| Toronto_FTIR     | YYZ_IAGOS | 60  | -5.51 | 4.40 | 0.57 | 0.45 | 13.01 |
| Jungfrauoch_FTIR | FRA_IAGOS | 230 | 10.78 | 2.63 | 0.81 | 1.20 | 7.22  |
| Zugspitze_FTIR   | MUC_IAGOS | 50  | 5.53  | 3.74 | 0.59 | 0.65 | 12.79 |
| Lauder_FTIR      | 256_Umk   | 235 | -2.34 | 3.49 | 0.10 | 0.07 | 16.65 |
| Boulder_FTIR     | 067_Umk   | 121 | -5.72 | 2.66 | 0.67 | 0.49 | 6.68  |
| Jungfrauoch_FTIR | 035_Umk   | 250 | 2.37  | 2.34 | 0.72 | 0.82 | 5.56  |
| Kiruna_FTIR      | 105_Umk   | 171 | -3.71 | 4.75 | 0.53 | 0.46 | 14.58 |
| 031_Umk          | HNL_IAGOS | 21  | 9.95  | 2.84 | 0.73 | 1.58 | 2.02  |
| 067_Umk          | DEN_IAGOS | 15  | 3.86  | 3.55 | 0.15 | 0.19 | 18.79 |
| 035_Umk          | FRA_IAGOS | 303 | 7.92  | 3.16 | 0.70 | 0.97 | 8.62  |
| KIX_IAGOS        | NGO_IAGOS | 16  | -0.30 | 3.65 | 0.78 | 0.94 | 1.50  |
| KIX_IAGOS        | NKM_IAGOS | 36  | -0.38 | 4.15 | 0.78 | 1.01 | -0.67 |
| KIX_IAGOS        | NRT_IAGOS | 111 | 1.18  | 4.18 | 0.69 | 0.64 | 11.56 |
| NGO_IAGOS        | NRT_IAGOS | 20  | 0.27  | 5.60 | 0.38 | 0.26 | 21.36 |
| NKM_IAGOS        | NRT_IAGOS | 23  | 1.52  | 4.03 | 0.83 | 0.58 | 13.68 |
| DTW_IAGOS        | YYZ_IAGOS | 36  | -0.31 | 3.45 | 0.70 | 0.63 | 9.88  |
| PDX_IAGOS        | YVR_IAGOS | 18  | 0.80  | 4.58 | 0.12 | 0.18 | 21.42 |
| CDG_IAGOS        | BRU_IAGOS | 45  | 0.54  | 1.53 | 0.94 | 1.11 | -2.46 |
| TMF_Lidar        | SAN_IAGOS | 20  | 7.23  | 2.89 | 0.69 | 0.81 | 11.28 |
| TMF_Lidar        | LAX_IAGOS | 33  | 7.97  | 2.80 | 0.79 | 1.19 | 4.09  |
| OHPO3T_Lidar     | ohp_O3S   | 235 | 5.20  | 3.17 | 0.69 | 0.90 | 7.37  |
| OHPO3T_Lidar     | 040_Umk   | 206 | 0.02  | 4.19 | 0.49 | 0.71 | 6.61  |

**Table S6.** TrOC QR and MLR L3 trend calculations for the original and subsampled (2 randomly chosen daily measurements a month, denoted by “Sub” in the column headers) monthly mean time series. Trends are estimated in ppb per decade, and trend uncertainties (2 standard deviations) and  $p$  values (QR) are given. Bold trends have  $p < 0.05$ . Left columns display the station name, instrument type, and mean monthly sampling frequency of the original time series.

