Dear Editor,

According to the reviewers' comments and suggestions, we have made major revision to our manuscript. The main changes in the manuscript are as follows:

1) As suggested by one reviewer, we add an analysis of uncertainty reduction and add a new section of "uncertainty reduction".

2) We reorganize Section 2.1 to further clarify the differences between GCASv1 and GCASv2.

3) We add more discussions about the use of 1-week assimilation window.

4) We added 4 sensitivity experiments for the year of 2010 to test the impact of assimilation window, spurious signals, and uncertainty in fossil fuel carbon emissions on the inverted global and regional BIO carbon fluxes.

The point-by-point response to the reviews and the detailed changes are listed in the attachments. Many thanks to you and the referees for the time and effort you expend on this paper.

Best Regards,

Sincerely yours,

Fei Jiang

Referee #1

We would like to thank the anonymous referee for his/her comprehensive review and valuable suggestions. These suggestions help us to present our results more clearly. In response, we have made changes according to the referee's suggestions and replied to all comments point by point. All the page and line number for corrections are referred to the revised manuscript, while the page and line number from original reviews are kept intact.

General comments.

Authors present estimates of regional carbon dioxide flux variability based on assimilating GOSAT satellite observations of CO_2 with ensemble-based data assimilation system. The estimated CO_2 fluxes where evaluated by comparison to indexes of climate variability, and published top-down and bottom-up estimates. The analysis of the carbon cycle variability and comparison with data on climate variability makes a strong point of the study. On the other hand, the description of the ensemble-based data assimilation system can be improved. The paper is well written and can be accepted after minor revisions addressing the review suggestions.

Detailed comments.

Lines 130-139 Suggest clarifying, what becomes a state vector to optimize, currently it is implicit. Some details emerge much later on Lines 358-366, when uncertainties are discussed. Response: Thank you for this suggestion. In this study, the terrestrial ecosystem (BIO) and ocean (OCN) carbon fluxes are treated as state vector and optimized. Indeed, as you said, the state variables had been mentioned in two places in the article. The first place is in the section of system description, and the second is in the section of "Experimental Design". In the first place, we are introducing the current system (GCASv2) that we have improved, we set 4 state vector schemes in this system for different applications: 1) only the BIO flux is state vector; 2) both BIO and OCN fluxes are treated as state vectors; 3) the BIO, OCN and FOSSIL fluxes are optimized at the same time; and 4) only net flux is optimized. In this study, we chose to optimize both BIO and OCN, which were introduced in the section of "Experimental Design". To further clarify the state vector of this study, we added a sentence of "*In this study, the second scheme was selected.*" at the end of the 2nd paragraph in section 2.1 (see Line 178, Page 7).

Lines 232-236 The logic behind selecting 1-week data assimilation window doesn't look solid, as the other ensemble-based assimilation systems use longer window in order of 12 weeks, (Peters et al. 2005, Feng et al. 2009, Jacobson et al. 2020. The notice that there was a problem reproducing CO₂ growth rate with a longer window in Zhang et al (2015) doesn't look like a strong argument, if considered in comparison with other studies. Response: Many thanks for this suggestion. We have added more discussions about the assimilation window, and shown the mean observation (only GOSAT XCO₂) number (Figure S2) during the study period that each grid could have within the 1 week assimilation window and the 3000 km localization scale. We also conduct a test in the year of 2010 for different DA windows (1, 2 and 4 weeks) and evaluate the posterior results using surface observations (see Table 1). We have revised that paragraph (see Lines 303-307, Lines 309-340, Pages 11-12) as follows:

"The DA window is set to one week in GCASv2, which is the same as before. Theoretically, a longer DA window is better, because CO_2 is a stable species. The longer window, the farther CO₂ will be transported. In this way, more observation stations will sense the flux change of one area, and thus more observations can be used to optimize the flux of that place. Therefore, many previous ensemble-based assimilation systems used a longer DA window (e.g., Peters et al. 2005, Feng et al. 2009, Jacobson et al. 2020). However, the farther away, the weaker signal the stations can sense. Bruhwiler et al. (2005) clearly shown that a pulse traveling from a faraway place would contribute relatively little signal compared to recent pulses from nearby source regions. In addition, Limited by the method of EnKF, this weak signal will be masked by the method's own unphysical signal (spurious correlation), and in order to reduce this influence, we must increase the ensembles, thereby greatly increasing the computational cost. Miyazaki et al. (2011) tested the differences of 3 days and 7 days DA windows, and pointed that with a longer DA window, more observation data will be available to constrain the surface flux, but a longer window can make the effect of model error more obvious. Thus, the assimilation result can be improved as long as the observations with spurious correlations can be neglected. However, spurious correlations can be more serious with increases in the DA window, because of a limited number of ensembles. As a result, a longer window is not necessarily better than a shorter window system. To avoid the influence of spurious signals, Kang et al. (2012) used a very short DA window (6 hours) in their assimilation system (LETKF C) and pointed out that the flux inversion with a long window (3 weeks) is not as accurate as the one obtained with a 6 h DA window, particularly in smaller-scale structures. During the development of GCASv1, Zhang et al. (2015) tested different DA windows and found that the longer the window, the larger optimized terrestrial carbon sink will be, resulting in a smaller optimized annual atmospheric CO₂ growth rate as compared to the observed rate. Considering the fact that at present, due to the release of satellite XCO₂ retrievals like GOSAT and OCO-2, the atmospheric CO_2 observations and coverages have increased significantly compared to before, which means that we do not need to extend the DA window to include more observation data now. Figure S2 shows the mean super observation (see section 2.1.1, only GOSAT XCO₂) numbers during the study period that each grid could have within the 1week DA window and a localization scale (3000 km, see the next paragraph). In most land areas and pan-tropical waters, each grid can already have more than 3 super observations. On average, each grid over the land could has 4 super observations. Two sensitivity tests in 2010 were conducted using 2- and 4- weeks DA windows but the same localization scale, the results are shown in Table S3. When the length of DA window increases from 1 week to 4 weeks, the mean super observation number increases from 4 to 9, accordingly, the inverted global BIO flux increased from -4.16 PgC yr⁻¹ to -4.49 PgC yr⁻¹, resulting in a larger deviation of the simulated and observed atmospheric CO₂ growth rate (AGR) and larger simulation error against the surface observations. Therefore, we still use the 1-week DA window in GCASv2."



Figure 1. Mean observation numbers within a DA window (1 week) during May 2009 ~ Dec 2015 (This figure has been added in the revised Supporting Information, and named as Figure S2)

Table 1. Results of sensitivity tests in the year of 2010 (1week, 2weeks and 4weeks are three additional experiments using 1 week, 2 weeks, and 4 weeks assimilation windows, respectively) (This Table has been added in the revised Supporting Information, and named as Table S4)

		Prior	1 week	2 weeks	4 weeks
Super Obs.	Total	-	730	1039	1360
Num. per window	Each grid over land	-	4	6	9
	BIO	-2.07	-4.16	-4.46	-4.49
Global Flux	OCN	-2.08	-2.33	-2.32	-2.35
(PgC/yr)	FOSSIL	9.07	9.07	9.07	9.07
	Net	7.25	4.91	4.62	4.55
	North America Boreal	-0.29	-0.43	-0.41	-0.35
	North America Temperate	-0.42	-1.25	-1.75	-2.41
Designal	Tropical South America	-0.17	-0.26	-0.32	-0.27
Flux (PaC/ur)	Temperate South America	-0.24	-0.4	-0.36	-0.19
(PgC/yr)	Northern Afirca	0.21	0.32	0.36	0.62
	Southern Africa	0.22	-0.3	-0.59	-1.04
	Boreal Asia	-0.4	-0.46	-0.3	0.11
	Temperate Asia	-0.3	-0.29	-0.15	-0.06

	Southeast Asia	-0.29	-0.23	-0.21	-0.2	
	Australia	-0.17	-0.4	-0.48	-0.53	
	Europe	-0.19	-0.41	-0.21	-0.12	
independent	BIAS	1.43	-0.44	-0.4	-0.38	
	MAE	1.92	1.37	1.39	1.51	
evaluation	RMSE	2.36	2.11	2.18	2.39	
Deviation from the observed AGR		2.08	0.26	0.55	0.62	
(PgC yr ⁻¹)		2.08	-0.20	-0.55	-0.02	

Technical corrections

Lines 119-120 Need to clarify, written that fluxes "are perturbed with a Gaussian random distribution" – better add more detail on whether perturbation is applied independently to each grid or over regions.

Response: Thank you! We have rewritten that sentence (see Lines 123-124, Page 5), as follows:

"the prior fluxes of X^b in each grid are independently perturbed with a Gaussian random distribution"

Line 216 As resolutions of the transport model and fluxes are apparently different, suggest writing which of them are referred as 'model grids'.

Response: Thanks for this suggestion. We have changed 'model grids' as 'transport model grids' (see Lines 268 and 269, Page 10).

Line 584 Revise 'a very stronger carbon sink' as 'a stronger carbon sink' or 'a very strong carbon sink'

Response: Thanks! We have changed 'a very stronger carbon sink' as 'a very strong carbon sink' (see Line 857, Page 29).

Line 594 Suggest revising 'weak' to 'weaker' Response: Thanks! We have changed 'weak' to 'weaker' (see Line 870, Page 30).

Referee #2

We thank the anonymous referee for his/her valuable comments and constructive suggestions. We have made changes according to the referee's suggestions and replied to all comments point by point. All the page and line number for corrections are referred to the revised manuscript, while the page and line number from original reviews are kept intact. The references related to the responses are listed in the end of this document.

General comments:

In this study, Jiang et al. upgraded the Global Carbon Assimilation System (GCAS) with new assimilation algorithms, a localization scheme, and a higher assimilation parameter resolution, namely GCASv2. The global terrestrial ecosystem (BIO) and ocean (OCN) carbon fluxes from 2009 to 2015 were constrained by the GOSAT ACOS XCO2 retrievals. Following this, the posterior carbon fluxes from 2010 to 2015 were evaluated using 52 surface flask observations. The errors in the posterior carbon fluxes in the new inversion system were compared to those in a previous version. The authors indicated that the pattern of regional carbon sinks was significantly different from previous studies (CT2017). The inter-annual variations of carbon fluxes in most land regions, and the relationship with the changes of severe drought area the plant indexes, and drought were re-visited. These results are interesting. However, the improvement of the inversion methodology is not presented, and the reduction of the uncertainty by the inversions remains unclear (Figure 3) in the current paper. I, therefore, recommend that this work cannot be published before the following comments are addressed.

Specific comments:

Figure 3: What is the source for error bars in these two plots? Are they coming from the uncertainty in the prior and posterior estimates? If yes, it seems that the uncertainty is not reduced from the prior estimates to the posterior estimates. One main purpose of inversion is to reduce the uncertainty in the prior estimates. If the uncertainty is not reduced, the effectiveness of the inversions should be evaluated.

Response: Thank you for this suggestion. The error bars represent the standard deviations of all biases at each latitude and each site, respectively. Indeed, the uncertainty reduction is very important for an inversion study. We analyzed the uncertainty reduction rate (UR), and added a section of "4.2 Uncertainty reduction" in the revised manuscript (see Lines 663 - 705, Pages 21 - 23). The annual mean URs of the BIO fluxes over different TRANSCOM regions are in the range of $6\% \sim 27\%$, with global mean of 17%. The highest monthly UR is 51% in temperate South America.

Figures 4/5: Evaluation of the reduction of the uncertainty from the prior estimates to the posterior estimates is more important than evaluation of the bias itself for an inversion system.

Response: Thank you for this suggestion. We have analyzed the reductions of the uncertainties from the prior estimates to the posterior estimates and added a section of "4.2 Uncertainty reduction" in the revised manuscript (see Lines 663 – 705, Pages 21 - 23).

Tables 2/3: What is the uncertainty for the prior and posterior estimates? Response: Thank you! We have added the uncertainties of the prior and posterior estimates in the revised manuscript (see Lines 719 – 723, Page 24 and Line 828, Page 28).

Line 473-488: What is the uncertainty for the estimates from this study? To evaluate the effectiveness of an inversion system, the uncertainty of the posterior estimates is more important than the central value. Such information is missing in the current manuscript, which is better considered / discussed in previous studies (e.g. the literature cited in line 586). Response: Thank you for this suggestion! As shown above, we have analyzed the uncertainty reductions and added a section of "4.2 Uncertainty reduction" in the revised manuscript (see Lines 663 – 705, Pages 21 - 23).

Figures 7/9/10: What is the uncertainty for the prior and posterior estimates? Response: Thank you for this suggestion. We have added the prior and posterior uncertainties in Figures 7, 9 and 10, which are named as Figure 8, 10 and 11 in the revised manuscript (see Lines 893-896, Page 31; Lines 934-937, Page 33; and Lines 1045-1049, Page 37).

Figure 1: The authors suggested that a new assimilation scheme is developed in this paper. Why not directly compare the flow charts between the GCAS and GCASv2 systems and show the difference?

Response: Many thanks for this suggestion. We have modified Figure 1 and given the differences in the flow charts between GCASv1 and GCASv2 (see Lines 147-148, Page 6).

Line 124: It seems that a major advance of GCASv2 against GCAS is that "In the second step, the MOZART-4 model is run again using the optimized fluxes of Xa, to generate new CO2 concentrations for the initial field of the next DA window. This DA flow chart is different from the previous version of GCAS, which runs the MOZART-4 model only once, and optimizes the fluxes and the initial field of the next window synchronously." However, I do not understand how this improves the inversion system. The old GCAS system produces the posterior global gridded carbon fluxes, which were used as prior fluxes as input to any other forward models to simulate the CO2 field. If the difference of GCASv2 was just that the posterior global gridded carbon fluxes were used by MOZART-4 to simulate the CO2 field, I cannot see how and why the inversing methodology is improved.

