



# Investigating the differences in calculating global mean surface CO<sub>2</sub> abundance: the impact of analysis methodologies and site selection

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16 Abstract. The World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) coordinates high-quality 17 atmospheric greenhouse gas observations globally and provides these observations through the WMO World Data Centre for Greenhouse Gases (WDCGG) supported by Japan Meteorological Agency. The WDCGG and the National Oceanic and 18 19 Atmospheric Administration (NOAA) analyse these measurements using different methodologies and site selection to 20 calculate global annual mean surface CO2 and its growth rate as a headline climate indicator. This study proposes a third hybrid method named semi-NOAA, which is used as an independent validation of the methods as described by NOAA and 21 22 WDCGG. We apply the semi-NOAA to incorporate observations from most WMO GAW stations and 3D modelled CO2 23 fields from CarbonTracker Europe (CTE). We found that different observational networks (i.e., the NOAA, GAW, and CTE 24 networks) and analysis methods result in differences in the calculated global surface CO2 mole fractions equivalent to the 25 current atmospheric growth rate over a three-month period. However, the CO2 growth rate derived from these networks and 26 CTE model output shows good agreement. Over the long-term period (40 years), both networks with and without continental 27 sites exhibit the same trend in the growth rate  $(0.030 \pm 0.002 \text{ ppm per year})$ . However, a clear difference emerges in the short-28 term (one month) change of the growth rate. The network that includes continental sites improves the early detection of 29 changes in biogenic emissions.

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#### 1 Introduction

38 Global mean surface temperature averaged over 2011-2020 has increased by about 1.09°C relative to the average temperature 39 of 1850-1900 (Gulev et al., 2021). The increasing amount of atmospheric carbon dioxide (CO<sub>2</sub>), together with increases in 40 other greenhouse gases, is the main driver of the warming (Eyring et al., 2021). After being relatively stable between 180 ppm 41 (ice age) and 280 ppm (interglacial) for the last 800,000 years (Lüthi et al., 2008), the annual average CO2 level of the 42 atmosphere has increased since the industrial revolution from roughly 277 ppm in 1750 to 415.7±0.2 ppm in 2021 (WMO, 43 2022), due to emissions of CO<sub>2</sub> related to human activities like burning of fossil fuels and land use changes (Friedlingstein et 44 al., 2022). Mean global atmospheric CO2 annual growth rate (GATM) is an important constraint on the global carbon cycle. Based on the most recent Global Carbon Budget (GCB) analysis (Friedlingstein et al., 2022), the total emission of CO<sub>2</sub> due to 45 human activities was  $10.2 \pm 0.8$  GtC yr<sup>1</sup> in 2020, of which  $3.0 \pm 0.4$  GtC yr<sup>1</sup> was captured by the ocean sink and  $2.9 \pm 1$  GtC 46 47 yr¹ by the terrestrial sink, leaving a net increase of 5.0 ± 0.2 GtC yr¹ of CO₂ in the atmosphere, corresponding to an 48 atmospheric CO<sub>2</sub> mole fraction increase of  $2.4 \pm 0.1$  ppm yr<sup>-1</sup>. 49 As the atmosphere mixes the contributions of all sources and sinks, an observational global average CO2 mole fraction can be 50 constructed if there are enough observations to represent the spatial and temporal variation across the globe. Since most land 51 masses are concentrated in the Northern Hemisphere, and the highest anthropogenic emissions (e.g. during winter) occur in 52 the relatively narrow latitudinal band between 30 °N and 60 °N, relatively large spatial and temporal gradients in CO<sub>2</sub> mole 53 fraction exist in and around that region. Due to convective and advective mixing, the average mixing time of air within the 54 same latitudinal bands varies from several weeks to a month. However, mixing between latitudinal bands is slower, especially 55 the exchange between the northern and southern hemispheres, which has an approximate interhemispheric transport time of 56 1.4 ± 0.2 years (Patra et al., 2011). The interplay of the latitudinal and interhemispheric differences in fossil fuel emissions 57 and seasonal exchange with land biota (Denning et al., 1995) creates a latitudinal and interhemispheric gradient that requires 58 a sufficiently dense network to capture a representative global annual mean. 59 However, measurement stations that are close to sources or sinks may not be representative of a large atmosphere volume and 60 the average signal at their latitude. Therefore, inclusion of these observations might introduce significant biases on the global 61 mean CO2 and its growth rate. These biases can be avoided by filtering of data and a careful selection of spatially representative 62 stations, as done by NOAA in their use of 43 stations (Fig. 1) that are considered to be representative for the Marine Boundary 63 Layer (MBL reference network, https://www.esrl.noaa.gov/gmd/ccgg/mbl/mbl.html). An additional data processing step 64 developed by NOAA to further avoid biases due to unrepresentative local signals is filtering and smoothing, by using a 65 combination of a low pass filter and decomposition into a fitted long-term trend and seasonal cycle (Thoning et al., 1989), 66 hereafter refered to as the NOAA analysis. These fits can also be used to fill gaps for missing data, though care must be taken 67 to avoid extrapolation errors before and beyond the time covered by the data record of the station. The WMO Global 68 Atmosphere Watch (GAW) World Data Centre for Greenhouse Gases (WDCGG) also publishes global averages mole fraction 69 for CO<sub>2</sub> and the other major greenhouse gases. They use curve fitting and filter methods that are very similar to those developed 70 by NOAA, but WDCGG includes continental locations that are potentially influenced by local sources and sinks (Tsutsumi et 71 al., 2009). 72 The NOAA MBL observations are all part of the NOAA cooperative global air sampling network and analysed in the same

laboratory. All NOAA flask-air observations are traceable to the WMO X2019 CO2 scale that is maintained by NOAA Global

Monitoring Laboratory (GML). In contrast, the WDCGG data originate from multiple independent laboratories (including

NOAA GML), that together form a network of hundreds of stations coordinated by WMO GAW. Having a multitude of