| Northern Hemisphere (180-20° W) TrOC (surface to 300hPa) trends   |                 |                               |                                            |                          |                                                |                                 |                                             |                                                    |
|-------------------------------------------------------------------|-----------------|-------------------------------|--------------------------------------------|--------------------------|------------------------------------------------|---------------------------------|---------------------------------------------|----------------------------------------------------|
| Station                                                           | Instru-<br>ment | Mon-<br>thly<br>Sam-<br>pling | QR L3<br>Trend $\pm 2\sigma$<br>(ppbv/dec) | QR<br>L3<br>$p$<br>value | QR L3 Sub<br>Trend $\pm 2\sigma$<br>(ppbv/dec) | QR<br>L3<br>Sub<br>$p$<br>value | MLR L3<br>Trend $\pm 2\sigma$<br>(ppbv/dec) | MLR L3<br>Sub Trend<br>$\pm 2\sigma$<br>(ppbv/dec) |
| Alert                                                             | O3S             | 4.04                          | $0.74 \pm 1.76$                            | 0.42                     | $-0.07 \pm 2.26$                               | 0.95                            | $0.62 \pm 1.63$                             | $0.42 \pm 1.89$                                    |
| ATL                                                               | IAGOS           | 5.51                          | $-0.78 \pm 2.22$                           | 0.50                     | N/A                                            | N/A                             | $-0.53 \pm 2.44$                            | N/A                                                |
| Boulder                                                           | O3S             | 4.51                          | <b><math>-1.41 \pm 1.14</math></b>         | <b>0.01</b>              | $-0.17 \pm 1.43$                               | 0.81                            | <b><math>-1.30 \pm 0.79</math></b>          | $-0.91 \pm 1.02$                                   |
| Boulder (067)                                                     | Umkehr          | 13.47                         | $0.44 \pm 1.30$                            | 0.52                     | $-0.82 \pm 1.43$                               | 0.26                            | $-0.02 \pm 1.08$                            | $-0.99 \pm 1.41$                                   |
| Churchill                                                         | O3S             | 3.63                          | $-1.64 \pm 2.42$                           | 0.17                     | $-2.85 \pm 3.02$                               | 0.08                            | <b><math>-3.01 \pm 1.98</math></b>          | $-3.02 \pm 2.27$                                   |
| DAL                                                               | IAGOS           | 3.08                          | <b><math>2.41 \pm 1.66</math></b>          | <b>0.01</b>              | N/A                                            | N/A                             | $2.16 \pm 2.63$                             | N/A                                                |
| Edmonton                                                          | O3S             | 3.93                          | $0.03 \pm 0.96$                            | 0.95                     | $-0.11 \pm 1.47$                               | 0.89                            | $-0.64 \pm 0.95$                            | $-0.80 \pm 1.11$                                   |
| Eureka                                                            | O3S             | 5.40                          | $0.32 \pm 1.36$                            | 0.65                     | $-0.03 \pm 1.89$                               | 0.98                            | $-0.30 \pm 1.37$                            | $-0.16 \pm 1.51$                                   |
| Fairbanks (105)                                                   | Umkehr          | 8.72                          | $0.02 \pm 2.28$                            | 0.99                     | $-0.15 \pm 3.70$                               | 0.94                            | $0.98 \pm 2.77$                             | $0.84 \pm 3.72$                                    |
| Goose Bay                                                         | O3S             | 4.09                          | $-0.80 \pm 1.28$                           | 0.21                     | $-0.80 \pm 1.49$                               | 0.28                            | $-0.26 \pm 1.20$                            | $-0.47 \pm 1.34$                                   |
| Hilo                                                              | O3S             | 4.13                          | $-0.43 \pm 1.30$                           | 0.50                     | $-0.86 \pm 1.71$                               | 0.32                            | $-0.41 \pm 1.03$                            | $-1.06 \pm 1.32$                                   |