Response: Thank you for this comment. Indeed, as you said, the descriptions of the differences between GCASv2 and GCASv1 are rather vague. We have revised Section 2.1 to further clarify their differences. The main differences between GCASv2 and GCASv1 are as follows:

1) Optimization of the initial field of each window. In GCASv1, it is directly optimized using the observations, while in GCASv2, it is simulated using the posterior fluxes of the previous window. The advantage of this method in GCASv2 is that the assimilation errors could be

transported from one window to the next. If the fluxes are overestimated in one window because of some reasons, by this method, they will affect the concentrations of the next window, thereby the posterior fluxes of the next window will compensate the overestimations. While in GCASv1, since the initial field of each window is directly optimized using the observations, which means in each window, there are relatively perfect initial fields, the inversions of each window are independent, and the amount of overestimation or underestimation in one window will continue to accumulate until the end, leading to an overall overestimation or underestimation. In addition, due to the perfect initial field, the differences between the simulated and observed concentrations are only contributed by the errors in the prior fluxes of current window, resulting in a relatively smaller model – data mismatch, so as to weaken the assimilation benefits on fluxes. This difference is given in Lines 128 - 143, Page 5 in the revised manuscript.

2) State vector. In GCASv1, only BIO is state vector, while in GCASv2, we set 4 state vector schemes for different applications: 1) only the BIO flux is state vector; 2) both BIO and OCN fluxes are treated as state vectors; 3) the BIO, OCN and FOSSIL fluxes are optimized at the same time; and 4) only net flux is optimized. This difference is given in Lines 172 - 178, Page 7 in the revised manuscript.

3) Resolution of the state vectors. In GCASv1, the scaling factor λ is defined in different land and ocean areas based on 22 TRANSCOM regions (Gurney et al., 2002) and 19 Olson ecosystem types, as in CarbonTracker (Peters et al., 2007), while in GCASv2, we change to use a λ in each grid, meaning that for each grid, the perturbations of prior fluxes are independent, and the grid cell of λ could be set freely. This difference is given in Lines 154-161, Page 6 in the revised manuscript.

4) observation data. In GCASv1, only flask/in situ observations were assimilated, while in GCASv2, we added a module to assimilate the satellite XCO_2 retrievals, and allow users to simultaneously or separately assimilate the flask/in situ concentrations and the XCO_2 retrievals. See Lines 186 – 201, Pages 7 -8 in the revised manuscript. Besides, a 'super-observation' approach is also adopted in GCASv2, See Lines 202-215, Page 8 in the revised manuscript.

5) assimilation algorithm, in GCASv2, we added another EnKF algorithm, i.e., EnSRF. See Lines 223-227, Page 9.

Line 143: It seems that the carbon emission from cement production, a large part of CO2 source, is missed in this inversion system. This could be a big weakness of the current system. Response: Sorry, that description is not accurate enough. The carbon emission from cement production has been included in this study. The fossil fuel carbon emissions are obtained from NOAA's CarbonTracker, version CT2017, which is an average of the Carbon Dioxide Information Analysis Center (CDIAC) product (Andres et al., 2011) and the Open-source Data Inventory of Anthropogenic CO2 (ODIAC) emission product (Oda et al., 2018). We have checked the document of CT2017 and the introduction of CDIAC database, compared the annual global fossil fuel emissions in our system with the global emissions from the CDIAC website (https://cdiac.ess-dive.lbl.gov/), and confirmed that the carbon emission from

cement production has been included in this study. We have changed the sentence of "... atmosphere and ocean (OCN) carbon exchange, fossil fuel (FOSSIL) carbon emission and biomass burning (FIRE) carbon emission..." to "... atmosphere and ocean (OCN) carbon exchange, fossil fuel and cement production (FOSSIL) carbon emission and biomass burning (FIRE) carbon emission..." (see Lines 166-167, Page 7)

Line 143: What is the relationship between BIO and FIRE? Biomass sequestrates carbon from the atmosphere, and releases CO2 in biomass burning. Should FIRE be a part of BIO? Response: Yes, biomass burning carbon emission is a part of terrestrial ecosystem carbon flux. Terrestrial ecosystems uptake carbon through photosynthesis (GPP) and release carbon through respiration (ER) and biomass combustion (FIRE). The BIO flux defined in this study is the net flux of GPP and ER (ER-GPP). In many previous inversion studies, it is directly defined as net ecosystem exchange [NEE = ecosystem respiration (ER) – gross primary production (GPP)] (e.g., Hu et al., 2019; Peters et al., 2007, 2010), and the sum of NEE and FIRE is defined as net biosphere exchange (NBE, Liu et al., 2017). In the revised manuscript, we have changed the sentence of "... name terrestrial ecosystem (BIO) carbon flux, ..." to "namely terrestrial ecosystem (BIO) carbon flux (i.e., net ecosystem exchange (NEE) = ecosystem respiration (ER) – gross primary production (GPP)), ..." (see Lines 163-166, Pages 6-7)

Line 147: "FOSSIL and FIRE fluxes are assumed to have no errors, only BIO and OCN fluxes are optimized in an assimilation system". I do not think that this is the case in other inversion systems: (1) It needs clear justification by summarizing and tabulating the methodology in the literature. (2) The difference relative to a system with errors considered for FOSSIL and FIRE need to be calculated to show how much the conclusion of the present study are sensitive to this assumption.

Response: Thank you for this comment. Yes, there are considerable uncertainties for the fossil fuel and biomass burning carbon emissions, which are about 6% and 20% for global mean, respectively. Ideally, we would like the inversion to partition the deviations from the a-priori fluxes among all the four type of carbon fluxes. NEE and ocean fluxes can, since they are geographically separated, readily be accounted for in statistically independent deviation terms. However, the inversion cannot be expected to distinguish between land biosphere fluxes and fossil fuel emissions, because both are inextricably localized on land, and the CO₂ data alone do not discern fossil and non-fossil carbon (Rödenbeck et al., 2003). Therefore, most inversion studies for surface carbon fluxes focused on the NEE and ocean fluxes, and the fossil fuel and biomass burning were prescribed (e.g., Gurney et al., 2002, 2003; Peters et al., 2007; Nassar et al., 2011; Feng et al., 2009; Monteil et al., 2020). As shown in Table 1, we have reviewed a lot of studies, in which only Deng et al. (2014, 2015) considered the uncertainties of fossil fuel and biomass burning carbon emissions, Liu et al. (2019) and Kang et al. (2012) directly optimized the net carbon flux, and Some studies (Monteil et al., 2020, Scholze et al., 2019) only optimized the NEE. Although Deng et al. (2014)'s state vector includes emissions of CO_2 from fossil fuel combustion, when they reported their posteriori flux estimates, they removed the a priori fossil fuel estimate from the reported total land flux.

As shown in section 2.1, we have added a scheme to simultaneously the fossil fuel and cement production carbon emissions in GCASv2. We have tried to use it to optimize the fossil fuel emissions in China. We tested different emission inventories, but GCASv2 did not make them converge, but only made the emissions of each inventory slightly lower. Therefore, we think that under the current resolution of atmospheric transport model, spatial coverage of observational data, and the assimilation settings, GCASv2 cannot optimize it well.

According to your suggestion, we added a sensitivity test for optimizing fossil fuel carbon emissions, using the same localization scheme as BIO and OCN, giving fossil fuels a global uncertainty of 5%. The results showed that the impact on both the inverted global and regional scale BIO fluxes are very small (Table 2).

The following sentences has been added in the revised manuscript:

"... and the FOSSIL and FIRE carbon emissions are kept intact (*the impact of this assumption* on both the inverted global and regional BIO fluxes are very small (Table S4)). Following Wang et al. (2019), ..." (see Lines 558-560, Pages 16-17)

System Name	Transport model/Res.	Assimilati on method	Obs.	State Vector*	Reference
CT/CTE/CT- China	TM5,global 3x2, region, 1x1	EnSRF	obspack	NEE, OCN	Peters et al., 2007; Peters et al., 2010; Zhang et al., 2014
UoE	GEOS- Chem,4x5	EnKF	in situ or GOSAT	NEE, OCN	Feng et al., 2009, 2016, 2017
CAMS CO2 inversion system	LMDz,3.75x1.8 75	variational	surface observations, GOSAT, OCO- 2	NEE, OCN	Chevallier, et al., 2019
CCDAS	TM3,4x5	4D-Var	in situ CO2, SM, and L- VOD	NEE	Scholze et al., 2019
Jena CarboScope	TM3,4x5	time- independe nt Bayesian inversion	surface observations	NBE, OCN	Rödenbeck, 2005; Rödenbeck et al., 2003
TransCom 3 inversions	16 Atmospheric Transport Models,2.0x2.5 to 7.5x7.5	Bayesian synthesis inversion	GLOBALVIEW data	NEE, OCN	Baker et al., 2006; Gurney et al., 2002, 2003

Table 1. a summary of the inversion methodology in the literature.

Nasser et al., 2011	GEOS- Chem,2x2.5	time- independe nt Bayesian inversion	TES and surface flask measurements	NEE, OCN	Nassar et al., 2011
EUROCOM (include 6 systems)	CHIMERE, FLEXPART, STILT, TM5, NAME/0.5x0.5 ~1x1	Variational , EnKF, MCMC	flask	NEE OCN (4 prescribed)	Monteil et al., 2020
Deng et al., 2007	NIES,2.5x2.5	Time- dependent Bayesian synthesis	GLOBALVIEW data	NEE, OCN	Deng et al., 2007
Niwa et al., 2012	NICAM- TM,~240 km	Time- dependent Bayesian synthesis	GLOBALVIEW , CONTRAIL	NEE, OCN	Niwa et al., 2012
Miyazaki et al., 2011	AGCM,2.8x2.8	LETKF	OSSEs (GOSAT, CONTRAIL, and surface sites)	NEE, OCN	Miyazaki et al., 2011
TM5-4DVAR inversion system	TM5,6x4	4D-Var	GOSAT	NEE, OCN	Basu et al., 2013
GEOS-Chem- 4DVAR inversion system	GEOS- Chem,4x5	4D-Var	GOSAT, Flask	NEE, OCN, FOSSIL, FIRE	Deng et al., 2014; 2016
CMS-Flux inversion framework	GEOS- Chem,4x5	4D-Var	GOSAT, OCO- 2, SIF	NBE, OCN	Liu et al., 2017
LETKF_C	GEOS- Chem,4x5	LETKF	OSSEs (GOSAT, CONTRAIL, and surface sites)	Net flux	Liu et al., 2019; Kang et al., 2012

*NEE: net ecosystem exchange, ecosystem respiration (ER) – gross primary production (GPP); NBE: net biosphere exchange, NEE + biomass burning carbon emission (FIRE); OCN: atmosphere - ocean carbon exchange; FOSSIL: fossil fuel and cement production carbon emission; Net flux: NEE + OCN + FOSSIL+ FIRE

Table 2. Results of sensitivity tests in the year of 2010 (Wfossil is an experiment with the

		Prior	1 week	Wfossil
Super Obs.	Total	-	730	730
Num. per window	Each grid could use	-	4	4
	BIO	-2.07	-4.16	-4.15
Global Flux	OCN	-2.08	-2.33	-2.31
(PgC/yr)	FOSSIL	9.07	9.07	9.05
	AGR	7.25	4.91	4.92
	North America Boreal	-0.29	-0.43	-0.44
	North America Temperate	-0.42	-1.25	-1.21
	Tropical South America	-0.17	-0.26	-0.27
	Temperate South America	-0.24	-0.4	-0.41
Designal Flux	Northern Afirca	0.21	0.32	0.34
Regional Flux	Southern Africa	0.22	-0.3	-0.29
(PgC/yr)	Boreal Asia	-0.4	-0.46	-0.48
	Temperate Asia	-0.3	-0.29	-0.27
	Southeast Asia	-0.29	-0.23	-0.24
	Australia	-0.17	-0.4	-0.4
	Europe	-0.19	-0.41	-0.43
la dense dens	BIAS	1.43	-0.44	-0.43
independent	MAE	1.92	1.37	1.35
evaluation	RMSE	2.36	2.11	2.08
Deviation from	Deviation from the observed AGR (PgC yr ⁻¹)		-0.26	-0.25

FOSSIL carbon emissions being synchronously optimized) (This Table has been added in the revised Supporting Information)

Line 209: How does GCASv2 consider the spatial representativeness errors in the inversion system?

Response: Many thanks for this question. GCASv2 do not consider the spatial representativeness errors for the GOSAT XCO₂ retrievals in this study. Generally, the spatial representation error must be considered when the resolution of the model grid is inconsistent with the spatial range represented by the observation data. In this study, we only use the XCO₂ retrievals. The reason of why we do not consider the spatial representativeness errors is that, first, the XCO₂ retrieval is a column averaged atmospheric CO₂ concentration, which is the result of full atmosphere mixing; 2) before we use the GOSAT data in GCASv2, it has been averaged within the grid cell of $1^{\circ}\times1^{\circ}$. 3) a 'super-observation' approach is adopted based on the optimal estimation theory (Miyazaki et al., 2012). A super-observation is generated by averaging all observations located within the same model grid within a DA window. Therefore, we believe that the spatial representation of the re-grided and averaged XCO₂ is constructed using the GOSAT retrieval error, which has been uniformly inflated by a factor of 1.9 with lowest error fixed as 1 ppm. Therefore, we did not consider the spatial representation error in this study.

Line 238: How many sites are subject to this spurious noise? Are these sites excluded from the inversion system? How much does removing data at these sites influence the inversion fluxes?

Response: We have conducted an additional assimilation for the year of 2010, in which we do not remove the spurious signals, namely all the data with the correlation coefficient with the perturbed fluxes greater than zero were used for assimilation. As shown in Table 3, on average, 87% of the observations were spurious noise and removed in this study. The spurious observations will increase the inverted global land sink and enlarge the deviation of the simulated and observed atmospheric CO_2 growth rate. For different TRANSCOM regions, the impact for the BIO fluxes could be in the range of -32% to 40%. We have added the following sentences in the revised manuscript (see Lines 351-355, Page 12) and added Table 3 in the revised Supporting Information.

"...Otherwise, the relationship is assumed to be spurious noise. On average, 87% of the observations were spurious noise and removed in this study. The spurious observations will increase the inverted global land sink and enlarge the deviation of the simulated and observed AGR. For different TRANSCOM regions, the impact for the BIO fluxes could be in the range of -32% to 40% (Table S4). The scale of 3000 km ..."