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76 independent laboratories carries an additional risk of biases due to differences in sampling, measurement, and analysis 77 methods, for example calibration scales, although much care is taken to avoid these by coordination in the network and use of 78 a common calibration scale from the WMO Central Calibration Laboratory (CCL) guided by a set of strict measurement 79 compatibility goals (WMO, 2022). The different selection of stations results in a larger seasonal cycle amplitude in WDCGG results compared to those of NOAA and a small but quite consistent bias in global surface annual mean CO2 mole fraction 80 81 (Tsutsumi et al., 2009). The NOAA estimate of global surface annual mean CO2 mole fraction is expected to be negatively biased (e.g. ~0.35 ppm lower than the WDCGG estimate, Tsutsumi et al., 2009) compared to a full global surface average 82 83 because areas with large sources are not represented. However, none of the two afore mentioned approaches represents those parts that have the atmosphere with low CO<sub>2</sub> mole fraction levels, i.e. the full troposphere (up to ~8-15 km altitude) and the 84 85 stratosphere or the regions of the world with substantial observational gaps. 86 In this paper we propose a data integration method to estimate the global mean surface CO2 and its growth rate, named semi-87 NOAA, which is used as an independent validation of the methods as described by NOAA and WDCGG through a completely 88 independent and open-source implementation. We apply the semi-NOAA methodology to incorporate CO2 data from the 89 GAW network (139 stations, Fig. 1) and a well-established 3D global transport model (TM5: Transport Model 5, Peters et al., 90 2004, Krol et al., 2005). We investigate the influence of small differences between the three methodologies and whether these 91 are significant or not for calculating the global mean surface CO2 and its growth rate, how consistent the semi-NOAA and 92 WDCGG approaches are with each other, and how they compare with NOAA analysis and estimates derived from a CO2 93 simulation with the 3D transport model TM5. These 3D CO2 results for 2001-2020 using TM5 are performed in the 94 CarbonTracker Europe framework (CTE, Peters et al., 2004, Van Der Laan-Luijkx et al., 2017), where the CO<sub>2</sub> uptake and 95 emission fluxes are optimized by the inversion system to minimize the mismatch between the in situ observations and the modelled CO2 mole fraction. CTE generally has a good representation of the CO2 field, with mean biases with respect to 96 97 independent aircraft measurements of generally less than 0.5 ppm (Friedlingstein et al., 2022). Furthermore, the inferred CO<sub>2</sub> 98 fluxes from CTE fit well within the ensemble of those of other inversions used for the evaluation of Global Carbon Budget 99 (e.g. Friedlingstein et al., 2022).



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#### 2 Methods and data

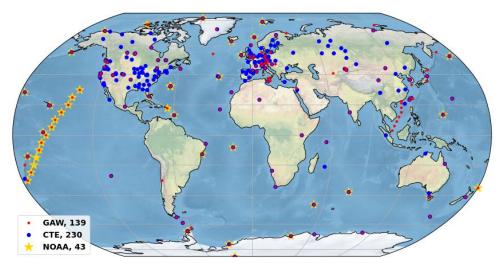


Figure 1. The selected GAW global network for  $CO_2$  measurement (139 sites, red dots), the global network for the CTE evaluations (230 sites, blue dots), and the NOAA network (43 sites, yellow stars).

#### 2.1 The WMO GAW observations and WDCGG analysis method

The WMO GAW network measurements are archived and distributed by WDCGG (World Data Center for Greenhouse Gases), hosted by the Japan Meteorological Agency. The GAW observations used in this study originate from 139 selected stations of the GAW network, and all observations are on the WMO standard scale, WMO-CO<sub>2</sub>-X2019 (Hall et al., 2021). The details on the station selection are described in Tsutsumi et al., (2009), which mainly excludes stations located in the northern hemisphere that show large standard deviations from the latitudinal fitted curve. The remaining 139 stations show a more reasonable latitudinal scatter range (Fig. 1).

The WDCGG global analysis method (hereafter WDCGG method), as described in Tsutsumi et al., (2009), includes the mentioned station selection, a data fitting and filter (involves data interpolation and extrapolation), and calculation of the zonal and global mean mole fractions, trends, and growth rates. The procedure is also summarized in Text S1.

114 The output from the global analysis by the WDCGG method are used to compare against an alternative method (semi-NOAA)

115 that we designed to follow as closely as possible the fit and filter method (Conway et al., 1994) deployed by NOAA and is

116 described in the section 2.3.

#### 2.2 CTE model output and station observations

CarbonTracker Europe (CTE) is a global model of atmospheric CO<sub>2</sub> and designed to keep track of CO<sub>2</sub> uptake and release at the Earth's surface over time (Van Der Laan-Luijkx et al., 2017). CTE incorporates an off-line atmospheric transport module (TM5, Peters et al., 2004, Krol et al., 2005) driven by ECMWF ERA5 data, and there are four prescribed fluxes (i.e. from ocean, biosphere, fire and fossil fuel), which are transported in the model, together with the transported initial CO<sub>2</sub> field. CTE also includes a data assimilation system that applies an ensemble Kalman filter to optimize the biogenic and ocean fluxes for a combination of plant-functional types and climate zones to improve the fit of the simulated concentrations with observations.





- 124 The optimized fluxes from the data assimilation have been used in Global Carbon Project (GCP) 2021 (Friedlingstein et al.,
- 125 2022), and CTE compares well to the other data assimilation systems used in GCP.
- 126 The CTE model data used here consisted of simulated monthly CO<sub>2</sub> mole fraction at 1x1 degree horizontal resolution and 25
- levels in the vertical, the data period ranges from 2001 to 2020 which has no influence of model spin-up (Krol et al., 2018).
- 128 From the CTE output a set of simulated synthetic atmospheric CO<sub>2</sub> mole fractions with monthly resolution can be extracted
- 129 within grid cells where stations are situated. This study analyses monthly observation data (1980-2020) and synthetic time
- 130 series (2001-2020) by using the semi-NOAA method (section 2.3) and attempts to estimate global mean CO2 mole fraction
- and its growth rate. The observed CO<sub>2</sub> mole fractions are taken from 230 out of 290 global-wide distributed stations (Fig. 1,
- 132 the station selection is summarized in Text S2), the data come from the GLOBALVIEW-plus ObsPack data product (Kenneth
- N., 2022), and include surface-based, shipboard-based and tower-based measurements.

#### 2.3 The semi-NOAA method

- 135 The temporal pattern of CO<sub>2</sub> measurement records at locations around the globe can be explained as the combination of
- roughly three components: a long-term trend, a non-sinusoidal yearly cycle (or seasonality), and short-term variations. This
- 137 study synchronizes monthly CO<sub>2</sub> records with the fitting and filter method obtained from the NOAA Global Monitoring
- 138 Laboratory (Thoning et al., 1989, Conway et al., 1994), without extrapolation. The station selection and CO<sub>2</sub> averaging method
- 139 are kept the same as in the WDCGG method (Text S1). This method will be referred to as the semi-NOAA method and will
- be compared to the WDCGG method without extrapolation. The only difference from WDCGG method without extrapolation
- 141 is the fitting and filter method. All code for the method described here was developed in Python and is available as a Jupyter
- notebook under a GPL license [https://doi.org/10.18160/Q788-9081]. The semi-NOAA method can be summarized by the
- 143 following three steps.