| Mauna Loa                                                         | FTIR            | 14.37                         | $1.26 \pm 2.48$                            | 0.30                     | $0.99 \pm 2.54$                                | 0.44                            | $0.88 \pm 2.33$                             | $1.15 \pm 3.37$                                    |
| Mauna Loa (031)                                                   | Umkehr          | 19.65                         | <b><math>1.62 \pm 0.96</math></b>          | <b>0.00</b>              | <b><math>1.73 \pm 1.12</math></b>              | <b>0.00</b>                     | <b><math>1.49 \pm 0.91</math></b>           | <b><math>1.50 \pm 1.17</math></b>                  |
| Paramaribo                                                        | O3S             | 3.40                          | $-0.42 \pm 1.04$                           | 0.45                     | $-0.16 \pm 1.18$                               | 0.78                            | $0.22 \pm 1.17$                             | $-0.10 \pm 1.36$                                   |
| Resolute                                                          | O3S             | 3.71                          | <b><math>-2.07 \pm 1.78</math></b>         | <b>0.03</b>              | $-0.80 \pm 1.80$                               | 0.37                            | <b><math>-2.12 \pm 1.80</math></b>          | $-1.63 \pm 1.87$                                   |
| Scoresbysund                                                      | O3S             | 4.23                          | <b><math>-2.73 \pm 1.40</math></b>         | <b>0.00</b>              | <b><math>-2.51 \pm 1.72</math></b>             | <b>0.00</b>                     | <b><math>-2.82 \pm 1.15</math></b>          | <b><math>-3.23 \pm 1.38</math></b>                 |
| TMF                                                               | Lidar           | 10.57                         | <b><math>1.24 \pm 1.08</math></b>          | <b>0.02</b>              | <b><math>1.28 \pm 1.08</math></b>              | <b>0.02</b>                     | <b><math>1.31 \pm 1.02</math></b>           | $1.08 \pm 1.36$                                    |
| Thule                                                             | FTIR            | 9.69                          | <b><math>-3.27 \pm 1.74</math></b>         | <b>0.00</b>              | $-1.77 \pm 1.89$                               | 0.06                            | <b><math>-3.59 \pm 1.92</math></b>          | $-1.41 \pm 2.75$                                   |
| Toronto                                                           | FTIR            | 8.98                          | $-1.15 \pm 2.16$                           | 0.27                     | $-0.72 \pm 3.50$                               | 0.68                            | $-1.70 \pm 2.08$                            | $-1.02 \pm 3.02$                                   |
| Trinidad Head                                                     | O3S             | 4.50                          | $-0.96 \pm 1.12$                           | 0.07                     | <b><math>-1.41 \pm 1.27</math></b>             | <b>0.03</b>                     | <b><math>-0.90 \pm 0.89</math></b>          | $-1.08 \pm 1.14$                                   |
| Wallops Island                                                    | O3S             | 4.67                          | <b><math>-2.83 \pm 1.50</math></b>         | <b>0.00</b>              | <b><math>-2.82 \pm 1.74</math></b>             | <b>0.00</b>                     | <b><math>-2.81 \pm 1.25</math></b>          | <b><math>-2.45 \pm 1.42</math></b>                 |
| Northern Hemisphere (19° W-79° E) TrOC (surface to 300hPa) trends |                 |                               |                                            |                          |                                                |                                 |                                             |                                                    |
| Arosa (035)                                                       | Umkehr          | 8.09                          | $0.63 \pm 1.36$                            | 0.34                     | $0.03 \pm 2.00$                                | 0.98                            | $0.68 \pm 1.05$                             | $-0.38 \pm 1.42$                                   |
| Ascension Island                                                  | O3S             | 3.83                          | $-1.06 \pm 1.76$                           | 0.22                     | $-0.70 \pm 1.54$                               | 0.37                            | $-0.88 \pm 1.74$                            | $-0.93 \pm 1.84$                                   |
| De Bilt                                                           | O3S             | 4.31                          | <b><math>1.50 \pm 1.20</math></b>          | <b>0.01</b>              | <b><math>2.08 \pm 1.38</math></b>              | <b>0.00</b>                     | <b><math>1.34 \pm 1.08</math></b>           | <b><math>1.34 \pm 1.34</math></b>                  |
| FRA                                                               | IAGOS           | 24.64                         | $0.09 \pm 1.10$                            | 0.87                     | $1.13 \pm 1.19$                                | 0.06                            | $-0.04 \pm 1.08$                            | $0.28 \pm 1.36$                                    |