		Prior	Posterior	Wnoise
Super Obs.	Total	-	730	730
Num. per window	Each grid could use	-	4	28
Clabel Elver	BIO	-2.07	-4.16	-4.31
(DaC/ur)	OCN	-2.08	-2.33	-2.42
(rgC/yl)	AGR	7.25	4.91	4.67
	North America Boreal	-0.29	-0.43	-0.42
	North America Temperate	-0.42	-1.25	-1.41
	Tropical South America	-0.17	-0.26	-0.3
	Temperate South America	-0.24	-0.4	-0.37
Desite and Floor	Northern Afirca	0.21	0.32	0.28
(DeC/arr)	Southern Africa	0.22	-0.3	-0.42
(PgC/yr)	Boreal Asia	-0.4	-0.46	-0.33
	Temperate Asia	-0.3	-0.29	-0.31
	Southeast Asia	-0.29	-0.23	-0.27
	Australia	-0.17	-0.4	-0.4
	Europe	-0.19	-0.41	-0.28
in domon domé	BIAS	1.43	-0.44	-0.41
independent	MAE	1.92	1.37	1.4
evaluation	RMSE	2.36	2.11	2.2
Deviation from	n the observed AGR (PgC yr ⁻¹)	2.08	-0.26	-0.5

Table 3. Results of sensitivity tests in the year of 2010 (Wnoise is the experiment with spurious signals included)

Technical corrections:

Line 38: "BIAS" is not defined before it is used.

Response: Thanks! We have changed "BIAS" to "bias" in the revised manuscript (see Line 38, Page 2).

Line 63: "However, their carbon uptakes have significant spatial differences and interannual variations." References are needed.

Response: Thanks for this suggestion. We have added three references, namely *Bousquet et al.* (2000), *Takahashi et al.* (2009) and *Piao et al.* (2020). (see Lines 65-66, Page 3)

Line 95: "However, so far, on the one hand, most studies focused on the impact of GOAST XCO 2 retrievals on the inversion of surface carbon fluxes, but in many regions, there are still large divergences for carbon sinks between different inversions with the same GOSAT data or between inversions with GOSAT and in situ observations (Chevallier et al., 2014)". Is only one study considered and cited?

Response: Many thanks for this suggestion. We have added two references in the revised manuscript, i.e., Wang et al. (2018) and Feng et al. (2016). The sentence has been revised as follows (see Line 102, page 4 in the revised manuscript):

"...between inversions with GOSAT and in situ observations (e.g., Chevallier et al., 2014; Feng et al., 2016; Wang et al., 2018), on the other hand, ..."

Line 102. References are needed.

Response: Thank you! We have added two references, namely Feng et al. (2017) and Byrne et al., (2019). See Line 106, Page 4 in the revised manuscript.

Line 255: The references for the two emission inventories of FOSSIL and FIRE are out of date. ODIAC and GFEDv4 have been updated recently.

Response: We have revised the reference of ODIAC "Oda and Maksyutov (2011)" as "*Oda et al. (2018)*", and the references of GFEDv4 "van der Werf et al. (2010) and Giglio et al. (2013)" as "*Randerson et al., 2017*" (see Lines 377 and 379, Page 13)

Line 270: "The BIO carbon flux, which is the most important prior carbon flux". Why is the prior carbon flux of BIO more important than FOSSIL and FIRE to an inversion system? Response: This statement is problematic. From the perspective of the carbon cycle, the carbon flux of terrestrial ecosystems is not more important than others. In fact, what we want to express is that because the carbon flux of terrestrial ecosystems has the greatest uncertainty and the most significant interannual variation, when using observational data to optimize surface carbon flux, the carbon flux of terrestrial ecosystems is the most concerned. We have modified that sentence to "*The BIO carbon flux, which is one of the most concerned prior carbon fluxes in an assimilation system*" in the revised manuscript. (see Line 389, Page 13)

Line 340: When the averages of the modeled and the observational values/retrievals are equal, BIAS is zero, even if all data are distant to the 1:1 line in the comparison. BIAS cannot

effectively evaluate the performance of the model by showing how much the modeled values/retrievals agree with the observational values/retrievals. The average of absolute difference between the modeled and the observational values/retrievals is needed. Response: Thank you! We have added the mean absolute error (MAE) between the modeled and the observational values/retrievals in the revised manuscript. (see Line 532, Page 15; Lines 577-579, Page 17; Lines 599-601, Page 18; and Lines 614 – 616, Page 19)

Line 360: Does the study of Wang et al. (2019) account for the uncertainty in FOSSIL and FIRE?

Response: No, Wang et al. (2019) only optimized the terrestrial ecosystem and ocean carbon fluxes.

Line 448: What is "impact of accumulation"?

Response: As shown in the following figure (Figure 1), we find that there is a significant increasing trend for the annual BIAS between the simulated CO_2 concentration with the posterior flux and the observed concentration. We believe that this increasing trend is due to the accumulation of errors in the assimilation system, which may be caused by the slight overestimates of land sink in each year.





Figures 3/4: "Biases" in the caption is easily confused with "BIAS" defined in equation 10. Response: Thank you! We have modified the "Biases" in the caption Figures 3/4 to "BIAS". (see Line 595, page 18 and Line 605, page 19)

Table 1: BIAS cannot evaluate the performance of the model by showing how much the modeled values/retrievals agree with the observed values/retrievals. Response: Thank you for this suggestion! According to this suggestion, we have added the mean absolute error (MAE) in Table 1 in the revised manuscript. (see Line 532, Page 15; Lines 577-579, Page 17; Lines 599-601, Page 18; and Lines 614 – 616, Page 19)

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3	retrievals using a new version of Global Carbon Assimilation System
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5	Fei Jiang ^{1,7*} , Hengmao Wang ¹ , Jing M. Chen ² , Weimin Ju ¹ , Xiangjun Tian ³ , Shuzhang
6	Feng ¹ , Guicai Li ⁴ , Zhuoqi Chen ⁵ , Shupeng Zhang ⁵ , Xuehe Lu ¹ , Jane Liu ^{2,6} , Haikun
7	Wang ⁶ , Jun Wang ¹ , Wei He ¹ , Mousong Wu ¹
8	
9	
10	1 Jiangsu Provincial Key Laboratory of Geographic Information Science and Technology,
11	International Institute for Earth System Science, Nanjing University, Nanjing, 210023, China
12	2 Department of Geography and Planning, University of Toronto, Toronto, Ontario M5S3G3,
13	Canada
14	3 The Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, 100029, China
15	4 National Satellite Meteorological Center, China Meteorological Administration, Beijing
16	100101, China
17	5 College of Global Change and Earth System Science, Beijing Normal University, Beijing,
18	100875, China
19	6 School of Atmospheric Sciences, Nanjing University, Nanjing, 210023, China
20	7 Jiangsu Center for Collaborative Innovation in Geographical Information Resource
21	Development and Application, Nanjing, 210023, China
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* Corresponding author: Tel.: +86-25-83597077; Fax: +86-25-83592288; E-mail address: jiangf@nju.edu.cn 1

27 Abstract

28 Satellite XCO2 retrievals could help to improve carbon flux estimation because of their good spatial coverage. In this study, to assimilate the GOSAT XCO₂ retrievals, 29 the Global Carbon Assimilation System (GCAS) is upgraded with new assimilation 30 algorithms, procedures and a localization scheme, a higher assimilation parameter 31 resolution and so on, and hence is named as GCASv2. Based on this new system, the 32 33 global terrestrial ecosystem (BIO) and ocean (OCN) carbon fluxes from May 1, 2009 to Dec 31, 2015 are constrained using the GOSAT ACOS XCO₂ retrievals (Version 7.3). 34 35 The posterior carbon fluxes from 2010 to 2015 are independently evaluated using CO₂ observations from 52 surface flask sites. The results show that the posterior carbon 36 fluxes could significantly improve the modeling of atmospheric CO₂ concentrations, 37 38 with global mean <u>bias</u> decreases from a prior value of 1.6 ± 1.8 ppm to -0.5 ± 1.8 ppm. 39 The uncertainty reduction (UR) of the global BIO flux is 17%, and the highest monthly regional UR could reach 51%. Globally, the mean annual BIO and OCN carbon sinks 40 41 and their interannual variations inferred in this study are very close to the estimates of 42 CT2017 during the study period, and the inferred mean atmospheric CO₂ growth rate and its interannual changes are also very close to the observations. Regionally, over the 43 44 northern lands, there are the strongest carbon sinks in North America Temperate, followed by Europe, Boreal Asia, and Temperate Asia; and in tropics, there are strong 45 sinks in Tropical South America and Tropical Asia, but a very weak sink in Africa. This 46 pattern is significantly different from the estimates of CT2017, but the estimated carbon 47 sinks in each continent and some key regions like Boreal Asia and Amazon are 48 comparable or in the range of previous bottom-up estimates. The inversion also changes 49 50 the interannual variations of carbon fluxes in most TRANSCOM land regions, which have a better relationship with the changes of severe drought area or LAI, or are more 51 consistent with previous estimates for the impact of drought. These results suggest that 52 the GCASv2 system works well with the GOSAT XCO2 retrievals, and has a good 53 performance in estimating the surface carbon fluxes, meanwhile, our results also 54 indicate that the GOSAT XCO2 retrievals could help to better understand the 55

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57 interannual variations of regional carbon fluxes.

58 1. Introduction

Atmospheric carbon dioxide (CO₂) is one of the most important greenhouse gases, 59 60 and fossil fuel burning and land use change are mostly responsible for its increase from 61 the preindustrial concentration. Terrestrial ecosystems and oceans play very important roles in regulating the atmospheric CO₂ concentration. In the past half century, about 62 60% of the anthropogenic CO₂ emissions have been absorbed by the terrestrial 63 ecosystems and oceans (IPCC, 2014). However, their carbon uptakes have significant 64 65 spatial differences and inter-annual variations (Bousquet et al., 2000; Takahashi et al., 2009; Piao et al., 2020). Therefore, it is very important to quantify the global and 66 67 regional carbon fluxes.

Atmospheric inversion is an effective method for estimating the surface CO₂ fluxes 68 69 using the globally distributed atmospheric CO2 concentration observations (Enting and Newsam, 1990; Gurney et al., 2002). It has robust performance on global or hemisphere 70 scale carbon budget estimates (Houweling et al., 2015), but on regional scales, due to 71 72 the uneven distribution of in situ observations, the reliability of inversion results varies greatly in different regions. Generally, the inversions have large uncertainties in tropics, 73 74 southern hemisphere oceans and most continental interiors such as South America, Africa, and Boreal Asia (Peylin el al., 2013). Satellite observation has a better spatial 75 76 coverage, especially over remote regions, and studies show that it can be used to improve the carbon flux estimates (e.g., Chevallier et al., 2007; Miller et al., 2007; 77 78 Hungershoefer et al., 2010). The Greenhouse Gases Observing Satellite (GOSAT) (Kuze et al., 2009), being the first satellite mission dedicated to observing CO₂ from 79 space, was launched in 2009. Many inversions have utilized the GOSAT retrievals for 80 column-averaged dry air mole fractions of CO₂ (XCO₂) to infer surface carbon fluxes 81 (e.g., Basu et al., 2013; Maksyutov et al., 2013; Saeki et al., 2013a; Chevallier et al., 82 83 2014; Deng et al., 2014; Deng et al, 2016; Wang et al., 2018a; Wang et al., 2019). Takagi et al. (2011) found that GOSAT XCO2 retrievals could significantly reduce the 84 uncertainties in estimates of surface CO2 fluxes for regions in Africa, South America, 85

and Asia, where the sparsity of the surface monitoring sites is most evident. Basu et al. 86 87 (2013) shown that assimilating only GOSAT data can well reproduce the observed CO2 time series at the surface and TCCON sites in the tropics and the northern extra-tropics, 88 but enhance seasonal cycle amplitudes in the southern extra-tropics. Wang et al. (2019) 89 also showed that GOSAT XCO2 retrievals can effectively improve carbon flux 90 estimation, and the performance of the inversion with GOSAT data only was 91 92 comparable with the one using in situ observations. Meanwhile, based on the inversions with GOSAT XCO2 retrievals, Liu et al. (2018) quantified the impacts of the 2011 and 93 2012 droughts on terrestrial ecosystem carbon uptake anomalies over the contiguous 94 US, Deng et al. (2016) compared the distributions of drought and posterior carbon 95 fluxes in South America for 2010-2012, Detmers et al. (2015) studied the impact of the 96 strong La Niña episode on the carbon fluxes in Australia from the end of 2010 to early 97 98 2012. However, so far, on the one hand, most studies focused on the impact of GOAST XCO₂ retrievals on the inversion of surface carbon fluxes, but in many regions, there 99 are still large divergences for carbon sinks between different inversions with the same 100 101 GOSAT data or between inversions with GOSAT and in situ observations (e.g., 102 Chevallier et al., 2014; Feng et al., 2016; Wang et al., 2018a), on the other hand, 103 although some studies reported the impact of drought or extreme wetness on the 104 changes of carbon fluxes using inversions based on GOSAT, few studies have comprehensively investigated the impacts of GOSAT data on the interannual variations 105 106 of inverted land sinks in different regions (Feng et al., 2017; Byrne et al., 2019).

In this study, we present a 6-year inversion from 2010 to 2015 for the global and 107 108 regional carbon fluxes using only the GOSAT XCO2 retrievals. The Global Carbon Assimilation System (GCAS) is employed to conduct this inversion, which was 109 110 developed in China in 2015 (Zhang et al., 2015) and updated in this study with a new scheme to assimilate XCO2 retrievals. The inverted multi-year averaged carbon fluxes 111 112 for the globe, global land and ocean, each TRANSCOM region (Gurney et al., 2002) as well as some key areas are shown and compared with previous top-down and bottom-113 up (Jiang et al., 2016) estimates. The estimated interannual variations of carbon fluxes 114

in each TRANSCOM region are given and discussed against changes in drought and
LAI. This manuscript is organized as follows: Section 2 details the GCASv2 system as
well as the prior fluxes, GOSAT retrievals and uncertainty settings. Section 3 briefly

118 introduces the experimental design. Results and discussions are presented in Section 4,

119 and Conclusions are given in Section 5.