#### 144 2.3.1. Fitting and filter

- 145 CO<sub>2</sub> records from each station can be abstracted as a combination of long-term trend and seasonality, which can be fitted by
- 146 a function consisting of polynomial and harmonics. We applied a linear regression analysis based on 3 polynomial coefficients
- and 4 harmonics (Eq. 1) to fit CO<sub>2</sub> data using general linear least-squares fit (LFIT, Press et al., 1988).

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$$f(x) = a_0 + a_1 t + a_2 t^2 + \dots + a_k t^k + \sum_{n=1}^{n_h} (A_n \cos 2\pi n t + B_n \sin 2\pi n t)$$
 (1)

- where  $a_k$ ,  $A_n$  and  $B_n$  are fitted parameters, t is the time from the beginning of the observation and it is in months and
- 150 expressed as a decimal of its year. k denotes polynomial number, k = 2.  $n_h$  denotes harmonic number,  $n_h = 4$ . Fig. 2
- 151 illustrates the function fit to CO<sub>2</sub> data to gain the annual oscillation (red line in Fig. 2a), is a combination of a polynomial fit
- to the trend (blue line in Fig. 2a) and harmonic fit to the seasonality (green line in Fig. 2b).
- 153 The residuals are the difference between raw data and the function fit (black dots in Fig. 2c). The filtering method is based on
- 154 Thoning et al. (1989) which transforms CO<sub>2</sub> data from time domain to frequency domain using a Fast Fourier Transform
- 155 (FFT), then applies of a low pass filter to the frequency data to remove high-frequency variations, and then transform the
- 156 filtered data back to the time domain using an inverse FFT. The short term (a cut-off value of 80 days, red line in Fig. 2c) and
- 157 long term (a cut-off value of 667 days, blue line in Fig. 2c) filters used here are the same as in NOAA method, and applied to
- 158 obtain the short term and interannual variations that are not determined by the fit function. The original part of the code is also
- available as Python code from the NOAA website [https://gml.noaa.gov/aftp/user/thoning/ccgcrv/].





### 2.3.2. Calculate smoothed CO2 and long-term trend

The results of the filtering residuals are then added to the fitted curve to obtain smoothed  $CO_2$  and its long-term trend. The smoothed  $CO_2$  comprises the fitted trend, the fitted seasonality and the smoothed residuals (red line in Fig. 2d), which only removes short-term variations or noise. The long-term trend comprises fitted trend and residual trend, which removes seasonal cycle and noise (blue line in Fig. 2d).

#### 2.3.3. Calculate CO2 growth rate, GATM

 $G_{ATM}$  is determined by taking the first derivative of the long-term trend. However, the growth is made up of discrete points, e.g. the black dots in Fig. 3a shows the trend points. In this case, a cubic spline interpolation is applied to the trend points, in which the spline curve passes through each trend points, as the blue line in Fig. 3a.  $G_{ATM}$  is obtained by taking the derivative of the spline at each trend point (Fig. 3b).

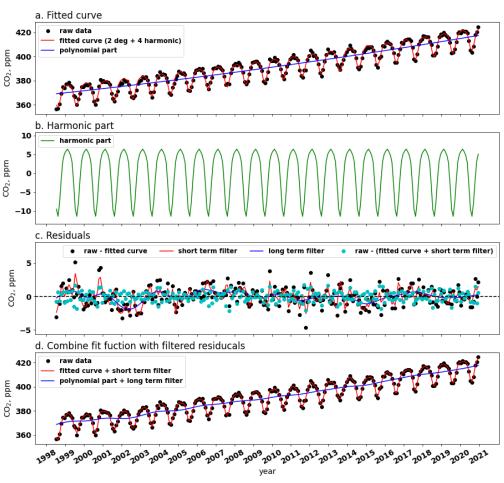


Figure 2. Example of analysed  $CO_2$  data from PAL station (Pallas, Finland), illustrating semi-NOAA curve fitting and filter method. Panel (a) shows monthly averaged  $CO_2$  (dots), curve fitting with 2-degree polynomial and 4-degree harmonics (red line), and long-term trend estimated by a 2-degree polynomial (blue line). Panel (b) shows seasonality estimated by 4-degree harmonics. Panel (c) shows the residuals of raw data from the function fit (black dots), the red





line is obtained by the short-term filter and the blue line is obtained by the long-term filter. The cyan dots show the residuals of raw data from the sum of fitted curve and smoothed residuals. Panel (d) shows final processed CO<sub>2</sub>, which comprises fitted trend, fitted seasonality and smoothed residuals (red line). The blue line shows final trend which comprises fitted trend and residuals trend.

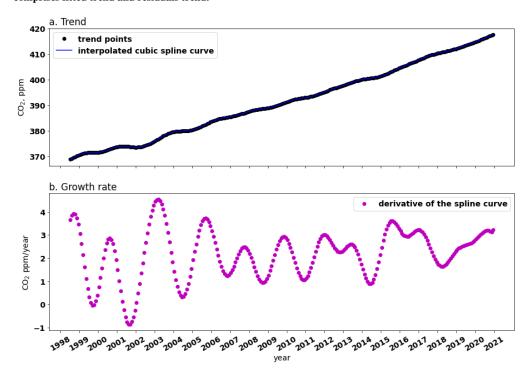


Figure 3. Example of  $CO_2$  growth rate, the raw data is the same as used in Fig. 2 from station PAL (Pallas, Finland). Panel (a) shows the trend points (black dots) and its cubic spline interpolation (blue line). Panel (b) shows the  $G_{ATM}$  at each trend point.

# 3 Results

Global averaged surface CO<sub>2</sub> and its G<sub>ATM</sub> are calculated from the GAW observations from 139 sites (Fig. 1) using the WDCGG method with and without extrapolation and our semi-NOAA method, namely GAW (WDCGG+), GAW (WDCGG) and GAW (semi-NOAA). The semi-NOAA method is also applied to three CTE datasets: 1) observations from 230 sites selected in the CTE dataset (hereafter these sites are named as CTE network, Fig. 1) which comes from the ObsPack data product (Kenneth N., 2022), namely CTE\_obs (semi-NOAA); 2) CTE model output at the sites (sampled at the same location, altitude and time), namely CTE\_output (semi-NOAA); and 3) model output for full global grids (averaged over the first three levels, 0 to 0.35 km Alt.), namely CTE\_global (semi-NOAA). We calculated the global means and its G<sub>ATM</sub> by area-weighted averaging the zonal means over each latitudinal band (30°), as same as the CO<sub>2</sub> averaging method in Tsutsumi et al. (2009). A bootstrap method is used to estimate the uncertainties of global CO<sub>2</sub> mean and its G<sub>ATM</sub>, which is an almost identical uncertainty analysis as presented by Conway et al. (1994) who constructed 100 bootstrap networks for the NOAA analysis. We construct 200 bootstrap networks which is consistent with the WDCGG analysis in Tsutsumi et al., (2009). For each bootstrap network, we randomly draw the same number of sites (as the actual network, e.g. 139 sites for GAW network) with replacement (or restitution) from the actual network, which means some sites are missing whereas others will be represented





twice or more often. We calculate global mean  $CO_2$  mole fraction and its  $G_{ATM}$  for each network, and then calculate the statistics (i.e. mean and 68% confidence interval, CI) on the 200 networks. All uncertainties in this paper are reported as  $\pm$  68% CI.