| Hohenpeissenberg                                                  | O3S             | 10.58                         | $0.55 \pm 0.94$                            | 0.23                     | $0.35 \pm 1.24$                                | 0.63                            | $0.26 \pm 0.76$                             | $0.35 \pm 0.97$                                    |
| Izana                                                             | FTIR            | 8.55                          | $1.08 \pm 1.30$                            | 0.08                     | $1.05 \pm 2.26$                                | 0.37                            | $0.73 \pm 1.07$                             | $0.85 \pm 1.53$                                    |
| Izana                                                             | O3S             | 4.00                          | <b><math>2.12 \pm 1.18</math></b>          | <b>0.00</b>              | <b><math>1.92 \pm 1.52</math></b>              | <b>0.01</b>                     | <b><math>2.30 \pm 0.87</math></b>           | <b><math>1.79 \pm 1.06</math></b>                  |
| Jungfraujoch                                                      | FTIR            | 8.56                          | <b><math>-1.93 \pm 1.78</math></b>         | <b>0.03</b>              | <b><math>-1.83 \pm 1.64</math></b>             | <b>0.03</b>                     | $-1.08 \pm 1.34$                            | $-1.33 \pm 1.86$                                   |



|                                                                        |        |       |                     |             |                     |             |                     |                     |
|------------------------------------------------------------------------|--------|-------|---------------------|-------------|---------------------|-------------|---------------------|---------------------|
| Kiruna                                                                 | FTIR   | 7.26  | <b>-1.77 ± 1.48</b> | <b>0.02</b> | -1.26 ± 1.33        | 0.06        | <b>-1.73 ± 1.15</b> | <b>-1.77 ± 1.42</b> |
| Legionowo                                                              | O3S    | 4.86  | <b>-1.26 ± 1.18</b> | <b>0.04</b> | <b>-2.02 ± 1.32</b> | <b>0.00</b> | <b>-1.40 ± 1.06</b> | <b>-1.86 ± 1.05</b> |
| Lerwick                                                                | O3S    | 4.93  | -1.01 ± 1.54        | 0.18        | -0.77 ± 1.57        | 0.33        | -0.96 ± 1.24        | -1.02 ± 1.46        |
| Madrid                                                                 | O3S    | 3.96  | -0.74 ± 1.24        | 0.25        | -0.61 ± 1.29        | 0.38        | -0.62 ± 1.22        | -0.54 ± 1.39        |
| NyAlesund                                                              | O3S    | 6.50  | -0.75 ± 1.08        | 0.15        | <b>-1.21 ± 1.14</b> | <b>0.04</b> | <b>-0.93 ± 0.91</b> | -0.98 ± 1.18        |
| OHP (040)                                                              | Umkehr | 11.29 | 0.51 ± 2.10         | 0.62        | -0.19 ± 2.40        | 0.88        | -0.86 ± 1.88        | -0.76 ± 1.92        |
| OHP                                                                    | Lidar  | 6.72  | <b>2.24 ± 1.76</b>  | <b>0.01</b> | <b>3.00 ± 2.13</b>  | <b>0.00</b> | 1.90 ± 2.04         | <b>2.48 ± 1.97</b>  |
| OHP                                                                    | O3S    | 3.85  | <b>1.37 ± 1.26</b>  | <b>0.03</b> | 1.04 ± 1.39         | 0.12        | <b>1.96 ± 1.05</b>  | <b>1.67 ± 1.10</b>  |
| Payerne                                                                | O3S    | 12.75 | <b>-1.29 ± 1.02</b> | <b>0.01</b> | <b>-1.98 ± 1.57</b> | <b>0.01</b> | <b>-1.63 ± 0.94</b> | <b>-2.53 ± 1.22</b> |
| Sodankyla                                                              | O3S    | 4.17  | <b>-1.74 ± 1.40</b> | <b>0.01</b> | <b>-1.89 ± 1.42</b> | <b>0.01</b> | <b>-1.75 ± 1.08</b> | <b>-2.02 ± 1.20</b> |
| Uccle                                                                  | O3S    | 11.80 | <b>1.23 ± 1.10</b>  | <b>0.03</b> | 0.83 ± 1.51         | 0.27        | 0.57 ± 0.97         | 0.51 ± 1.31         |
| Valentia                                                               | O3S    | 4.06  | 1.37 ± 2.04         | 0.18        | 1.63 ± 2.68         | 0.23        | -0.36 ± 2.41        | -0.02 ± 3.20        |
| Zugspitze                                                              | FTIR   | 9.68  | -1.15 ± 1.82        | 0.22        | -1.75 ± 2.51        | 0.16        | -0.32 ± 0.60        | -1.69 ± 2.40        |
| <b>Northern Hemisphere (80-180° E) TrOC (surface to 300hPa) trends</b> |        |       |                     |             |                     |             |                     |                     |
| Kuala Lumpur                                                           | O3S    | 2.07  | <b>2.61 ± 1.74</b>  | <b>0.00</b> | <b>2.26 ± 1.91</b>  | <b>0.02</b> | <b>1.86 ± 1.56</b>  | <b>1.92 ± 1.55</b>  |
| Rikubetsu                                                              | FTIR   | 3.73  | -0.12 ± 1.24        | 0.85        | -0.29 ± 2.03        | 0.78        | -0.58 ± 1.37        | -1.37 ± 1.72        |