120 2. Method and Data

121 2.1 A new version of the Global Carbon Assimilation System (GCASv2)

Figure 1 shows the flow chart of the GCASv2 system. In each data assimilation 122 123 (DA) window, there are two steps. The first step, the prior fluxes of X^{b} in each grid 124 are independently perturbed with a Gaussian random distribution, and put into the 125 global atmospheric chemical transport model MOZART-4 to simulate CO2 concentrations, which are then sampled according to the locations and times of CO2 126 127 observations. The sampled data are used in the assimilation module together with the CO_2 observations to generate the optimized fluxes of X^a . In the second step, the 128 MOZART-4 model is run again using the optimized fluxes of X^a , to generate new CO₂ 129 130 concentrations for the initial field of the next DA window. By this method, if the flux in one window is overestimated because of some reasons, it will affect the 131 132 concentrations of the next window, thereby the posterior flux of the next window will 133 compensate the overestimation. This DA flow chart is different from the previous 134 version of GCAS (GCASv1), which runs the MOZART-4 model only once, and 135 optimizes the fluxes and the initial field of the next window synchronously, namely in 136 each window, there is relatively perfect initial field (directly optimized using observations), the inversions of each window are independent, and the amount of 137 138 overestimation or underestimation in one window will continue to accumulate until the end, leading to an overall overestimation or underestimation. In addition, due to the 139 140 relatively perfect initial field, the differences between the simulated and observed 141 concentrations are only contributed by the errors in the prior fluxes of current window, 142 resulting in a relatively smaller model - data mismatch, so as to weaken the assimilation 143 benefits on fluxes.

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Figure 1. Flow chart of the GCASv2 system

149 The perturbation of X^b represents the uncertainty of the prior carbon flux, which 150 is calculated using the following function.

151

$$\boldsymbol{X}_{i}^{b} = \boldsymbol{X}_{0}^{b} + \lambda \times \boldsymbol{\delta}_{i} \times \boldsymbol{X}_{0}^{b} , i = 1, 2, \dots, N$$
(1)

where δ_i represents random perturbation samples, which is drawn from Gaussian 152 153 distributions with mean zero and standard deviation of one. N is the ensemble size. λ is 154 a set of scaling factors, which represents the uncertainty of each prior flux. In GCASv1, 155 λ is defined in different land and ocean areas based on 22 TRANSCOM regions 156 (Gurney et al., 2002) and 19 Olson ecosystem types, as in CarbonTracker (CT, Peters 157 et al., 2007). This means that in the same area, the error of a prior flux is the same. Through assimilation, the flux will be integrally enlarged or reduced. In GCASv2, we 158 change to use a λ in each grid, meaning that for each grid, the perturbations of prior 159 fluxes are independent. In addition, the grid cell of λ is different from those of the prior 160 flux and the transport model, which could be set freely. X_0^b is prior carbon flux. 161 162 Generally, there are 4 types of carbon fluxes, namely terrestrial ecosystem (BIO) carbon 163 flux (i.e., net ecosystem exchange (NEE) = ecosystem respiration (ER) - gross primary 6

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166	production (GPP)), atmosphere and ocean (OCN) carbon exchange, fossil fuel_and		删除了: carbon flux
167	cement production (FOSSIL) carbon emission and biomass burning (FIRE) carbon		
168	emission, which are used to drive the transport model to simulate the atmospheric CO_2		
169	concentration. And in general, FOSSIL and FIRE fluxes are assumed to have no errors,		
170	only BIO and OCN fluxes are optimized in an assimilation system (e.g., Gurney et al.,		
171	2002; Peters, et al., 2007; Nassar et al., 2011; Jiang et al., 2013; Chevallier, et al., 2019).	_	删除了: Wang et al., 2019
172	In GCASv1, only the BIO flux was treated as state vector and optimized, the OCN flux		
173	was directly from the output of CarbonTraker (CT). In GCASv2, it is set to be an		
174	optional item. Four schemes are set (Functions 2 - 5). The first one is the same as the		
175	previous version, only the BIO flux is optimized; the second one is the same as general,		
176	namely both BIO and OCN fluxes are state vectors; the third one is that BIO, OCN and	_	删除了: optimized
177	FOSSIL fluxes are optimized at the same time; and the fourth one is that only net flux		
178	is optimized. In this study, the second scheme was selected.		
179	$X_{i}^{b} = (X_{bio}^{b} + \lambda_{bio} \times \delta_{i,bio} \times X_{bio}^{b}) + X_{ocn}^{b} + X_{fossil}^{b} + X_{fire}^{b}, i = 1, 2, \dots, N $ (2)		
180	$X_{i}^{b} = \left(X_{bio}^{b} + \lambda_{bio} \times \delta_{i,bio} \times X_{bio}^{b}\right) + \left(X_{ocn}^{b} + \lambda_{ocn} \times \delta_{i,ocn} \times X_{ocn}^{b}\right)$		
181	$+X_{fossil}^{b}+X_{fire}^{b}, i = 1, 2,, N$ (3)		
182	$X_{i}^{b} = \left(X_{bio}^{b} + \lambda_{bio} \times \delta_{i,bio} \times X_{bio}^{b}\right) + \left(X_{ocn}^{b} + \lambda_{ocn} \times \delta_{i,ocn} \times X_{ocn}^{b}\right)$		
183	+ $(X^{b}_{fossil} + \lambda_{fossil} \times \delta_{i,fossil} \times X^{b}_{fossil}) + X^{b}_{fire}, i = 1, 2,, N$ (4)		
184	$X^b_i = \left(X^b_{bio} + X^b_{ocn} + X^b_{fossil} + X^b_{fire}\right) + \lambda_{netflux} \times \delta_{i,netflux} \times (X^b_{bio} + X^b_{bio}) + \lambda_{netflux} \times (X^b_{bio} + X^b_{bio}) + \lambda_{n$		
185	$X_{ocn}^{b} + X_{fossil}^{b} + X_{fire}^{b}$), i = 1, 2,, N (5)		
186	For the CO ₂ observations, in GCASv1, only the flask and in situ observations were		
187	assimilated. In GCASv2, we added a module to use satellite XCO ₂ retrievals. With this		
188	module, simulated CO ₂ concentration profiles are converted to XCO ₂ concentrations,		移动了(插入) [2]
189	and users can choose to assimilate flask/in situ observations or satellite XCO ₂ retrievals	$\left\langle \right\rangle$	删除了: For the modeled XCO ₂ , the
190	alone, or simultaneously assimilate these two data. The simulated CO ₂ concentration		设置了格式: 下标 设置了格式: 下标
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195 profiles are mapped into the satellite retrieval levels and then vertically integrated based 196 on satellite averaging kernel according to the following equation (Connor, et al., 2008). $XCO_{2}^{m} = XCO_{2}^{a} + \sum_{i} h_{i}a_{i}(A(x) - y_{a,i})$ (6) 197 where *j* denotes the retrieval level; *x* is the simulated CO_2 profile, and A(x) is a 198 199 mapping matrix; XCO_2^a is the prior XCO_2 ; h_i is a pressure weighting function, a_i and y_a are the satellite column averaging kernel and the prior CO₂ profile for retrieval, 200 201 respectively. 202 To reduce the computational cost and the influence of representative errors, a 'super-observation' approach is also adopted in GCASv2 based on the optimal 203 estimation theory (Miyazaki et al., 2012). A super-observation is generated by 204 205 averaging all observations located within the same model grid within a DA window. 206 We assume that the observation errors of different stations at different times are independent of each other. The standard deviation of the *j*th observation y_j is r_j . The 207 208 super-observation ynew, standard deviation rnew and corresponding simulations $x_{new,i}$ from one perturbed prior flux X_i^b are calculated: 209 $\underline{\qquad \qquad }^{1}/r_{new}^{2} = \sum_{j=1}^{m} \frac{1}{r_{j}^{2}}$ (7) 210 $\underline{y_{new}} = \sum_{j=1}^{m} w_j y_j / \sum_{j=1}^{m} w_j \underline{y_j}$ 211 (8) $\underline{x_{new,i}} = \sum_{j=1}^{m} w_j \, x_{j,i} / \sum_{j=1}^{m} w_j \tag{9}$ 212 where $w_j = \frac{1}{r_i^2}$ is the weighting factor; *m* is the number of observations within a 213 214 super-observation grid. The super-observation error decreases as the number of 215 observations used for the super-observation increases. 216

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223 Besides the Local Ensemble Transform Kalman Filter (LETKF), which has been 224 implemented in GCASv1, to avoid storing and inverting very large matrices during analysis, in GCASv2, we added another assimilation algorithm, namely the Ensemble 225 226 square root filter (EnSRF) algorithm (Whitaker and Hamill, 2002), which has been successfully used in CT (Peters et al., 2005). EnSRF obviates the need to perturb the 227 observations in contrast to the traditional EnKF algorithm and assimilates observations 228 229 in a sequential way. It has a better performance than the method to assimilate observations simultaneously as long as the observation errors are uncorrelated 230 231 (Houtekamer and Mitchell, 2001). The implementation process and setup are detailed 232 below.

After obtaining an ensemble of state vectors as described in Section 2.1, ensemble runs of MOZART-4 are conducted to propagate these errors in the model with each ensemble sample of a state vector. The background error covariance P^b is calculated based on the forecast ensemble from Eq. (2):

$$\boldsymbol{P}^{\boldsymbol{b}} = \frac{1}{n-1} \sum_{i=1}^{n} (\boldsymbol{X}_{i}^{\boldsymbol{b}} - \overline{\boldsymbol{X}}^{\boldsymbol{b}}) (\boldsymbol{X}_{i}^{\boldsymbol{b}} - \overline{\boldsymbol{X}}^{\boldsymbol{b}})^{T}$$

where \overline{X}^{b} represents the mean of the ensemble samples. Based on the background error covariance, the response of the uncertainty in the simulated concentrations to the uncertainty in emissions is obtained. Combing observational vector y, the state vector is updated according to the following formulations:

$$\overline{X^a} = \overline{X^b} + \mathbf{K}(\mathbf{v} - H\overline{X^b})$$

$$\mathbf{K} = \mathbf{P}^{\mathbf{b}}\mathbf{H}^{\mathrm{T}}(\mathbf{H}\mathbf{P}^{\mathbf{b}}\mathbf{H}^{\mathrm{T}} + \mathbf{R})^{-1}$$

$$\delta X_i^a = \delta X_i^b - \widetilde{K} H \delta X_i^b$$

²⁴⁵ While employing sequential assimilation and independent observations

246
$$\widetilde{K} = (1 + \sqrt{R/HP^bH^T + R})^{-1}K$$

237

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247 where H is the observation operator that maps the state variable from model space to 9 删除了: introduced by Whitaker and Hamill (2002),

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observation space. K is the Kalman gain matrix of ensemble mean depending on background and observation error covariance R, representing the relative contributions to analysis. \tilde{K} is the Kalman gain matrix of ensemble perturbation, and then emission perturbations after inversion δX_i^a can be calculated. At the analysis step, the ensemble mean $\overline{X^a}$ is taken as the best estimate of the carbon flux,

261 2.1.2 Atmospheric transport model

262 Same as the GCASv1 (Zhang et al., 2015), the global chemical transport Model for 263 OZone And Related chemical Tracers (MOZART-4; Emmons et al., 2010) is adopted as the atmospheric transport model in GCASv2. MOZART-4 is a flexible model, it can 264 be run at essentially any resolution, and can be driven by essentially any meteorological 265 data set and with any emission inventories (Emmons et al., 2010). In this system, we 266 267 preset two horizontal resolutions for MOZART runs, one being approximately 268 $2.8^{\circ} \times 2.8^{\circ}$, with <u>transport model grids</u> of 128×64 , and another being approximately $1.0^{\circ} \times 1.0^{\circ}$, with the model grids of 360×180 . In the vertical direction, we use 28 layers. 269 270 The ERA-Interim reanalysis datasets from the European Centre for Medium-Range Weather Forecasts (ECMWF) are used to drive the model. ERA-Interim data set 271 272 includes as many as 128 meteorological variables, and has the highest spatial resolution 273 of approximately 80 km (T255 spectral) on 60 vertical levels from the surface up to 0.1 hPa. Only the variables required for MOZART-4 with a spatial resolution of $1.0^{\circ} \times 1.0^{\circ}$, 274 and 28 vertical levels for 3-D variables from the surface to approximately 2.5 hPa are 275 selected in this system. The selected variables and vertical levels are shown in Table S1 276 and S2 in the supporting information. 277

278 2.1.3 DA window and localization

The DA window is set to one week in GCASv2, which is the same as before. Theoretically, a longer DA window is better, because CO_2 is a stable species. The longer window, the farther CO_2 will be transported. In this way, more observation stations will sense the flux change of one area, and thus more observations can be used to optimize the flux of that place. Therefore, many previous ensemble-based assimilation systems

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To reduce the computational cost and the influence of representative errors, a 'super-observation' approach is adopted based on the optimal estimation theory (Miyazaki et al., 2012). A super-observation is generated by averaging all observations located within the same model grid within a DA window. We assume that the observation errors of different stations at different times are independent of each other. The standard deviation of the *j*th observation y_j is r_j . The super-observation y_{new} , standard deviation r_{new} and corresponding simulations $x_{new,i}$ from one perturbed prior flux X_i^b are calculated:

$$\frac{1}{r_{new}^2} = \sum_{j=1}^m \frac{1}{r_j^2} \quad (7)$$

$$y_{new} = \sum_{j=1}^m w_j \, y_j / \sum_{j=1}^m w_j \quad (8)$$

 $x_{new,i} = \sum_{j=1}^{m} w_j \, x_{j,i} / \sum_{j=1}^{m} w_j \quad (9)$

where $w_j = \frac{1}{r_j^2}$ is the weighting factor; *m* is the number of

observations within a super-observation grid. The superobservation error decreases as the number of observations used for the super-observation increases.