## 3.1 Globally averaged surface CO2 mole fraction and its GATM

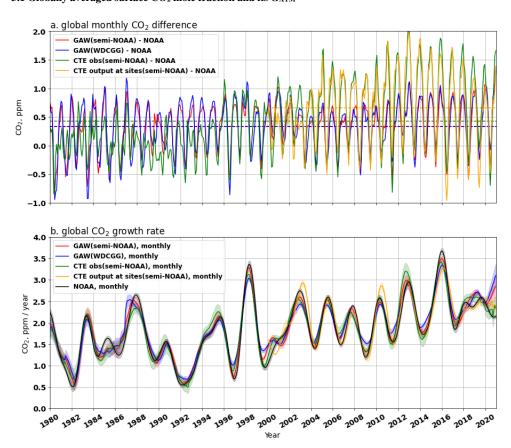


Figure 4. Comparison of globally averaged  $CO_2$  mole fraction (a) and its  $G_{ATM}$  (b) from 1980 to 2020. Panel (a) shows the global monthly  $CO_2$  mole fraction from 139 GAW sites (estimated from observations only) and those from 230 sites used in CTE (either from observations or model output) differs from NOAA estimates based on 43 MBL sites. Red and blue lines show the  $CO_2$  derived from the GAW observations using semi-NOAA and WDCGG method without extrapolation, respectively. Green and orange lines show the  $CO_2$  derived from observations and model output at the 230 sites assimilated by CTE using semi-NOAA method, respectively. The dash lines show the mean over the available period. Panel (b) compares the global  $CO_2$  growth rate derived from GAW observations using semi-NOAA (red line) and WDCGG method without extrapolation (blue line), CTE observations (green line) and model output (orange line) using semi-NOAA method, and the NOAA analysis (black line). The shadow area shows the uncertainty as 68% confidence interval obtained by the bootstrap analysis.

Global averaged surface CO<sub>2</sub> mole fraction derived from the GAW network (GAW (semi-NOAA) or GAW (WDCGG)) is 0.329 or 0.336 ppm significantly (p<0.05) higher than the NOAA analysis during 1980-2020 (red or blue line in Fig. 4a, Table S1a-b), this result is consistent with Tsutsumi et al., (2009) who found a 0.350 ppm higher global average in the GAW network during 1983-2006. The higher estimate from the GAW network can be explained by inclusion of more diverse sites, not only NOAA's MBL sites, but also more continental sites (Fig. 1). Both global CO<sub>2</sub> and its G<sub>ATM</sub> derived from the GAW (semi-

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217 NOAA) and GAW (WDCGG) are nearly overlapping (the red and blue lines) in Fig. 4a and 4b (as can also be seen by 218 comparing Fig. S1 and S2). The statistic metrics (Table S1a) show high agreement (r=0.999, RMSE=0.053 ppm, ME=0.007 ppm for the CO<sub>2</sub> mole fraction; r=0.991, RMSE=0.081 ppm yr<sup>-1</sup>, ME=0.005 ppm yr<sup>-1</sup> for the G<sub>ATM</sub>) between these two 219 220 methods, which confirms that the semi-NOAA method agrees well with WDCGG method without extrapolation. The 221 WDCGG method with extrapolation (i.e. GAW (WDCGG+)), where the long-term trend of each station is extrapolated to the 222 most long-running station period and added to its average seasonal variation to synchronize data period of all stations (Tsutsumi et al., 2009), produces ~0.096 ppm significantly (p<0.05) higher values than the global surface CO<sub>2</sub> mole fraction 223 224 derived from the GAW (WDCGG) during the common period 1984-2020 (see Table S2), while the extrapolation has tiny 225 effect (RMSE=0.062 ppm yr<sup>-1</sup>, ME=-0.011 ppm yr<sup>-1</sup>, Table S2) on the CO<sub>2</sub> growth rate. 226 Global averaged surface CO<sub>2</sub> derived from CTE\_obs (semi-NOAA) and CTE\_output (semi-NOAA) are 0.422 ppm (1980-227 2020) and 0.656 ppm (2001-2020) significantly (p<0.05) higher compared to the NOAA analysis, respectively (green and orange lines in Fig. 4a). Comparing the global mean of CTE\_obs (semi-NOAA) with CTE\_output (semi-NOAA) during the 228 229 common period 2001-2020, we find a low bias (0.069 ppm in CTE\_output, Table S1d-e and Table S3), which indicates that 230 the CTE model results can reproduce the global mean CO2 levels reasonably well. The global annual CO2 mole fraction from 231 CTE\_obs (semi-NOAA), CTE\_output (semi-NOAA) and CTE\_global (semi-NOAA) is 0.368 (2001-2020), 0.299 (2001-232 2020) and 0.186 (2001-2020) ppm significantly (p<0.05) higher than the result of the GAW (semi-NOAA), respectively (Table 233 S1d-f). The higher global mean from CTE\_obs (semi-NOAA) and CTE\_output (semi-NOAA) is mainly due to more sites in 234 the Northern Hemisphere in the CTE network compared to the GAW network. The lower bias between GAW (semi-NOAA) 235 and CTE\_global (semi-NOAA) indicates that the GAW network provides a good representation of the low-level atmosphere (i.e. 0 to 0.35 km altitude) at global scale (Table S1f), or the CTE model has a good performance in the low-level atmosphere. 236