| <b>Southern Hemisphere TrOC (surface to 300hPa) trends</b>             |        |       |                     |             |                     |             |                     |                     |
| Arrival Heights                                                        | FTIR   | 6.24  | <b>-1.25 ± 1.20</b> | <b>0.04</b> |                     | <b>0.00</b> | <b>-1.69 ± 1.32</b> | <b>-2.20 ± 1.48</b> |
| Fiji                                                                   | O3S    | 2.58  | -1.04 ± 1.80        | 0.29        | 0.0 ± 1.70          | 1.00        | -1.33 ± 2.28        | -1.01 ± 2.27        |
| Irene                                                                  | O3S    | 2.48  | 0.48 ± 2.36         | 0.68        | 0.49 ± 2.11         | 0.66        | -0.16 ± 2.41        | 0.31 ± 2.63         |
| Lauder                                                                 | O3S    | 3.84  | 0.01 ± 0.70         | 0.98        | 0.24 ± 0.75         | 0.52        | 0.13 ± 0.61         | -0.16 ± 0.79        |
| Lauder                                                                 | FTIR   | 10.90 | <b>1.64 ± 0.86</b>  | <b>0.00</b> | <b>1.85 ± 1.10</b>  | <b>0.00</b> | <b>1.67 ± 0.86</b>  | <b>1.21 ± 1.08</b>  |
| Lauder (256)                                                           | Umkehr | 8.97  | 0.38 ± 1.20         | 0.55        | 0.69 ± 1.61         | 0.41        | 0.58 ± 0.86         | 0.35 ± 0.92         |
| Nairobi                                                                | O3S    | 3.90  | 0.47 ± 1.56         | 0.54        | 0.69 ± 1.62         | 0.40        | 0.75 ± 1.37         | 0.60 ± 1.41         |
| Natal                                                                  | O3S    | 3.67  | 0.76 ± 1.22         | 0.21        | 0.90 ± 1.83         | 0.33        | 1.04 ± 1.37         | 1.49 ± 1.73         |
| Reunion                                                                | O3S    | 3.27  | 1.17 ± 1.62         | 0.15        | 0.51 ± 1.65         | 0.54        | <b>1.93 ± 1.27</b>  | <b>1.45 ± 1.40</b>  |
| Samoa                                                                  | O3S    | 3.31  | -0.49 ± 1.10        | 0.35        | -0.97 ± 1.60        | 0.23        | -0.52 ± 0.99        | -0.52 ± 1.03        |
| South Pole                                                             | O3S    | 4.89  | <b>-0.90 ± 0.56</b> | <b>0.00</b> | -0.55 ± 0.76        | 0.15        | <b>-1.01 ± 0.73</b> | -0.36 ± 0.77        |

In Table S6, we compare the original L3 trends values with the L3 trends from a single realization of the datasets subsampled to exactly two random daily values a month. It should be mentioned that our single “subsampled” dataset contains fewer sites, as the IAGOS airports DAL and ATL have too many months with only one measurement a month, so that any time series sampled to exactly two daily values a month contain too much gaps for a reliable trend estimation. From Table S6, we can conclude that, for this specific subsampling strategy, the differences between the trend values estimated from both samples are not large (mean absolute trend difference of  $0.46 \pm 0.37$  and  $0.37 \pm 0.38$  ppb per decade for QR and MLR, respectively). For both methods, only at around five sites, there is a trend sign reversal, but the estimated trends have large uncertainties. The most striking and consistent feature of the comparison is the higher trend uncertainties ( $2\sigma$ ) for the subsampled dataset (i.e. for

46/49 sites, or 87/92%, for QR/MLR). Also the QR  $p$  values of the “subsampled” trend estimations are higher for the majority of the sites (32 sites, 60%). As a consequence, the number of sites with trends significantly different from zero (taking a  $p$  value lower than 0.05 as criterion) decreases, although moderately, from 22/21 (40/38%) to 16/15 (30/28%) for QR/MLR when subsampling the data to exactly two monthly measurements for calculating the monthly mean. This is not unexpected because, based on the sampling theory (Thompson, 2012), if the samples are chosen randomly and have no structural bias, the results are not expected to be biased, but smaller samples lead to larger uncertainty. As a matter of fact, the differences in trend values and trend uncertainties between the two L3 datasets are comparable with those between QR L1 and QR L3: a similar amount of sites with larger than smaller trend estimates, a mean absolute trend differences of  $0.46 \pm 0.40$  ppb per decade, 7 (out of 55 sites) switching trend sign, all but one sites having larger trend uncertainties (2 sigma values) for QR L3 compared to QR L1, 40 sites having larger QR L3 trend  $p$  values, the number of sites with trends significantly different from zero (taking a  $p$  value lower than 0.05 as criterion) being 32 for QR L1 (and 22 for QR L3). On top of that, the trend (uncertainty) differences between the original and subsampled datasets lie also in the same order of magnitude than those between the two trends estimation methods (QR and MLR) used, with a mean absolute trend difference of  $0.38 \pm 0.37$  ppb per decade, and exactly the same number of sites having trends significantly different from zero.