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304 used a longer DA window (e.g., Peters et al. 2005, Feng et al. 2009, Jacobson et al. 305 2020). However, the farther away, the weaker signal the stations can sense. Bruhwiler et al. (2005) clearly shown that a pulse traveling from a faraway place would contribute 306 307 relatively little signal compared to recent pulses from nearby source regions. In addition, 308 Limited by the method of EnKF, this weak signal will be masked by the method's own 309 unphysical signal (spurious correlation), and in order to reduce this influence, we must 310 increase the ensembles, thereby greatly increasing the computational cost. Miyazaki et 311 al. (2011) tested the differences of 3 days and 7 days DA windows, and pointed that 312 with a longer DA window, more observation data will be available to constrain the 313 surface flux, but a longer window can make the effect of model error more obvious. 314 Thus, the assimilation result can be improved as long as the observations with spurious 315 correlations can be neglected. However, spurious correlations can be more serious with 316 increases in the DA window, because of a limited number of ensembles. As a result, a longer window is not necessarily better than a shorter window system. To avoid the 317 318 influence of spurious signals, Kang et al. (2012) used a very short DA window (6 hours) 319 in their assimilation system (LETKF_C) and pointed out that the flux inversion with a long window (3 weeks) is not as accurate as the one obtained with a 6 h DA window, 320 321 particularly in smaller-scale structures. During the development of GCASv1, Zhang et 322 al. (2015) tested different DA windows and found that the longer the window, the larger optimized terrestrial carbon sink will be, resulting in a smaller optimized annual 323 324 atmospheric CO2 growth rate (AGR) as compared to the observed rate. Considering the 325 fact that at present, due to the release of satellite XCO2 retrievals like GOSAT and OCO-326 2, the atmospheric CO₂ observations and coverages have increased significantly 327 compared to before, which means that we do not need to extend the DA window to 328 include more observation data now. Figure S2 shows the mean super observation (see section 2.1.1, only GOSAT XCO₂) numbers during the study period that each grid 329 $(3^{\circ}\times3^{\circ})$ could have within a 1-week DA window and a localization scale (3000 km, see 330 the next paragraph). In most land areas and pan-tropical waters, each grid can already 331 have more than 3 super observations. On average, each grid over the land could has 4 332

333 super observations. Two sensitivity tests in 2010 were conducted in this study using 2-334 and 4-weeks DA windows but a same localization scale, the results are shown in Table S4. When the length of DA window increases from 1 week to 4 weeks, the mean super 335 observation number increases from 4 to 9, accordingly, the inverted global BIO flux 336 increased from -4.16 PgC yr⁻¹ to -4.49 PgC yr⁻¹, resulting in a larger deviation of the 337 simulated and observed AGR and larger simulation error against the surface 338 339 observations. Therefore, we still use the 1-week DA window in GCASv2, 340 As discussed before, in the EnKF method, there are inevitably spurious correlations.

Therefore, a localization scale, which determines that only measurements located 341 within a certain distance (cutoff radius) from a grid point will influence the analysis of 342 this grid, must be set to reduce the effect of spurious correlations. The localization 343 technique in this study is based on both the distance between one site and one grid cell 344 345 of λ , and the linear correlation coefficient between the simulated concentrations and the perturbed fluxes for each parameter (λ)/observation pair. If the distance is less than 500 346 km and the correlation coefficient is greater than zero, the observations will be accepted 347 for assimilation, and if the distance is greater than/equal to 500 km and less than 3000 348 km and the relationship between a parameter deviation and its modeled observational 349 350 impact is statistically significant (p < 0.05), then that relationship is retained. Otherwise, 351 the relationship is assumed to be spurious noise. On average, 87% of the observations were spurious noise and removed in this study. The spurious observations will increase 352 the inverted global land sink and enlarge the deviation of the simulated and observed 353 354 AGR. For different TRANSCOM regions, the impact for the inverted BIO fluxes could 355 be in the range of -32% to 40% (Table S4). The scale of 3000 km is set simply according 356 to the globally-averaged 80-m wind speed during the day (4.96 m/s, Archer and Jacobson, 2005) and the length of DA window (1 week). 357

358 2.2 Prior carbon fluxes

- 359 360
- As described in Section 2.1, there are 4 types of prior carbon fluxes in GCASv2. In this study, FOSSIL carbon emissions are obtained from NOAA's CT, version 2017
- 361 (<u>CT2017</u>, Peters et al. 2007, with updates documented at http://carbontracker.noaa.gov),

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In addition, Zhang et al. (2015) tested different DA window lengths and found that the longer the window, the larger optimized terrestrial carbon sink will be, resulting in a smaller optimized annual atmospheric CO₂ growth rate as compared to the observed rate. Therefore, they pointed out that the 1week DA window seems to be most suitable. For this reason, this study also uses the same DA window of one week as before. ...

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375 which is an average of the Carbon Dioxide Information Analysis Center (CDIAC) 376 product (Andres et al., 2011) and the Open-source Data Inventory of Anthropogenic 377 CO₂ (ODIAC) emission product (Oda et al., 2018). The FIRE CO₂ emissions are also taken from CT2017, which are the average of the Global Fire Emissions Database 378 379 version 4.1 (GFEDv4) (Randerson et al., 2017) and the Global Fire Emission Database from the NASA Carbon Monitoring System (GFED_CMS). The OCN CO2 exchange 380 381 is from the pCO₂-Clim prior of CT2017, which is derived from the Takahashi et al. 382 (2009) climatology of seawater pCO2. In addition, as shown in Figure 7 of the 383 CarbonTracker Documentation CT2017 release (https://www.esrl.noaa.gov/gmd/ccgg/ carbontracker/CT2017/, accessed on 4 Mar, 2020), there are no data in many seas like 384 Japan Sea, Mediterranean, Gulf of Mexico, East China Sea, and so on, and therefore, 385 the fluxes in 2009 modeled using the global ocean circulation (OPA) and the 386 387 biogeochemistry model (PISCES-T) (Buitenhuis et al., 2006; Jiang et al., 2013) is used 388 to fill the no data areas.

389 The BIO carbon flux, which is one of the most concerned prior carbon fluxes in an assimilation system, was simulated using the Boreal Ecosystems Productivity 390 Simulator (BEPS) model (Chen et al., 1999; Ju et al., 2006) in this study. BEPS is a 391 392 process-based, remote sensing data driven, and mechanistic ecosystem model. In this study, BEPS model was run starting from 2000. To simplify the initialization, the initial 393 values of the different carbon pools are from a previous BEPS simulation (Chen et al., 394 2019). In short, all carbon pools were assumed to be in a state of dynamic equilibrium 395 from 1901 to 1910. And all carbon pools were determined by solving a set of equations 396 describing the dynamics of carbon pools (Chen et al., 2003). Then the simulation 397 398 forwarded using historical data. Due to the lack of historical data of remote sensed LAI 399 data, the averaged LAI from 1982 to 1986 represented that over the 1901-1981 period. Then, all our initial carbon pools were set to states of carbon pools in 2000 according 400 to Chen et al. (2019). The BEPS model was also driven by the 1°×1° ERA-Interim 401 reanalysis datasets, including relative humidity, wind speed, air temperature, incoming 402 solar radiation, and total precipitation. The other data include LAI data and clumping 403

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index. LAI was inverted from surface reflectance datasets of Moderate Resolution
Imaging Spectroradiometer (MODIS) (Liu et al., 2012), and the clumping index was
derived from the MODIS Bidirectional Reflectance Distribution Function (BRDF)
products, which provided the finest pseudo multi-angular data for the land surface,
according to Normalized Difference between Hotspot and Darkspot (NDHD) (Chen et
al., 2005, He et al., 2012).

413 2.3 GOSAT XCO2 retrievals

414 The GOSAT XCO₂ retrievals of the ACOS Version 7.3 Level 2 Lite product (O'Dell et al., 2012; Crisp et al., 2012) at the pixel level during May 2009 ~ Dec 2015 415 is used in this study, which is bias-corrected (Wunch et al., 2011). In order to achieve 416 417 the most extensive spatial coverage with the assurance of using best quality data available, before being used in the inversion system, the XCO2 retrievals are filtered 418 with two parameters of warn levels and xco2 quality flag, which are provided along 419 420 with the product. Only the data with xco2 quality flag greater than 0 are selected. The selected data are then divided into three groups according the value of warn levels, that 421 are with warn levels less than 8, greater than 9 and less than 12, and greater than 13, 422 respectively. The group with smallest warn levels has the best data quality, while that 423 with the largest is the worst. Then, the pixel data are averaged within the grid cell of 424 425 1°×1°, and in each grid, only the group with best data quality is selected and then 426 averaged. The other variables like column-averaging kernel, retrieval error and so on which are provided along with the XCO₂ product are also dealt with the same method. 427 This process is the same as Wang et al. (2019). Except the XCO2, the other quantities, 428 429 provided along with the ACOS product were also filtered and averaged to 1°×1° grid 430 according to the above method.

431 2.4 Evaluation data and method

Generally, direct validation of the optimized flux is impossible, and instead, we
indirectly evaluate the posterior flux by comparing the forward simulated atmospheric
CO₂ mixing ratios against measurements (e.g., Jin et al., 2018; Wang et al., 2019; Feng

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上移了 [2]: For the modeled XCO₂, the simulated CO₂ concentration profile should be first mapped into the satellite retrieval levels and then vertically integrated according to the following equation.

 $XCO_2^m = XCO_2^a + \sum_j h_j a_j (A(x) - y_{a,j})$ (10)

where *j* denotes the retrieval level; *x* is the simulated CO₂ profile, and A(x) is a mapping matrix; XCO₂^a is the prior XCO₂; h_j is a pressure weighting function, a_j and y_a are the satellite column averaging kernel and the prior CO₂ profile for retrieval, respectively. Except the simulated CO₂ profile,

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concentration profile should be first mapped into the satellite retrieval levels and then vertically integrated according to the following equation.

 $XCO_{2}^{m} = XCO_{2}^{a} + \sum_{j} h_{j}a_{j}(A(x) - y_{a,j})$ (10)

where *j* denotes the retrieval level; *x* is the simulated CO₂ profile, and A(x) is a mapping matrix; XCO₂^a is the prior XCO₂; *h_j* is a pressure weighting function, *a_j* and *y_a* are the satellite column averaging kernel and the prior CO₂ profile for retrieval, respectively. Except the simulated CO₂ profile, the other quantities are provided along with the ACOS product and filtered and averaged to 1°×1° grid according to the above method.

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上移了 [1]: Except the simulated CO₂ profile, the other quantities are provided along with the ACOS product and filtered and averaged to 1°×1° grid according to the above

et al., 2020). First, the simulated XCO2 are compared against the corresponding GOSAT 511 512 XCO2 retrievals to test the effectiveness of the assimilation system (see Section 2.3 for the description of the GOSAT XCO2 retrieval). Second, Surface CO2 observations used 513 independent evaluations in this study are obtained 514 for from the obspack co2 1 GLOBALVIEWplus v5.0 2019-08-12 product. It is a subset of the 515 Observation Package (ObsPack) Data Product (ObsPack, 2019), and contains a 516 517 collection of discrete and quasi-continuous measurements at surface, tower and ship 518 sites contributed by national and universities laboratories around the world. In this study, 519 surface CO2 measurements from 52 flask sites are selected to evaluate the posterior CO2 concentrations, which are all provided by the NOAA Global Monitoring Laboratory 520 (with lab number of 1 in each filename). The locations of the 52 sites could be found in 521 522 Figure 2 and the corresponding sites code as well as the information latitude and 523 longitude are listed in Table S3 in the Supporting Information.

524 During the evaluation, <u>4 basic statistical measures</u>, namely mean bias (BIAS), 525 mean absolute error (MAE), root mean square error (RMSE), and correlation 526 coefficient (CORR), are calculated against the surface CO₂ observations and GOSAT 527 XCO₂ retrievals, respectively. The BIAS, <u>MAE</u>, RMSE, and CORR reflect the overall 528 model tendency, both the model bias and error variance, and the linear correspondence 529 between the modeled and observational values/retrievals, respectively. The functions of 530 these <u>4</u> basic statistical measures are expressed as:

$$BIAS = \frac{1}{M} \sum_{j=1}^{M} (x_j - y_j) = \bar{y} - \bar{x}$$

$$MAE = \frac{1}{M} \sum_{j=1}^{M} |x_j - y_j|$$
(16)

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$$RMSE = \sqrt{\frac{1}{M} \sum_{j=1}^{M} (x_j - y_j)^2}$$

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$$CORR = \frac{\sum_{j=1}^{M} (x_j - \bar{x}) (y_j - \bar{y})}{\sqrt{\sum_{j=1}^{M} (x_j - \bar{x})^2} \sqrt{\sum_{j=1}^{M} (y_j - \bar{y})^2}}$$

535 where x_j and y_j denote the modeled and the observational values/retrievals,

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541 respectively, at the *j*th out of *M* records, and the overbars denote averages.

Figure 2. Distributions of the observation sites used in this study. Red solid circles are
the 52 surface flask sites used for evaluations, the shaded shows the 11 TRANSCOM
regions, the blue rectangle shows the Amazon region, which is defined the same as
Botta et al. (2012)

547 3. Experimental Design

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The assimilation system was run from May 1, 2009 to Dec 31, 2015. Two forward simulations with the prior and posterior fluxes were also conducted from May 1, 2009 to Dec 31, 2015, respectively. For both assimilation and forward runs, the initial field of 3-D CO₂ concentrations at 00:00 UTC May 1, 2009 was from the product of CT2017 as well, and the MOZART-4 model was run with the resolution of $2.8^{\circ} \times 2.8^{\circ}$. The first 8 months are considered as a spin-up run, and the results from Jan 1, 2010 to Dec 31, 2015 are analyzed and evaluated in this study.

555 During the assimilation, the resolution of λ is the same as the transport model. For 556 the state vector, the second scheme (Function 3) was adopted, namely the BIO CO₂ 557 exchanges and OCN fluxes are optimized in this study, and the FOSSIL and FIRE 558 carbon emissions are kept intact (the impact of this assumption on both the inverted

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560 global and regional BIO fluxes are very small (Table S4)). Following Wang et al. 561 (2019), global annual uncertainties of 100% and 40% are assigned to BIO and OCN CO_2 exchanges, respectively. Accordingly, the uncertainties of the scaling factor (λ) 562 for the prior BIO and OCN fluxes in each DA window at the grid cell level are assigned 563 to 3 and 5, respectively. The model-data mismatch error of XCO₂ is constructed using 564 the GOSAT retrieval error, which is provided along with the ACOS product. According 565 566 to the previous works of Wang et al. (2019) and Deng et al. (2014), all retrieval errors 567 are also uniformly inflated by a factor of 1.9 in this study, which is the same as Wang et al. (2019), but a lowest error is added in this study, which is fixed as 1 ppm. 568

569 4. Results and Discussions

570 4.1 Evaluation for the inversion results

571 4.1.1 Evaluation using assimilated GOSAT XCO₂ retrievals

Figure 3a shows the zonal mean XCO₂ model-data mismatch errors at different 572 573 latitudes during the study period. Compared with the GOSAT XCO2 retrievals, basically 574 all the zonal mean BIAS of the prior XCO₂ in different latitudes are greater than 1 ppm, 575 with a global mean of 1.8 ± 1.3 ppm (average \pm standard deviation), but for the posterior 576 XCO₂, most zonal average BIAS are within ±0.5 ppm, with global mean of -0.0±1.1 577 ppm. The global mean MAE and RMSE between the simulated and GOSAT retrieved 578 XCO₂ concentrations also decreases from a prior value of 2.0 and 2.2 ppm to 0.8 and 579 1.1 ppm, respectively (Table 1), indicating that the model-data mismatch errors between 580 the simulated and retrieved XCO2 are significantly reduced. Overall, for both prior and 581 posterior concentrations, the BIAS in the southern hemisphere is smaller than that in 582 the northern hemisphere. In the same hemisphere, the BIAS at low latitudes is smaller than that at high latitudes. Figure 4 shows the spatial distribution of the posterior XCO₂ 583 biases. It could be found that in most grids (~80%), the biases are within ±1ppm. In 584 Tropical Pacific, North Pacific, North Atlantic and Tropical Land, most biases are 585

586 positive, and in the northern extra-tropical lands, negative biases are dominant. This

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pattern may be related to the retrieval errors, and the large BIAS in the high latitudes may be attributed to the large retrieval errors in those areas, which are caused by the lower solar elevation angle. Overall, this small posterior BIAS, which is less than the retrieval error (Crisp et al., 2012), indicates that the GCAS<u>v2</u> system works well with the GOSAT XCO₂ retrievals in this study.