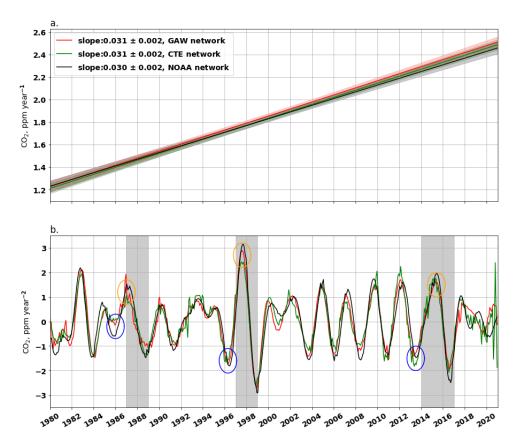


Figure 5. Trend analysis of the global CO<sub>2</sub> growth rate from 1980 to 2020. Panel (a) shows the trends of CO<sub>2</sub> growth rate for the GAW network (red line), the CTE network (green line) and the NOAA network (black line) during the whole period 1980-2020, the CO<sub>2</sub> growth rate is derived from GAW (semi-NOAA), CTE\_obs (semi-NOAA) and NOAA analysis (Fig. 4b). Panel (b) shows the trend of CO<sub>2</sub> growth rate for each month during 1980-2020, calculated as the derivative of the growth rate. The grey bands mark the period of three strong El Niño events, i.e 1987-1988, 1997-1998 and 2014-2016.

Despite differences in the global averaged surface  $CO_2$  mole fractions derived from different networks and analysis methods, the  $G_{ATM}$  derived from GAW network, CTE network and its model output, and NOAA network agree well (r>0.903, RMSE<0.192 ppm yr<sup>-1</sup>, MAE<0.158 ppm yr<sup>-1</sup>, ME<0.025 ppm yr<sup>-1</sup>, Table S1) during the common period (Fig. 4b). The trend analysis shows that the  $G_{ATM}$  increased steadily  $(0.030 \pm 0.002$  ppm per year each year) from 1980 to 2020 (Fig. 5a) based on the observations from the three networks (i.e. GAW, CTE and NOAA). This implies that over long-term period (here 40 years) the networks with and without continental sites show the same trend of the  $G_{ATM}$ . Hence, the  $CO_2$  advective transport and mixing plays a negligible role in estimating the long-term change of the  $G_{ATM}$ . However, there is a clear difference in the short-term (here one month) change of the  $G_{ATM}$  between the networks with and without continental sites (Fig. 5b). The El Niño event often diminishes net global C uptake (due to e.g. droughts, floods and fires) and increases global  $CO_2$  growth rate (Sarmiento et al., 2010). The  $G_{ATM}$  derived from the GAW and CTE network (red and blue lines) increases earlier before the three strong El Niño events (marked as blue circles in Fig. 5b) and reaches the peak earlier during the El Niño events (marked as orange circles in Fig. 5b), compared to the  $G_{ATM}$  derived from the NOAA network (black line). This indicates that continental sites can help early detection of the change of  $G_{ATM}$  which is caused by biogenic emission or uptake changes. The



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257 CTE network (green line) detects the change even earlier than the GAW network (red line) for the three El Niño events (Fig.

5b), which is due to even more continental sites included in the CTE network (Fig. 1), although the more continental sites also

259 induce the larger variability.

Table 1 shows the global annual  $CO_2$  and its  $G_{ATM}$  derived from GAW (semi-NOAA), together with the uncertainty estimated by the bootstrap method. The global average surface  $CO_2$  mole fraction has increased from 339.17±0.38 ppm in 1980 to 413.06±0.16 ppm in 2020 (Table 1, Fig. S1). The uncertainty before 1990 is larger than after 1990, due to fewer measurement stations over the globe before 1990. The average  $G_{ATM}$  for the two decades before 2000 is about 1.54±0.08 ppm yr<sup>-1</sup>, however, in the following two decades it increases to 1.91±0.05 ppm yr<sup>-1</sup> (2000-2009) and 2.41±0.06 ppm yr<sup>-1</sup> (2010-2019) (Table 1, Fig. S1).

Table 1. Annual global averaged CO<sub>2</sub> mole fraction (Mean, ppm) and its G<sub>ATM</sub> (ppm yr<sup>-1</sup>) derived from GAW observations using semi-NOAA method. U(Mean) and U(G<sub>ATM</sub>) respectively indicate the uncertainty of Mean and its G<sub>ATM</sub> as 68% confidence interval. The annual value is averaged over the monthly values of the year.

Year	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989
Mean	339.17	340.16	341.03	342.59	344.46	345.69	347.08	348.99	351.45	353.15
U(Mean)	0.38	0.24	0.19	0.24	0.26	0.22	0.14	0.15	0.12	0.15
$G_{ATM}$	1.65	1.07	0.88	2.02	1.32	1.38	1.55	2.38	2.08	1.23
U(G <sub>ATM</sub> )	0.12	0.10	0.15	0.13	0.08	0.11	0.14	0.08	0.09	0.06
Year	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Mean	354.22	355.64	356.37	357.09	358.51	360.52	362.27	363.40	366.14	368.10
U(Mean)	0.10	0.11	0.10	0.10	0.11	0.12	0.12	0.10	0.10	0.10
$G_{ATM}$	1.41	1.03	0.65	1.22	1.72	2.06	1.16	1.82	2.89	1.34
U(G <sub>ATM</sub> )	0.08	0.06	0.05	0.05	0.05	0.08	0.07	0.05	0.05	0.05
Year	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009
Mean	369.30	370.77	372.92	375.45	377.22	379.28	381.38	383.20	385.26	386.78
U(Mean)	0.12	0.11	0.10	0.10	0.10	0.10	0.09	0.10	0.10	0.11
$G_{ATM}$	1.58	1.58	2.33	2.17	1.66	2.42	1.75	2.20	1.71	1.68
U(G <sub>ATM</sub> )	0.05	0.06	0.06	0.04	0.04	0.03	0.05	0.04	0.05	0.04
Year	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
Mean	389.01	390.97	393.14	396.00	397.79	400.12	403.47	405.70	407.93	410.57
U(Mean)	0.12	0.12	0.14	0.11	0.10	0.10	0.11	0.09	0.10	0.13
$G_{ATM}$	2.32	1.73	2.74	2.30	1.91	2.98	2.95	2.04	2.50	2.61
U(G <sub>ATM</sub> )	0.05	0.06	0.09	0.05	0.04	0.05	0.06	0.06	0.07	0.05
Year	2020									
Mean	413.06									
U(Mean)	0.16									
$G_{ATM}$	2.60									
U(G <sub>ATM</sub> )	0.16									