We can therefore conclude that the trend uncertainty due to a monthly sampling frequency of around 2 is comparable to the trend uncertainty that is associated with the choice of the trend estimation method and with the one due to the sampling frequency (all measurements vs. monthly means) for the QR trend estimation.

**Table S7.** Summary of trend differences, drifts, and their causes between the time series at collocated sites. The sampling is described by percentage of months with data and by the average number of observations for each month with data.

| station        | technique | sampling            | QR trend [ppbv/dec] | drift [ppbv/dec]                                | site comments                                                | general comment                                                                                                                                                                                    |
|----------------|-----------|---------------------|---------------------|-------------------------------------------------|--------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| <b>Izaña</b>   | O3S       | 97.8%<br>4.0/month  | <b>+2.59±0.68</b>   |                                                 | launch at coast (36 m a.s.l.)                                | despite drift between series, no significant trend difference                                                                                                                                      |
|                | FTIR      | 96.3%<br>8.6/month  | <b>+1.88±0.88</b>   | -1.53±0.88                                      | FTIR located at mountain top (2373m)                         |                                                                                                                                                                                                    |
| <b>Boulder</b> | O3S       | 99.7% 4.5           | <b>-1.14±0.86</b>   |                                                 | DLM trends always negative over the years                    | interannual variability not always similar, no significant DLM trend differences over the years                                                                                                    |
|                | Umkehr    | 98.6%<br>13.5/month | +0.11±0.72          | +1.24±0.84                                      | DLM trends close to zero over the years                      |                                                                                                                                                                                                    |
| <b>Lauder</b>  | O3S       | 91.9%<br>3.8/month  | +0.13±0.50          |                                                 | ends in mid 2022, while other two continue until 2023        | Umkehr different seasonal cycle than the other two techniques? Very low correlation coefficients between Umkehr and O3S/FTIR time series. Less positive trends compared to Bjorklund et al. (2024) |
|                | FTIR      | 98.1%<br>10.9/month | <b>+1.54±0.44</b>   | +1.97±0.80 (vs. O3S)<br>+0.97±0.81 (vs. Umkehr) | positive drift of FTIR vs. other two                         |                                                                                                                                                                                                    |
|                | Umkehr    | 95.3%<br>9.0/month  | +0.36±0.70          |                                                 | Umkehr turns from negative to positive trends in 2014 (DLM)  |                                                                                                                                                                                                    |
| <b>OHP</b>     | O3S       | 97.9%<br>3.9/month  | <b>+1.95±1.08</b>   |                                                 |                                                              | especially large differences in beginning of time series                                                                                                                                           |
|                | Umkehr    | 88.5%<br>11.3/month | <b>-1.49±1.00</b>   | -2.74±1.44 (vs. O3S)                            | DLM negative trends over the entire period                   |                                                                                                                                                                                                    |
|                | Lidar     | 85.9%<br>6.7/month  | <b>+1.93±1.02</b>   |                                                 | significantly positive trend by DLM during the entire period |                                                                                                                                                                                                    |

|        |        |                     |                   |                         |                                                                                                                                                     |                                                                                                                                                                                       |
|--------|--------|---------------------|-------------------|-------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Hawaii | O3S    | 100%<br>4.1/month   | -0.28±0.98        |                         | launch at coast<br>(11 m a.s.l.),<br>drop in TCO<br>since 2014                                                                                      | due to TCO<br>drop in<br>ozonesonde<br>time series,<br>positive drifts<br>of other<br>techniques vs.<br>O3S. Umkehr<br>and FTIR<br>trends, located<br>at same spot,<br>are consistent |
|        | Umkehr | 96.7%<br>19.7/month | <b>+1.83±0.44</b> | +1.97±0.88<br>(vs. O3S) | located at<br>mountain top<br>(3397 m),<br>significant<br>positive trends<br>since 2004<br>(DLM).                                                   |                                                                                                                                                                                       |
|        | FTIR   | 60%<br>14.4/month   | <b>+2.03±1.30</b> | +1.71±1.45<br>(vs. O3S) | located at<br>mountain top<br>(3397 m),<br>sparse data<br>coverage in<br>beginning of<br>time period,<br>constant trend<br>of 0.2 ppbv/yr<br>by DLM |                                                                                                                                                                                       |