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and observed CO₂ mixing ratios; error bar represents the standard deviations of the

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biases at each latitude and each site, respectively)

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599 Table1. Statistics of the simulated surface CO₂ and XCO₂ concentrations against the

600 surface flask observations and GOSAT retrievals, respectively

	BIAS		N	MAE		RMSE		 CORR		
	<u>Prior</u>	Post.	Prior	Post.		Prior	Post.	Prior	Post.	
XCO ₂	<u>1.8±1.3</u>	<u>-0.0±1.1</u>	<u>2.0</u>	<u>0.8</u>		<u>2.2</u>	<u>1.1</u>	<u>0.95</u>	<u>0.96</u>	
Surface CO ₂	<u>1.6±1.8</u>	-0.5±1.8	<u>2.1</u>	<u>1.4</u>		<u>2.4</u>	<u>1.9</u>	<u>0.96</u>	<u>0.96</u>	

601 *mean \pm standard deviation

602





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shaded) XCO₂ concentrations (simulations minus observations/retrievals)

607 4.1.2 Evaluation using independent surface observations

608 Figure 3b shows the mean biases of the simulated surface CO2 mixing ratios at each flask site at different latitudes. It could be found that the BIAS of the prior CO2 mixing 609 610 ratios are basically greater than 1 ppm at different latitudes, with global mean of 1.6±1.8 ppm, after constraining using the GOSAT XCO2 retrievals, the BIAS at most sites are 611 within ± 1 ppm, with a global mean of -0.5 ± 1.8 ppm. These BIAS are similar to those 612 613 of Basu et al. (2013), in which the average model-observation bias decreased from a 614 prior value of 1.95 ppm to -0.55 ppm. The MAE and RMSE between the simulated and 615 surface flask concentrations are also reduced in most sites, with the global mean MAE and RMSE decreasing from 2.1 and 2.4 to 1.4 and 1.9 ppm, respectively (Table 1). The 616 617 BIAS in the northern hemisphere are significantly larger than those in southern 618 hemisphere, because the carbon flux in the northern hemisphere is more complex than 619 that of the southern hemisphere (Wang et al., 2019). In addition, the posterior BIAS in most sites are negative, especially in the middle latitudes in the northern hemisphere. 620 The significant negative biases (less than 1 ppm) are mainly distributed in North 621

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America, Europe, central Asia, while positive biases are mainly located along east Asian coast (Figure 4), indicating that the carbon sinks in North America and Europe might be overestimated in this study, while those in the upwind areas of east Asian coastal sites, mainly eastern China, may be underestimated.

629 Moreover, it also could be found that the global mean prior BIAS of XCO₂ (about 630 1.8 ppm) is greater than the surface concentrations (1.6 ppm), while the BIAS of XCO₂ reduced by inversion (about 1.8 ppm) is less than the reduction of BIAS in the surface 631 concentrations (about 2.1 ppm). This may be attributed to the fact that, on the one hand, 632 although the GOSAT XCO2 retrievals were bias-corrected, there may still be some 633 systematic deviations; on the other hand, the responses of surface observations to 634 635 changes in the surface carbon flux is faster than the XCO₂ concentrations, so that larger flux adjustments are needed to match XCO2 concentration with ground data. A similar 636 situation was reported in Wang et al. (2019). In their study, GOSAT XCO₂ retrievals 637 were used to optimize the terrestrial carbon flux in 2015. Their inversion reduced the 638 BIAS of simulated surface and XCO₂ (compared against TCCON sites) concentrations 639 640 by about 1.1 and 0.9 ppm, respectively.

Figure 5 shows the time series of simulated and observed CO₂ mixing ratios at four 641 sites, i.e., mlo, nwr, tik, and nat. The mlo and nwr sites are two mountain stations located 642 643 in the center of Pacific and western US, respectively, and nat and tik are two coastal 644 sites located in Amazon and Siberia, respectively (Figure 2). Overall, the posterior mixing ratios have a better agreement with the observations at all 4 sites. The mlo site 645 646 is an atmospheric baseline station. At mlo, the posterior mixing ratio well reproduces the observed concentration, while the prior concentrations are overestimated all the 647 time since the summer of 2010, especially during the summertime every year. Besides, 648 the posterior concentrations during the wintertime are underestimated, and the 649 650 underestimation gradually increases along with time. A similar situation also could be found at the nat site as well as other sites located in tropical and southern hemisphere 651 oceans (Figure not shown). Figure S1 shows the interannual variations of the global 652 mean BIAS. Clearly, the biases of surface CO2 are gradually accumulated, leading to 653

删除了:2 删除了:0 the relatively large mean bias (-0.5 ppm). If we remove the impact of accumulation, the annual <u>BIAS</u> is about -0.1 ppm per year (about -0.2 PgC yr⁻¹). There are no error accumulations at most land sites like nwr and tik. These indicate that the global net carbon sinks are slightly overestimated every year, but in different lands, there are interannual variations.





670 where $\sigma_{posterior}$ and σ_{prior} are the posterior and prior uncertainties, respectively. 671 The URs on regional carbon flux estimates vary significantly over time and space (Deng 672 et al., 2014; Takagi et al., 2011). Table 2 lists the annual mean 1-o URs relative to the prior uncertainties during 2010 ~ 2015, which were aggregated in the 22 TRANSCOM 673 regions and 4 large-scale regions. It shows that over land regions, the annual mean URs 674 675 are in the range of 6% ~ 27%. The regions with large UR are temperate South America, 676 southern Africa, temperate North America, Europe. The UR over tropical and boreal 677 regions are relatively small due to the lower spatial coverage of XCO2. This distribution 678 is similar to the results of Deng et al. (2014), which are mainly related to the spatial 679 coverage of GOSAT XCO₂. For the monthly UR, in high latitudes, there are high URs in the warm season and very low ones in cold seasons; in mid-latitudes, the UR is 680 significant throughout the year; and in tropical areas, it is related to the rainy season. In 681 682 the rainy season, the URs are very low due to the massive cloud coverage, while in the dry season, the monthly UR are significant, with the highest UR reaching 25%. Figure 683 6 shows the monthly uncertainties in temperate North America and Europe. It could be 684 found that in Europe, high URs are mainly during May ~ September, and in temperate 685 North America, there are high URs in each month, with the highest UR reaching 45%. 686 687 The highest monthly UR is in temperate South America, with value of 50%. The highest monthly and annual URs are lower than the ones given in previous studies (40%-70%, 688 Takagi et al., 2011; Deng et al., 2014; Saeki et al., 2013a), which may be related to the 689 690 grided state vector and shorter DA window used in this study. 691 Over the ocean regions, the URs are very low, with values in the range of $0.12\% \sim$ 3.7%. As shown in formula (14), the UR is mainly determined by the observation 692 uncertainty R and background error covariance P^b (prior uncertainty). Usually, a small 693

R and large *P^b* corresponds to a large UR, and vice versa. Since we used a scheme in
 which the prior uncertainties were proportional to the prior fluxes, thereby the regions
 with small prior fluxes would have small prior uncertainties and small URs. Compared

697 to those over the lands, there are much weaker fluxes and much larger XCO₂

698 <u>uncertainties (Wunch et al., 2017) over the oceans, resulting in the significantly lower</u>

- 699 URs over the oceans. Previous studies (e.g., Takagi et al., 2011; Kadygrov et al., 2009)
- 700 <u>also showed very low URs over the oceans.</u>

701 Table 2. Annual mean prior uncertainties and reduction rates (UR) aggregated in

702 different TRANSCOM Regions during 2010~2015

Decion	Prior Unc.	UR	Dagion	Prior Unc.	UR
Region	<u>(PgC yr⁻¹)</u>	<u>(%)</u>	Kegion	(PgC yr ⁻¹)	<u>(%)</u>
Boreal North America	<u>0.82</u>	7.8	North Pacific	<u>0.49</u>	<u>0.29</u>
Temperate North America	<u>1.62</u>	<u>26.4</u>	West Pacific	<u>0.15</u>	<u>0.47</u>
Tropical South America	<u>1.28</u>	6.4	East Pacific	0.42	<u>3.71</u>
Temperate South America	<u>1.27</u>	<u>27.2</u>	South Pacific	<u>0.33</u>	<u>0.42</u>
Northern Africa	<u>1.5</u>	5.9	Arctic Ocean	<u>0.30</u>	<u>0.14</u>
Southern Africa	<u>1.35</u>	<u>15.9</u>	North Atlantic	0.27	<u>0.17</u>
Boreal Asia	<u>1.24</u>	15.6	Tropical Atlantic	<u>0.13</u>	<u>0.60</u>
Temperate Asia	<u>1.23</u>	10.3	South Atlantic	<u>0.25</u>	<u>0.46</u>
Tropical Asia	<u>0.77</u>	8.0	Southern Ocean	<u>0.40</u>	<u>0.12</u>
Australia	<u>0.50</u>	10.0	North Indian Ocean	<u>0.17</u>	<u>0.43</u>
Europe	<u>1.31</u>	<u>19.8</u>	South Indian Ocean	<u>0.35</u>	<u>0.33</u>
Northern Lands	<u>2.91</u>	19.9	Northern Oceans	0.65	<u>0.13</u>
Tropical Lands	<u>2.57</u>	<u>9.0</u>	Tropical Oceans	<u>0.51</u>	<u>2.82</u>
Southern Lands	<u>1.38</u>	24.4	Southern Oceans	<u>0.68</u>	<u>0.27</u>
Global Lands	<u>4.24</u>	17.1	Global Oceans	<u>1.11</u>	<u>0.84</u>



707	Table $\frac{2}{2}$ presents the mean prior and posterior global carbon budgets during $2010 \sim$
708	2015 of this study. For comparison, the mean global carbon budgets from Global
709	Carbon Budget 2018 (GCP2018, Le Quéré et al., 2018), CT2017, and Jena CarboScope
710	(JCS, Rödenbeck, 2005) are also shown. Both CT2017 and JCS estimates of the
711	surface-atmosphere CO2 exchange were based on the atmospheric measurements of
712	CO ₂ concentrations. In this study, the JCS product of s04oc_v4.3 is adopted. It should
713	to be noted that JCS only provides the net biosphere exchange (NBE), which is the sum
714	of BIO carbon flux and FIRE carbon emissions, and no individual FIRE carbon
715	emissions data is available. To compare, the FIRE carbon emissions used in this study,
716	which is from CT2017, is also applied to the JCS data, namely the BIO carbon flux of
717	JCS in this manuscript is obtained from the <u>NBE</u> of JCS minus the FIRE carbon
718	emission of this study.

719 Table 3. Mean global carbon budgets during 2010 ~2015 estimated in this study as well

	Prior	Posterior	GCP2018	CT2017	JCS
Fossil fuel and industry (FOSSIL)	9.58	9.58	9.49	9.62	9.31
Biomass burning (FIRE)	2.02	2.02	1.52*	2.03	2.02
Terrestrial ecosystem (BIO)	-4.07 <u>±4.24</u>	-4.24 <u>+3.51</u>	-3.13	-4.29	-4.07
Ocean (OCN)	-2.47 <u>±1.11</u>	-2.56 <u>±1.10</u>	-2.46	-2.57	-2.25
Budget imbalance	-	-	-0.52	-	-
Net biosphere exchange_ (<u>NBE)</u> ***	-2.05 <u>±4.24</u>	-2.22 <u>+3.51</u>	-2.12	-2.27	-2.05
Global net carbon flux (AGR)	5.06 <u>±4.38</u>	4.80 <u>±3.67</u>	4.91**	4.79	5.01

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4.3, Global Carbon Budget

* land-use change emissions, **atmospheric growth in GCP2018, *** for GCP2018, it is the sum of BIO, FIRE and budget imbalance, and for the others, it is the sum of BIO

723 flux and FIRE emission.