#### 3.2 Vertical profile of global CO<sub>2</sub> mole fraction

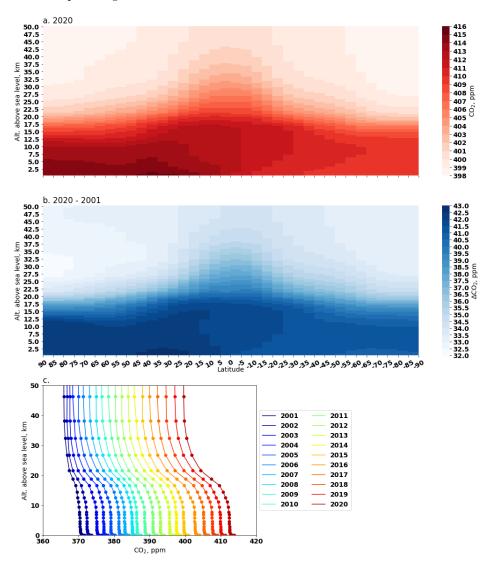


Figure 6. Global vertical profile of CO<sub>2</sub> mole fraction derived from CTE model output. Panel (a) shows the vertical profile in 2020. Panel (b) shows the difference of the vertical profile between 2001 and 2020. Panel (c) shows the annual mean vertical profile from 2001 to 2020, the dots mark CTE vertical level heights and lines are the linear interpolation between the heights.

The CTE model simulates CO<sub>2</sub> mole fraction over a global 3D grid, which allows us to view the modelled vertical CO<sub>2</sub> profile. In the lower atmosphere, highest CO<sub>2</sub> mole fraction are found in the Northern mid-latitude (dark red between 30 °N and 40 °N, Fig. 6a), where more anthropogenic emissions take place, which are subsequently transported towards northern and southern latitudes. The latitudinal and interhemispheric gradient of atmospheric CO<sub>2</sub> found in Fig. 6a, is not only determined by the latitudinal and interhemispheric differences in fossil fuel emissions and seasonal exchange with terrestrial biota (Denning et al., 1995), but is also due to atmospheric transport (Patra et al., 2011). With increasing altitude, the gradient





between Northern and Southern hemisphere becomes small and levels out at higher altitudes (e.g. >50 km). When comparing the vertical profile change between 2001 and 2020 (Fig. 6b and 6c), the CO<sub>2</sub> mole fraction increases slower at the higher atmosphere (>25 km altitude) than the increase at the lower atmosphere (<25 km altitude). Fig. 6c shows that the vertical gradient (difference between 50 km and 0.05 km) changes from ~5 ppm for 2001 to ~13 ppm for 2020. The high vertical gradient in 2020 reflects the accumulation of CO<sub>2</sub> in the lower atmosphere, which is caused by continuous CO<sub>2</sub> emissions from the surface during 2001-2020 and slow vertical transport. The low vertical gradient in 2001 is partly due to low surface emission.

#### 3.3 Relationship between the surface CO<sub>2</sub> mole fraction and atmospheric CO<sub>2</sub> mass

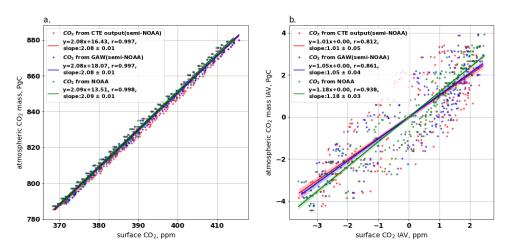


Figure 7. Relationship between the monthly surface  $CO_2$  mole fraction and atmospheric  $CO_2$  mass. The atmospheric  $CO_2$  mass calculated from the 3D CTE output. In panel (a), the monthly surface  $CO_2$  derived from the CTE\_output (semi-NOAA), GAW (semi-NOAA) and NOAA analysis, presented as blue, red and green dots, respectively. Panel (b) compare the corresponding interannual variability (IAV) of the atmospheric  $CO_2$  mass and the surface  $CO_2$ . The IAV is calculated as the anomaly departure from a quadratic trend.

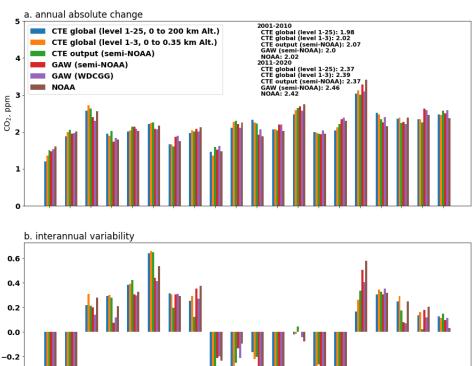
Atmospheric CO<sub>2</sub> mass calculated from the CTE output as a function of air mass and CO<sub>2</sub> concentration (Text S3), has increased from 789.46 PgC in 2001 to 877.88 PgC in 2020 (Fig. S3a). The spatial distribution of the atmospheric CO<sub>2</sub> mass can be seen in Fig. S3b and Fig. S3c. Monthly global surface CO<sub>2</sub> mole fraction derived from CTE output (red dots, Fig. 7a) at the 230 sites used in CTE with the semi-NOAA method (CTE\_output (semi-NOAA)) and GAW observations (blue dots, Fig. 7a) at 139 GAW sites with the semi-NOAA method (GAW (semi-NOAA)) has a similar linear relationship (showing the same slope of 2.08±0.01 PgC ppm<sup>-1</sup>) as the monthly atmospheric CO<sub>2</sub> total mass derived from the CTE output. The NOAA CO<sub>2</sub> (green dots, Fig. 7a) shows a similar linear relationship (has a slope of 2.09±0.01 PgC ppm<sup>-1</sup>). The slope or conversion factor in Fig. 7a is slightly lower than the factor 2.12 PgC ppm<sup>-1</sup> used in Ballantyne et al. (2012) for the period 1980-2010. The small difference in conversion factor is expected, considering the different model and data used. We further compare the interannual variability (IAV, calculated as the anomaly departure from a quadratic trend) of the atmospheric CO<sub>2</sub> mass and the surface CO<sub>2</sub> (Fig. 7b), the coefficient of the linear relationship is very close to ~1.0, which indicates the temporal change in atmospheric CO<sub>2</sub> mass agrees with temporal change in surface CO<sub>2</sub> mole fraction. The NOAA network tracks atmospheric CO<sub>2</sub> change slightly better (r=0.938) than the GAW (r=0.861) and CTE (r=0.812) networks, given the long resident time and well-mixed nature of atmospheric CO<sub>2</sub>. Overall, the relationship found in Fig. 7 implies that the current surface CO<sub>2</sub> network can be a good indicator of the CO<sub>2</sub> mass change in the whole atmosphere through a linear relationship.