**Table S8.** Same as Table S7, but now for different techniques at nearby sites.

| station            | technique | sampling            | Trend<br>[ppb/dec] | drift<br>[ppb/dec]                                  | site comments                                                                                                                                                                                                                                                  | general<br>comment                                                                                                                                         |
|--------------------|-----------|---------------------|--------------------|-----------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------|
| <b>Sodankylä</b>   | O3S       | 92.0%<br>4.2/month  | <b>-1.28±0.78</b>  |                                                     |                                                                                                                                                                                                                                                                | in<br>agreement                                                                                                                                            |
| <b>Kiruna</b>      | FTIR      | 85.2%<br>7.3/month  | <b>-2.26±0.88</b>  |                                                     |                                                                                                                                                                                                                                                                |                                                                                                                                                            |
| <b>Uccle</b>       | O3S       | 100%<br>11.8/month  | <b>+0.90±0.48</b>  |                                                     |                                                                                                                                                                                                                                                                | overall very<br>good<br>agreement<br>between the<br>3 time<br>series, De<br>Bilt most<br>deviant from<br>other 2 due<br>to lower<br>sampling<br>frequency? |
| <b>De Bilt</b>     | O3S       | 100%<br>4.3/month   | <b>+1.34±0.86</b>  |                                                     | higher variability<br>in monthly<br>anomalies due to<br>lower launch rate                                                                                                                                                                                      |                                                                                                                                                            |
| <b>Frankfurt</b>   | IAGOS     | 90.8%<br>24.6/month | <b>+0.65±0.36</b>  |                                                     | outliers associated<br>with large<br>statistical<br>uncertainty or<br>single monthly<br>value (2005)                                                                                                                                                           |                                                                                                                                                            |
| <b>Payerne</b>     | O3S       | 100%<br>12.8/month  | <b>-1.30±0.62</b>  |                                                     | altitude: 491m,<br>homogenized time<br>series starts in<br>2002, other 2 start<br>in 2000,<br>“smoother”<br>anomaly time<br>series (higher<br>monthly<br>sampling?), DLM<br>negative trends<br>during entire<br>period (but only<br>significant until<br>2009) | especially<br>large<br>differences<br>in beginning<br>of time<br>series                                                                                    |
| <b>Arosa</b>       | Umkehr    | 97.1%<br>8.1/month  | +0.56±0.78         | +1.93±0.96<br>(vs. O3S)<br>+1.88±1.18<br>(vs. FTIR) | altitude: 1840 m                                                                                                                                                                                                                                               |                                                                                                                                                            |
| <b>Jungfrauoch</b> | FTIR      | 93.9%<br>8.6/month  | <b>-1.78±0.66</b>  |                                                     | altitude: 3580 m,<br>free troposphere                                                                                                                                                                                                                          |                                                                                                                                                            |

|                          |      |                    |                   |  |                                                                                 |                                                                                                                                     |
|--------------------------|------|--------------------|-------------------|--|---------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------|
| <b>Hohen-peissenberg</b> | O3S  | 100%<br>10.6/month | <b>+0.50±0.46</b> |  | altitude: 975 m<br>Brewer-Mast type                                             | although different trend sign, no significant drift between both time series. Is altitude differences driving the trend difference? |
| <b>Zugspitze</b>         | FTIR | 95.7%<br>9.7/month | <b>-1.82±0.88</b> |  | altitude: 2960 m,<br>low values at beginning of time period, higher variability |                                                                                                                                     |