724 The mean posterior BIO carbon flux during 2010-2015 in this study is -4.24 ± 3.51 725 PgC yr⁻¹ (negative/positive mean carbon uptake/release from/to the atmosphere, same 726 thereafter), and the OCN flux is -2.56±1.10 PgC yr⁻¹, after considering the FOSSIL

carbon emission (9.58 PgC yr⁻¹) and FIRE carbon emission (2.02 PgC yr⁻¹), the mean 733 734 global net carbon flux (i.e., atmospheric CO2 growth rate) inverted in this study is 4.80±3.67 PgC yr⁻¹. Both the posterior BIO and OCN carbon fluxes are stronger than 735 the prior ones, and the posterior global net carbon flux is weaker than the prior one. 736 Compared with the others, both posterior BIO and OCN fluxes are close to the ones of 737 738 CT2017, but higher than the ones of JCS. The AGR of GCP2018 was estimated directly 739 from atmospheric CO₂ measurements, which were provided by the US National 740 Oceanic and Atmospheric Administration Earth System Research Laboratory (NOAA/ESRL) (Dlugokencky and Tans, 2018), and therefore, it could be considered 741 as a true value. The posterior AGR in this study (4.8 PgC yr⁻¹) is slightly lower than 742 GCP2018 and very close to CT2017. Compared with GCP2018, the deviations of prior 743 and JCS AGR are 0.15 and 0.10 PgC yr⁻¹, while the ones of posterior and CT2017 are 744 -0.11 and -0.12 PgC yr⁻¹, respectively. 745

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746 4.4 Regional Carbon Flux

747 Figure 7 shows the distributions of the mean prior and posterior annual BIO and OCN carbon fluxes as well as their differences during 2010 - 2015. For the prior BIO 748 749 flux, carbon uptakes mainly occur over eastern North America, Amazon, southern Brazil, western Europe, southern Russia, eastern China, South Asia and Malay 750 Archipelago; and carbon releases mainly occur in western North America, eastern 751 Amazon, Argentina, most Africa, Indo-China Peninsula, and parts of eastern Europe 752 and Russia. For the prior OCN flux, carbon uptakes mainly happen in mid-latitude 753 regions in both hemispheres, while carbon sources are mainly in tropical oceans and 754 755 Southern Ocean. After the constraint with the GOSAT XCO₂ retrievals, the overall patterns of carbon sinks and sources are similar to the prior ones. However, the BIO 756 sinks in East and Central America, eastern Amazon, tropical Africa, Indo-China 757 Peninsula, and southwestern Russia are obviously increased, on the contrary, in western 758 North America, temperate South America, extra-tropical Africa, South Asia, Southwest 759 China, North China, Siberia, and parts of southern and northern Europe, the carbon 760

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765 sources are increased. For the OCN flux, in most tropical and northern hemisphere

766 oceans, the carbon sinks are slightly increased, while in most southern hemisphere

767 oceans, the carbon sources are slightly enhanced.





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Figure 7. Distributions of mean annual terrestrial ecosystem and ocean carbon fluxes (a, prior; b, posterior and c, their differences (posterior - prior), unit: gC m⁻²yr⁻¹)

Table <u>4 lists</u> the aggregated mean annual prior and posterior BIO carbon fluxes

during 2010-2015 for the 11 TRANSCOM land regions (Figure 2, Gurney et al., 2002)

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as well as 3 aggregated large-scale regions, i.e., Northern Lands, Tropical Lands, and 780 781 Southern Lands. Northern lands include Boreal North America, Temperate North America, Boreal Asia, Temperate Asia and Europe; Tropical Lands include Tropical 782 South America, Tropical Asia, Northern Africa and Southern Africa; and Southern 783 784 Lands include Temperate South America and Australia. For the prior, there is a largest carbon sink in Tropical South America, followed by Boreal Asia and Temperate Asia, 785 786 and a weakest carbon flux in Southern Africa. After optimization using GOSAT XCO2 787 retrievals, the carbon sinks of Temperate North America, Southern Africa are significantly increased, and those in Australia and Europe are also enhanced. However, 788 789 in Temperate South America, Northern Africa, Boreal Asia, and Temperate Asia, the 790 carbon sinks are decreased. Very small changes are found in Boreal North America, Tropical South America, and Tropical Asia, especially for Tropical South America, 791 792 however, as shown in Figure 7, there are obvious changes over different areas in 793 Tropical South America, thus the zero change in statistics in this region may be just a 794 coincidence. For the Amazon region (Figure 2), the estimated BIO flux is decreased from a prior of -0.52±1.46 PgC yr⁻¹ to -0.45±1.28 PgC yr⁻¹. The largest carbon sink 795 occurs in Temperate North America, followed by Tropical South America and Europe, 796 797 and the weakest sink appears in Northern Africa.

798 For comparisons, Table 4 also lists the mean BIO carbon fluxes of CT2017 and 799 JCS for the same period. For the 3 large-scale regions, i.e., Northern Lands, Tropical Lands and Southern Lands, the same as the global total BIO carbon sink, the carbon 800 801 sinks in these 3 regions are also similar to CT2017. However, in each region, the distributions of carbon sinks between this study and CT2017 are significantly different. 802 803 In Northern Lands, the carbon sinks estimated by this study are more evenly distributed, although Temperate North America has the largest carbon sink, and those in Boreal Asia, 804 Temperate Asia and Europe are also very strong and comparable. However, in CT2017, 805 the carbon sinks are mainly distributed in Boreal Asia and Temperate Asia, accounting 806 for more than 70% of the total sink in Northern Lands. The sinks in Temperate North 807 808 America and Europe are very weak or even neutral. In Tropical Lands, this study shows

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strong carbon sinks in Tropical South America and Tropical Asia, and a weak sink in 811 812 Africa, while CT2017 shows an opposite pattern. In Southern Lands, this study shows comparable sinks in Temperate South America and Australia, while CT2017 shows a 813 strong sink in Temperate South America and very weak one in Australia. Compared 814 with JCS, except for Temperate North America and Southern Africa, the carbon sinks 815 are comparable in other regions. Constraining with different observations might be one 816 817 of the main reasons among these studies. Many studies have shown differences between the constraints with in situ observations and XCO₂ retrievals (e.g., Wang et al., 2019; 818 819 Deng et al., 2014). Besides, these differences may be also related to the different prior BIO carbon fluxes among these studies, especially for the tropical land. The distribution 820 821 of the posterior BIO fluxes in this study and CT2017 are consistent with the 822 corresponding prior fluxes in the tropical land (Table 4). Using the same GOSAT XCO2 823 retrievals, Deng et al. (2014) adopted a similar prior flux with this study, which was also simulated using the BEPS model but globally neutralized, to infer the land fluxes 824 of 2010, their distributions are roughly consistent with this study, while Wang et al. 825 826 (2019) applied the prior flux from CT2016 to optimizing the fluxes in 2015, and they showed a similar distribution of land sinks over tropical lands to that of CT2017. 827

828

 Table 4. Regional BIO and FIRE flux in the 11 TRANSCOM land regions (PgC yr⁻¹)

Desiens	E'm	This S	tudy	СТ	ICO	
Regions	Fire	Prior	Posterior	Prior	Posterior	JCS
Boreal North America	0.065	-0.26 <u>±0.82</u>	-0.28 <u>±0.75</u>	-0.05	-0.39	-0.31
Temperate North America	0.022	-0.49 <u>±1.62</u>	-0.88 <u>±1.19</u>	-0.14	-0.23	-0.21
Tropical South America	0.220	-0.66 <u>±1.28</u>	-0.66 <u>±1.20</u>	0.02	-0.11	-0.43
Temperate South America	0.142	-0.30 <u>±1.27</u>	-0.15 <u>±0.93</u>	-0.16	-0.42	0.13
Northern Africa	0.385	-0.18 <u>±1.50</u>	-0.05 <u>±1.41</u>	-0.47	-0.82	-0.11
Southern Africa	0.628	0.01 <u>±1.35</u>	-0.14 <u>±1.14</u>	-0.63	-0.55	-0.66
Boreal Asia	0.097	-0.61 <u>±1.24</u>	-0.45 <u>±1.05</u>	-0.18	-0.99	-0.51
Temperate Asia	0.065	-0.51 <u>±1.23</u>	-0.42 <u>±1.10</u>	-0.15	-0.66	-0.69
Tropical Asia	0.258	-0.45 <u>±0.77</u>	-0.47 <u>±0.71</u>	-0.05	-0.07	-0.73
Australia	0.097	-0.16 <u>±0.50</u>	-0.23 <u>±0.45</u>	-0.15	-0.07	-0.08
Europe	0.015	-0.46 <u>±1.31</u>	-0.52 <u>±1.05</u>	-0.18	0	-0.44
Northern Lands*	0.26	-2.33 <u>+2.91</u>	-2.55 <u>±2.33</u>	-0.7	-2.27	-2.16
Tropical Lands**	1.49	-1.28 <u>±2.57</u>	-1.32 <u>±2.34</u>	-1.13	-1.55	-1.93
Southern Lands***	0.24	-0.46 <u>±1.38</u>	-0.38 <u>±1.04</u>	-0.31	-0.49	0.05
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*Northern lands include Boreal North America, Temperate North America, Boreal Asia, Temperate
 Asia and Europe; **Tropical Lands include Tropical South America, Tropical Asia, Northern Africa
 and Southern Africa; ***Southern Lands include Temperate South America and Australia.

834 Compared with other studies, the land fluxes (including FIRE but excluding FOSSIL) in South America (-0.45 ± 1.51 PgC yr⁻¹), Europe (-0.51 ± 1.05 PgC yr⁻¹), 835 Boreal Asia (-0.35±1.05 PgC yr⁻¹), Temperate Asia (-0.35±1.10 PgC yr⁻¹), Tropical Asia 836 $(-0.21\pm0.71 \text{ PgC yr}^{-1})$, and Australia $(-0.13\pm0.45 \text{ PgC yr}^{-1})$ are comparable with the 837 forest sinks in these regions during 2000-2007 estimated using forest inventory data by 838 839 Pan et al. (2011). However, the land fluxes in Africa and North America are significantly different from the estimates of Pan et al. (2011). In North America, based 840 on inventory-based calculations, the Second State of the Carbon Cycle Report 841 842 (SOCCR2, Hayes et al., 2018) estimated that the average annual net land ecosystem flux was -0.96 PgC yr⁻¹, and after considering the outgassing and wood products 843 emissions, they reported the land-based carbon sink was -0.606 PgC yr⁻¹ (±75%) during 844 845 the 2004 to 2013 time period. The land flux estimated in this study (-1.07 PgC yr⁻¹) is 846 close to the bottom-up estimate of the net land ecosystem flux, but much stronger than the reported land-based carbon sink of SOCCR2. In Africa, Ciais et al. (2011) shown a 847 comprehensive estimate for its carbon balance, given a sink of - 0.2 PgC yr⁻¹ (excluding 848 land-use change emissions) based upon observations. Our estimate of the BIO flux in 849 Africa is very consistent with this result. Moreover, most recently, Palmer et al. (2019) 850 851 inferred the carbon fluxes of pan-tropical lands in 2015 and 2016 using both GOSAT and the NASA Orbiting Carbon Observatory (OCO-2) XCO2 retrievals, and their 852 853 estimated net carbon emissions from African biosphere dominate pan-tropical atmospheric CO2 signals are similar to the results of this study. In Boreal Asia, the land 854 sink estimated by bottom-up approaches was in the range of $-0.11 \sim -0.76 \text{ PgC yr}^{-1}$ 855 (Hayes et al., 2011; Nilsson et al., 2003; Dolman et al., 2012; Zamolodchikov et al., 856 857 2017). CT usually reports a very strong carbon sink (Jacobson et al. 2020; Peter et al., 2007; Zhang et al., 2014), one possible reason is that there are no enough surface 858 observations in Asia boreal regions. Saeki et al. (2013b) conducted an inversion with a 859 focus on the Siberia region, and also derived a large sink of -0.56 ± 0.79 PgC yr⁻¹ only 860

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864	using the NOAA data, but after adding additional observations in Siberia, they obtained
865	a weaker uptake of -0.35 ± 0.61 PgC yr ⁻¹ . Our estimate (-0.35 ± 1.05 PgC yr ⁻¹) is in the
866	range of bottom-up estimates, and very consistent with the Siberia-focused inversion
867	(Saeki et al., 2013b). In Europe, previous GOSAT-based inversions consistently derived
868	a very large European sink, which was in the range of -0.6 \sim -1.8 PgC yr^-1(Basu et al.,
869	2013, Chevallier et al., 2014; Deng et al., 2014), while the ones constrained using
870	surface observations were much weak \underline{er} , in the range of 0 \sim -0.4 PgC yr $^{-1}$ (Peters et al.,
871	2007, 2010; Peylin et al., 2013; Scholze et al., 2019). Our estimate of the BIO flux in
872	Europe is smaller than the previous GOSAT-based inversions, and close to the estimate
873	of Pelylin et al. (2013). In the Amazon region, the posterior land flux is -0.45 \pm 1.28 PgC
874	yr ⁻¹ , which is in the range of the previous long-term forest biomass sink estimates of -
875	$0.28 \sim$ -0.49 PgC yr^{-1} (Phillips et al., 2009; Brienen et al., 2015), but larger than the
876	other inversions (e.g., Deng et al., 2016; Gatti et al., 2014).

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878	4. <mark>5</mark> ,1 Global land and ocean fluxes	删除了: 4
879	Figure <u>8</u> shows the interannual variations of the prior and posterior BIO and OCN	(删除了:7
880	fluxes. Overall, from 2010 to 2015, the prior BIO fluxes show an increasing trend, but	
881	for the posterior fluxes, there is no significant trend. Large differences between the prior	
882	and the posterior fluxes mainly occur in 2010 and 2015. In 2010, the posterior sink is	
883	much stronger than the prior, while in 2015, the posterior sink is much weaker than the	
884	prior. For the OCN flux, both prior and posterior fluxes show consistently upward	
885	trends, and except for 2015, the posterior sinks are basically stronger than the prior ones	
886	every year. For the AGR (Figure 2), the prior sink shows a significant downward trend,	 一 删除了:8
887	while the posterior one shows a slightly increasing trend. The same as the BIO fluxes,	
888	large differences mainly occur in 2010 and 2015.	





Figure 8. Interannual variations of global (a) BIO and (b) OCN fluxes of the prior and posterior as well as GCP2018, CarbonTracker 2017 (CT2017) and Jena CarboScope 895 896 (JCS)

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897 Compared with the other products, the interannual variations of the posterior BIO fluxes (Figure <u>&a</u>) are consistent with the inversions of CT2017 and JCS, and the 898 estimates of GCP2018. For each year, the inversions of this study are all in the range of 899 CT2017 and JCS, but higher than GCP2018. However, because GCP2018 has the item 900 of budget imbalance and the land-use change emission is different from the FIRE 901 902 emission, the BIO flux in GCP2018 is different from this study, so direct comparison with GCP2018 is not meaningful. For OCN fluxes, overall, there are no significant 903 904 differences among different estimates, and the upward trend of this study is similar to that of GCP2018, and higher than those of CT2017 and JCS. The interannual variation 905 of AGR in this study is also very consistent with GCP2018 (Figure 2). Except for 2012 906 and 2015, the absolute deviations of AGR between this study and GCP2018 are within 907 0.3 PgC yr⁻¹. 908

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Figure 2. Interannual variations of the atmospheric CO₂ growth rates

915 4.5,2 Regional land fluxes

916 Figure 10a, b, and c show the prior and posterior interannual variations of the BIO 917 fluxes in Northern Lands, Tropical Lands and Southern Lands, respectively. In Northern 918 Lands, the interannual variations of both prior and posterior fluxes are similar to the 919 corresponding global land totals (Figure <u>8a</u>), i.e., upward trend for the prior flux and no trend with the posterior one, indicating that the interannual variations of global BIO 920 921 fluxes are dominated by the fluxes in Northern Lands. In Tropical Lands, the 922 interannual variations of posterior fluxes are similar to the prior ones, however, compared with the prior sinks in 2010 and 2011, the posterior sinks are much stronger, 923 while in 2013 and 2015, they are much weaker. In Southern Lands, there are large 924 925 differences for the interannual variations between the prior and posterior fluxes. For the prior flux, the highest sink is in 2011 and the weakest in 2012, and after that, it increases 926 927 year by year, while for the posterior flux, the sink decreases from 2010 to 2013, and then increases. 928

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Figure <u>10</u>. Prior and posterior interannual variations of the BIO fluxes in (a) Northern
Lands, (b) Tropical Lands, and (c) Southern Lands, respectively, and (d) severe
drought areas of above 3 regions.