CO<sub>2</sub> IAV, ppm



#### 3.4 Annual absolute change and interannual variability of global CO<sub>2</sub> mole fraction



-0.4 -0.6 200<sup>1</sup> 200<sup>2</sup> 200<sup>3</sup> 200<sup>4</sup> 200<sup>5</sup> 200<sup>6</sup> 200<sup>1</sup> 200<sup>8</sup> 200<sup>9</sup> 20<sup>10</sup> 201<sup>1</sup> 201<sup>2</sup> 201<sup>3</sup> 201<sup>4</sup> 201<sup>5</sup> 201<sup>6</sup> 201<sup>1</sup> 201<sup>8</sup> 201<sup>9</sup> 202<sup>0</sup>

Figure 8. Annual absolute change and interannual variability of global  $CO_2$  mole fraction derived from different data (CTE model, GAW observation and NOAA observation) and analysis methods (semi-NOAA method, WDCGG method and NOAA method) for 2000-2020. Panel (a) shows the annual absolute change which is the difference between annal mean. Averages over 2001-2010 and 2011-2020 are also shown. Panel (b) shows the IAV which is calculated as the anomaly departure from a quadratic trend.

Pressure-weighted average CO<sub>2</sub> in the lower atmosphere and whole atmosphere is derived from CTE output. The annual absolute change (calculated as the difference between annual mean) of CO<sub>2</sub> in the lower atmosphere (0 to 0.35 km altitude, orange bars in Fig. 8a) is more sensitive to surface sink and source than the change in the whole atmosphere (blue bars). The reason is that the whole atmosphere has a larger air volume than the lower atmosphere, and the change of the surface CO<sub>2</sub> is diluted due to horizontal and vertical transport. The CO<sub>2</sub> change derived from the observations of the GAW network (red bars for semi-NOAA method, purple bars for WDCGG method) and the NOAA network (brown bars), shows a small positive or negative difference from the CTE results over the different years. However, over the long term (e.g. decadal scale, 2001-2010 and 2011-2020), the CTE model derived change of lower and whole atmospheric CO<sub>2</sub> shows good agreement (<0.09 ppm yr<sup>-1</sup>) with the surface observation-based estimate, especially for the lower atmospheric CO<sub>2</sub> (<0.07 ppm yr<sup>-1</sup>). Fig. 8b shows the IVA derived from CTE (blue, orange and green bars) follows a similar temporal pattern as the observation-based IVA derived



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- 327 from the GAW and NOAA network (red, purple and brown bars), especially the IVA of the low-level atmosphere (orange
- bars) show good agreement with the observation-based IVA (r>0.971, RMSE<0.178 ppm).

#### 4 Discussion

During the past few decades, observational networks have been extended (e.g. from the NOAA MBL network) to the continents (e.g. GAW network and CTE network, Fig. 1) in order to monitor global CO2 concentrations and quantify CO2 sources and sinks. Although the continental observations include contributions from both big sources of anthropogenic emissions and big sources/sinks from terrestrial vegetation off/during the growing season, these continental observations show an overall higher global surface CO2 mole fraction in the global CO2 analysis which indicates that they are influenced by a net source. We find that the global mean derived from the GAW network is on average 0.329 (semi-NOAA method) or 0.336 (WDCGG method) ppm consistently higher than that derived from the NOAA network during 1980-2020, similarly ~0.350 ppm higher mole fraction in the GAW network was found in Tsutsumi et al. (2009) for 1983-2006. The CTE network even leads to a higher global mean (0.422 ppm during 1980-2020), which is likely due to more observational sites locate in the Northern Hemisphere where the highest anthropogenic emissions take place. This also explains the large fluctuation of CO2 concentration during the winters and summers during 2001-2020 (green and orange lines, Fig. 4a). In future, we expect that adding new observation sites (specially in Northern Hemisphere) into the current observational network (e.g. GAW network), would lead to higher global surface CO<sub>2</sub> and a larger amplitude of the global CO<sub>2</sub> seasonal cycle in the global CO<sub>2</sub> analysis. Although Friedlingstein et al. (2022) reported a 5.4% drop (~0.52 PgC) in fossil fuel CO2 emissions in 2020 (due to restrictions on e.g. transport, industry, power etc during the COVID-19 pandemic), the increase in annual CO<sub>2</sub> from 2019 to 2020 (2.60±0.16 ppm yr<sup>-1</sup>) remains at a similar level as from 2018 to 2019 (2.61±0.05 ppm yr<sup>-1</sup>). In principle, an equivalent drop of roughly 0.25 ppm yr<sup>-1</sup> (according to the conversion factor 2.08 PgC ppm<sup>-1</sup> in Fig. 7a) or roughly 0.13 ppm yr<sup>-1</sup> (according to the annual absolute change, red bars in Fig. 8a) in the growth rate should be visible for period 2019-2020 due to the declined CO<sub>2</sub> emission. However, such short-term human activity induced change of the CO<sub>2</sub> growth rate may be hidden by the natural variability. The bootstrap analysis is used in this study (also in Conway et al., (1994) and Tsutsumi et al., (2009)) to estimate the uncertainty of the CO2 temporal mean and its growth rate and to assess how sensitive the global value is to the distribution of sampling sites. The relatively large uncertainty (±0.16 ppm yr<sup>-1</sup>) at the end of 2020 compared to previous years (Table 1) is likely due to end-effect associated with the curve fitting and filter procedure. The end-effect is a tendency for the growth rate to turn toward the mean value at the end of the record (Conway et al., 1994), therefore Conway et al. (1994) suggested the last 6 months of the growth rate curves should be viewed with caution. Our analysis shows that basing the CO2 growth rate on GAW surface observations does not introduce a large bias (on average agreement within 0.015 ppm yr<sup>1</sup>) compared to a full atmospheric analysis (Fig. 4b and 8, Table S1e-f). This full atmosphere CO2 was provided by the CTE model, in which the global annual mean CO2 is significantly overestimated compared to GAW observations (e.g. 0.299 ppm higher in CTE\_output (semi-NOA), or 0.186 ppm higher in the CTE\_global (semi-NOAA) during 2001-2020). The overestimate derived from the CTE\_output (semi-NOAA), i.e. CTE outputs at the CTE 230 sites, is mainly due to more sites in the Northern Hemisphere in the CTE network than in the GAW network. The lower overestimate derived from the CTE\_global (semi-NOAA), i.e. CTE outputs at full global grids at the low-level atmosphere, implies that the biases in CTE are not uniform spatially and attempt to balance out. We estimate the CTE bias by comparing the observations and CTE outputs at the same sites, which results in 0.069 ppm low bias derived from the CTE outputs in calculating the global surface CO2 mole fraction.