Drought is one of the most important factors that affect terrestrial carbon sinks, and 938 generally, severe drought will significantly reduce carbon sinks (e.g., Ma et al., 2012; 939 Zhao and Running, 2010; Ciais et al., 2005; Gatti et al., 2014; Phillips et al., 2009; 940 941 Vicente-Serrano et al., 2013). Previous studies (e.g., Liu et al., 2018) have used the 942 GOSAT XCO2 retrievals to infer the impact of droughts on terrestrial ecosystem carbon 943 uptake anomalies. Figure 10d shows the severe drought areas (SDAs) in the 3 large 944 regions every year, which were calculated according to the monthly Standardised Precipitation-Evapotranspiration Index at 12-month time scales (SPEI12) (Beguería et 945 al., 2010). Here, the database of SPEIbase v2.5 is used, and the severe drought is 946 defined as SPEI12 less than -1.5 (Paulo et al., 2012). In addition, only the severe 947 drought that happens in forests, shrubs and crops are counted in this study. It could be 948 found that the posterior fluxes have better correlations with the SDAs in all 3 regions, 949 i.e. a larger SDA leads to a weaker carbon sink, and vice versa. The correlation 950 951 coefficients between carbon sinks and SDAs in Northern Lands, Tropical Lands and 33

Southern Lands increase from prior values of -0.1, -0.25 and -0.44 to -0.53, -0.67 and -955 956 0.76, respectively, indicating that the inversion has improved the interannual variations of BIO fluxes in large scales. In addition, strong El Niño event happened during 957 2015~2016, and many researches have studied the responses of tropical land carbon 958 fluxes to this strong El Niño event (e.g., Wang et al., 2018b; Liu et al., 2017; Bastos et 959 al., 2018; Koren et al., 2018). Liu et al. (2017) found that relative to the 2011 La Niña, 960 961 the pantropical biosphere released 2.5 ± 0.34 PgC more carbon into the atmosphere in 962 2015. Bastos et al. (2018) showed a smaller difference of carbon fluxes between 2015 963 and 2011 using both bottom-up and top-down approaches, which was in the range of $-0.7 \sim -1.9$ PgC yr⁻¹. In this study, compared with the prior, our inversion significantly 964 965 enhances the difference between 2011 and 2015 (Figure 10b), and shows that 2015 released 1.35 PgC more than 2011 in the pantropical region (defined as Liu et al., 2017), 966 967 which is much smaller than Liu et al.'s result, but agree well with the result of Bastos et al. (2018). 968

969 Moreover, Figure 11 shows the prior and posterior interannual variations of the 970 BIO fluxes on the 11 TRANSCOM land regions. In North America, including Temperate North America and Boreal North America, the prior fluxes show an upward 971 972 trend, while the posterior fluxes show a downward trend. In Boreal Asia and Temperate Asia, there are significant upward trends for the prior fluxes, but no significant trends 973 974 are found in the posterior fluxes. In Temperate South America, although the prior and posterior fluxes show trends of weakening first and then increasing, the years in which 975 976 the carbon sink is weakest are not consistent: the prior flux is weakest in 2012, while 977 the posterior one is in 2013. Similarly, in northern Africa, the prior and posterior fluxes show a trend of increasing and then decreasing, but the prior flux is the largest in 2014, 978 while the posterior one is strongest in 2011. In other regions, i.e., Tropical South 979 America, Tropical Asia, Southern Africa, Australia and Europe, the trends between the 980 prior and posterior fluxes are similar, especially in Tropical South America and Tropical 981 Asia, the prior and posterior fluxes are very close every year. Among them, in Southern 982 983 Africa and Australia, the posterior fluxes have more significant interannual variations

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than the prior fluxes, and in Europe, the posterior sink is much weaker in 2015, and stronger in 2010 and 2013 than the prior one.

The same as above, we also investigate the relationships between the interannual 988 989 variations of carbon sinks and SDAs in the 11 TRANSCOM land regions. As shown in 990 Table 5, in Temperate South America, Boreal Asia, and Europe, the posterior sinks have a better correlation with the SDAs than the prior sinks, especially in Europe, the 991 correlation coefficient increases from a prior value of -0.33 to -0.85. However, in other 992 regions, there is no obvious improvement, and in some regions, the relationships are 993 even getting worse, such as Boreal North America, Temperate North America, Northern 994 Africa and Southern Africa. One possible reason is that there are usually higher annual 995 996 mean temperatures in drought years, which might extend the growing season of vegetation, thereby enhance the carbon uptake and offset the impacts of drought. A 997 previous study (Wolf et al., 2016) showed that in 2012, Temperate North America 998 experienced an extreme summer drought event, and along with the warmest spring on 999 record. They quantified the impact of this climate anomaly on the carbon cycle and 1000 1001 concluded that the warm spring largely increased spring carbon uptake, and thus compensated for reduced carbon uptake induced by the summer drought. Liu et al. 1002 1003 (2018) reported that because of the compensating effect of the carbon flux anomalies between northern and southern US in 2011 and between spring and summer in 2012, 1004 the annual carbon uptake decreased by 0.10±0.16 PgC in 2011, and increased by 1005 0.10±0.16 GtC in 2012 over US compared with the averaged state. In this study, 1006 1007 compared with the mean flux during 2010-2015, the carbon sink in Temperate North America decreased by 0.09 PgC yr⁻¹ in 2011, and increased by 0.14 PgC yr⁻¹ in 2012, 1008 which is very close to the result of Liu et al. (2018). In Australia, both the prior and 1009 posterior fluxes have very good relationships with the SDAs. The significantly 1010 enhanced carbon uptake during 2010-2012 is consistent with the finding in Detmers et 1011 al. (2015), who inferred an even stronger carbon sink of -0.77±0.10 PgC yr⁻¹ from the 1012 end of 2010 to early 2012 using the GOSAT XCO2 product, and they confirmed that 1013 1014 this enhanced sink is related to the strong La Niña episode, which brought a record-

breaking amount of precipitation, resulting in an enhanced growth of vegetation. In 1016 1017 Tropical South America, the impacts of the 2010 drought on the carbon uptake over Amazon have been extensively studied (e.g., Doughty et al., 2015; Gatti et al., 2014; 1018 van der Laan-Luijkx et al., 2015). 2010 is a drought year, while 2011 is a wet year in 1019 the Amazon region, compared to 2011, Gatti et al. (2014) estimated the no-fire carbon 1020 exchange was reduced by 0.22 PgC yr⁻¹, van der Laan-Luijkx et al. (2015) derived a 1021 1022 decrease of biospheric uptake ranging from 0.08 to 0.26 PgC yr⁻¹, and Doughty et al. (2015) concluded that drought suppressed Amazon-wide photosynthesis by 0.23-0.53 1023 PgC yr⁻¹. In this study, our inversion reduces the difference of carbon uptake between 1024 2010 and 2011 from a prior of 0.62 PgC yr⁻¹ to 0.28 PgC yr⁻¹, which is much more 1025 1026 consistent with the previous estimates.

Carbon uptake occurs mainly through photosynthesis of vegetation leaves. Leaf 1027 1028 area index (LAI) is a measure of leaf area per unit area. Buchmann and Schulze (1999) shown that there are strong relationships between the interannual changes of carbon 1029 uptake and LAI in grasslands, C4 crops, and coniferous forests, but no significant 1030 relationship in broad-leaved forests; Chen et al. (2019) also showed that from 1981 to 1031 2016, the increase in LAI contributed significantly to the increase in global BIO carbon 1032 1033 sinks. Therefore, we further investigate the relationships between the interannual 1034 changes of carbon sinks and LAIs in the 11 TRANSCOM regions (Table 5). Here, the 1035 LAI data are from the GIMMS LAI3g product, which has a spatial resolution of 1/12 1036 degree and a time interval of 15 days (Zhu et al., 2013). As shown in Table 5, in Boreal 1037 North America, Temperate North America, Northern Africa and Southern Africa, compared with the prior fluxes, there are better relationships between the posterior 1038 1039 carbon sinks and LAIs, the correlation coefficients increase from prior values of -0.4, 1040 0.31 and 0.35 to 0.62, 0.73 and 0.90 respectively, suggesting that the inversion of this study may also improve the interannual variations of carbon sinks in these 4 regions at 1041 a certain extent. 1042

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1046Figure 11. Prior and posterior interannual variations of the BIO fluxes on (a) Boreal1047North America, (b) Temperate North America, (c) Tropical South America, (d)1048Temperate South America, (e) Northern Africa, (f) Southern Africa, (g) Boreal Asia,1049(h) Temperate Asia, (i) Tropical Asia, (j) Australia, and (k) Europe

Table 5. Correlation coefficients of severe drought areas (SDAs) and regional mean

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LAI with the BIO sinks in each region

Designs	S	DA	LAI		
Regions	Prior	Posterior	Prior	Posterior	
Boreal North America	-0.29	0.36	-0.4	0.62	
Temperate North America	-0.54	-0.27	0.31	0.73	
Tropical South America	-0.1	-0.2	0.64	0.49	
Temperate South America	-0.41	-0.74	0.72	0.24	
Northern Africa	0.51	0.2	0.81	0.89	
Southern Africa	-0.53	0.41	0.35	0.9	
Boreal Asia	-0.17	-0.35	0.49	0.1	
Temperate Asia	0.33	0.33	0.55	0.38	
Tropical Asia	-0.03	0.16	0.69	0.71	
Australia	-0.85	-0.73	0.88	0.83	
Europe	-0.33	-0.85	0.85	0.58	

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1058 5. Summary and Conclusions

In this study, we upgrade the GCAS system to GCASv2 with new assimilation 1059 algorithms, procedures and a localization scheme, a higher assimilation parameter 1060 resolution, and the ability to assimilate XCO₂ retrievals. Then, we use the GOSAT 1061 XCO₂ retrievals to constrain terrestrial ecosystem and ocean carbon fluxes from May 1062 1, 2009 to Dec 31, 2015, using the GCASv2 system. We compare the simulated prior 1063 and posterior XCO₂ against the corresponding GOSAT XCO₂ retrievals to test the 1064 effectiveness of the assimilation system and evaluate the posterior carbon fluxes by 1065 comparing the posterior CO₂ mixing ratios against observations from 52 surface flask 1066 sites. The distribution and interannual variations of the posterior carbon fluxes at both 1067 1068 global and regional scales from 2010 to 2015 are shown and discussed.

Compared with the GOSAT XCO₂ retrievals, the global mean BIAS and RMSE 1069 decrease from prior values of 1.8 ± 1.3 and 2.2 ppm to -0.0 ± 1.1 and 1.1 ppm, respectively, 1070 1071 indicating that the GCASv2 system works well with the GOSAT XCO2 retrievals. Independent evaluations using surface flask CO2 concentrations showed that the 1072 posterior carbon fluxes could significantly improve the modeling of atmospheric CO₂ 1073 concentrations, with the global mean BIAS and RMSE decreasing from prior values of 1074 1.6±1.8 and 2.4 ppm to -0.5±1.8 and 1.9 ppm, respectively. The large negative biases 1075 are mainly distributed in North America, Europe, indicating the overestimates of carbon 1076 1077 sinks over these areas. Evaluations also show that the biases gradually increase along with the time in most tropical and southern hemisphere ocean sites, but no accumulation 1078 1079 is found at most land sites, indicating that globally, the carbon sinks may be overestimated every year, but in different lands, the deviations of the estimates may 1080 differ each year. 1081

1082 Globally, the mean annual BIO carbon sink and the interannual variations 1083 inferred in this study are very close to the estimates of CT2017 during the study period, 1084 and the estimated mean AGR and interannual changes are also very close to the 1085 observations, with mean annual bias of -0.11 PgC yr⁻¹. Regionally, the inversion shows

that in the northern lands, the carbon sink of Temperate North America is the strongest, 1086 1087 and those in Boreal Asia, Temperate Asia and Europe are also very strong and comparable; in the tropics, there are strong sinks in Tropical South America and 1088 Tropical Asia, but a very weak sink in Africa. These distributions are significantly 1089 different from the estimates of CT2017, probably due to the different prior fluxes and 1090 CO2 observations used for inversion. However, our estimates in most regions or 1091 1092 continents are comparable or in the range of previous bottom-up estimates. The 1093 inversion also changed the interannual variations of carbon sinks in most TRANSCOM and hemisphere scale land regions, leading to their better relationship with the 1094 variations of severe drought or LAI, indicating that the inversion with GOSAT XCO2 1095 retrievals may help to better understand the interannual variations of regional carbon 1096 fluxes. 1097

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1099 Data availability

1100 The code of GCASv2 system and the inversion results of this study are available to the 1101 community and can be accessed upon request from Fei Jiang (jiangf@nju.edu.cn) at 1102 Nanjing University.

1103 Author contributions

FJ, JC and WJ designed the research; FJ run the model, analyzed the results and wrote
the paper; HW handled the GOSAT XCO₂ retrievals; WH analyzed the drought data;
XL run the BEPS model; FJ lead the update of the GCAS system, and XT, HW, JW, SF,
GL, ZC, SZ, JL, WH, and MW participated in it; RL, PS and PK provided the surface
CO₂ observations; JC, WJ and HW participated in the discussion of the inversion results
and provided input on the paper for revision before submission.

1110 Competing interests

1111 The authors declare that they have no conflict of interest.

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