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Different observational networks (i.e. NOAA network, GAW network and CTE network) are analysed in this study, which shows a difference in calculated global surface CO<sub>2</sub> mole fractions equivalent to the current atmospheric growth rate over a three-month period. This implies that the station selection, especially if and how many continental observations are used, has some but not a particularly strong influence on the derived global surface CO2. Nowadays more and more continental observations are established in order to monitor biogenic sources and sinks, and further provide insight into the climate change and the associated ecosystem processes (Ciais et al., 2005, Ramonet et al., 2020). Such continental observations carry more variability in measurements than the marine observations, which needs some caution when used in the mix of stations for determining global surface CO2 mole fraction. However, our study shows that continental sites can help early detect the change of CO2 growth rate caused by biogenic emission change (e.g. caused by El Niño events). Besides, current observational networks (with and without continental sites) and CTE model show a good agreement within 0.025 ppm yr<sup>-1</sup> on the global CO2 growth rate over long-term period. This implies that the current observation networks (e.g. as shown in Fig. 1, represent for multiple ecosystems, multiple sinks and sources, and different latitudes) have a similar good capacity to capture the global surface CO<sub>2</sub> changes, although there is the spatial and temporal variability of the CO<sub>2</sub> growth rate (e.g. Conway et al. 1994). We also notice that the uncertainty global CO2 growth rate is approximately 0.07 ppm yr<sup>-1</sup>, as derived from GAW (semi-NOAA) and averaged over 1980-2020 (Table 1). In order to reduce this uncertainty, we recommend adding more stations to the current observation network. We conducted an experiment (Fig. S4) which demonstrates that the uncertainty of the global CO<sub>2</sub> growth rate exponentially increases as the number of land observation sites is reduced. To reduce the uncertainty to 0.02 ppm yr1 (equivalent to 1% of the global CO2 growth rate), our experiment indicates that 332 land observation sites are required (Fig. S4). However, the required number of sites also depends on their geographical distribution (i.e. CO<sub>2</sub> footprint coverage of observation network, and the importance of the network design was addressed by Storm et al. (2022)), measurement accuracy, and consistency. Extrapolation beyond the measurement period extends knowledge gained from a limited period of measurements. During a limited period of measurement, we can define the average seasonality, long-term trend, and short-term variation at a measurement site. The long-term trend of individual site is extrapolated, for example by referring to the latitude reference time series (Masarie and Tans, 1995) or the mean long-term trend over sites within a certain (e.g. 30°) latitudinal zone (Tsutsumi et al., 2009), and then combining the extrapolated trend with average seasonality to produce the estimate beyond measurement period. The extrapolation requires the assumption that the relationship of an individual site to the latitude reference is invariant in time, however, the relationship between nearby sites is continuously changing (Masarie and Tans, 1995). Besides, the short-term variation is ignored or estimated from nearby sites, which introduces extra uncertainty from extrapolation. In this study, we find that the WDCGG method with extrapolation (GAW (WDCGG+)) results in ~0.096 ppm higher in the global surface CO<sub>2</sub> mole fraction than the WDCGG method without extrapolation (GAW (WDCGG)) using the same GAW observations, although the extrapolation has a tiny effect on the growth rate (Table S2). Therefore, extrapolation beyond the measurement period is not used in our analysis. With the increasing number of long-term measurements, this extrapolation becomes less and less necessary.

# 5 Conclusions

The WMO Global Atmosphere Watch CO<sub>2</sub> network documents the gradual global accumulation of CO<sub>2</sub> in the atmosphere due to human activities, and has been used to assess large-scale and long-term environmental consequence of fossil CO<sub>2</sub> emission and land use changes. Although the current CO<sub>2</sub> network is sparse due to operational costs and logistical constraints, it has a good capacity to represent global surface CO<sub>2</sub> mole fraction and its growth rate and trends in atmospheric CO<sub>2</sub> mass





404 changes. The three different analysis methods yield very similar global CO2 increase from 2001 to 2020, which gives 405 confidence to use either one of them in climate change study. The continuous monitoring the atmospheric CO2, basing on the 406 current GAW network together with reliable global data integration methods, provides essential information for policymakers 407 to support their efforts in mitigating the global warming. 408 6 Data and Code Availability 409 All data and code necessary to calculate the global mean surface CO2 mole fraction and Atmospheric CO2 mass is freely 410 available from ICOS Carbon Portal [https://doi.org/10.18160/Q788-9081]. The file list of results and code can be found in 411 Text S4. 412 Acknowledgments 413 We acknowledge Ingrid Luijkx for providing the TM5 data, WMO GAW Principal Investigators of the WMO GAW station network for providing the observational data, Ed Dlugokencky for providing NOAA data and comments. Thanks to the support 414 415 from ICOS, GAW, NOAA and CTE group. 416 **Author Contributions** 417 A.V. and Z.W. designed this study in discussion with Y.S., O.T and U.K.. 418 Z.W. performed analysis and led the writing. 419 Y.S., Y.N. and A.O. provided the GAW data, and commented on the manuscript. 420 W.P. and R.K. provided CTE model results and relevant ObsPack data, and commented on the manuscript. 421 X.L. provided NOAA data and commented on the manuscript. 422 All authors contributed to the writing of the paper and interpretation of the results. 423 **Competing Interests** 424 The authors declare no competing interests. 425 426 Financial support This research is a part of ICOS core work, there is no grant reference no. 427 428 References 429 BALLANTYNE, A. Á., ALDEN, C. Á., MILLER, J. Á., TANS, P. Á. & WHITE, J. 2012. Increase in observed net carbon dioxide uptake 430 by land and oceans during the past 50 years. Nature, 488, 70-72. https://doi.org/10.1038/nature11299 431 CIAIS, P., REICHSTEIN, M., VIOVY, N., GRANIER, A., OGÉE, J., ALLARD, V., AUBINET, M., BUCHMANN, N., BERNHOFER, 432 C. & CARRARA, A. 2005. Europe-wide reduction in primary productivity caused by the heat and drought in 2003. Nature, 437, 433 529-533. https://doi.org/10.1038/nature03972 434 CONWAY, T. J., TANS, P. P., WATERMAN, L. S., THONING, K. W., KITZIS, D. R., MASARIE, K. A. & ZHANG, N. 1994. Evidence 435 for interannual variability of the carbon cycle from the National Oceanic and Atmospheric Administration/Climate Monitoring 436 and Diagnostics Laboratory global air sampling network. Journal of Geophysical Research: Atmospheres, 99, 22831-22855.

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