Supplement of

Simultaneous shipborne measurements of CO_2, CH_4 and CO and their application to improving greenhouse gas flux estimates in Australia

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Table S1. Shipborne measurement time periods.

Trip	Measurement collection date
1	11 - 28 April 2012
2	2 - 10 May 2012
3	7 - 18 June 2012
4	17 - 29 June 2013
5	4 - 26 July 2013
6	27 July - 10 August 2013
7	27 September - 3 October 2013

Table S2. The mean measured and modelled mole fractions of the gases for different background regions with one standard deviation.

Region	Coral Sea		Tasman Sea		Indian Ocean	
	Measurements	Model	Measurements	Model	Measurements	Model
CO_2 (ppm)	389.3±0.3	390.4±0.6	389.2±0.2	390.4±0.2	393.2±0.3	393.7±0.4
CH_4 (ppb)	1761±3	1753±2	1766±2	1763±2	1783±1	1777±2
CO (ppb)	50±1	54.4±0.7	50±1	56.9±0.7	60±3	62±3

Table S3. Threshold values used during the separation of the total amount of each gas into background values and enhancements. The values refer to the change of each gas between latitudinal points that most clearly separate the background regions from the enhancements for each section.

Section	NB 2012	SB 2012	NB 2013	SB 2013
CO_2 (ppm)	0.2	0.15	0.2	0.2
CH_4 (ppb)	1	1	1	0.8
CO (ppb)	1	0.8	1	1

Table S4. Model and measurements statistics (mean bias (MB), standard deviation (σ), correlation coefficient (r) and the slope of the regression line)[1].

	2012		2013		2012		2013	
	NB	SB	NB	SB	NB	SB	NB	SB
Total	MB$\pm\sigma$				r(slope)			
CO_2	0.39±1.62	0.98±0.78	0.68±0.75	0.81±1.87	0.64(0.34)	0.51(0.3)	0.65(1.0)	0.11(0.08)
CH_4	-0.27±10.8	-3.43±9.62	-6.18±3.25	-8.53±13.8	0.74(0.93)	0.65(0.36)	0.82(0.87)	0.36(0.17)
CO	3.3±11.3	6.2±2.92	-2.18±17.6	-0.75±10.1	0.62(0.25)	0.7(0.43)	0.66(0.55)	0.51(0.27)
Background								
CO_2	0.94±0.46	1.09±0.38	0.35±0.57	1.18±0.78	-	-	-	-
CH_4	-0.12±8.15	-2.76±1.98	-5.99±2.78	-4.72±4.49	-	-	-	-
CO	7.8±3.66	6.95±1.1	1.31±2.47	2.8±5.44	-	-	-	-
Enhancements								
CO_2	-	-	-	-	0.58(0.29)	0.49(0.23)	0.72(0.87)	0.014(0.011)
CH_4	-	-	-	-	0.76(0.98)	0.67(0.27)	0.76(0.64)	0.29(0.1)
CO	-	-	-	-	0.66(0.24)	0.67(0.3)	0.58(0.48)	0.46(0.2)

[1] All values are based on the 0.1° latitude averaged data from the ship cruises. In order to quantify the model-measurement agreement of both the variability and total amount of each gas we separated out the total amount of each gas into background values and enhancements as described in Figure S5. Units for the MB and SD are ppm for CO_2 and ppb for CH_4 and CO.

Table S5. Contribution of different tracers to the total amount of each gas based on the simulated values with the GEOS-Chem model for the ship measurements during the 4 measurement sections in 2012 and 2013, and their average across the 4 sections.

Tracers	Contribution (%)				
	2012		2013		
	NB	SB	NB	SB	Average
CO_2	Median±(75th-25th)				Mean±1σ
Fossil Fuel	69.8±0.4	69.8±0.2	70.0±0.8	70.1±0.4	69.9±0.2
BB*	18.9±0.1	18.83±0.03	18.6±0.2	18.6±0.2	18.7±0.1
Biofuel	7.13±0.04	7.16±0.01	7.12±0.07	7.11±0.02	7.13±0.02
Ship	2.45±0.01	2.45±0.01	2.44±0.02	2.44±0.01	2.44±0.01
Aviation	1.77±0.01	1.77±0.01	1.77±0.02	1.76±0.01	1.78±0.01
Chemical Source	$3.3e^{-4}$±$1.2e^{-6}$	$3.3e^{-4}$±$5.3e^{-7}$	$3.3e^{-4}$±$2.12e^{-6}$	$3.2e^{-4}$±$1.6e^{-6}$	$3.3e^{-4}$±$1.8e^{-6}$
Balanced Biosphere	-1.1±0.7	-0.6±0.5	-0.2±0.4	0.07±1.14	-0.4±0.4
Net Terrestrial Exch	-46.2±0.1	-46.4±0.1	-46.1±0.5	-46.0±0.3	-46.2±0.1
Ocean	-17.7±0.2	-17.82±0.04	-18.0±0.3	-17.8±0.2	-17.8±0.1
CH_4					
Wetland	33.2±0.1	33.20±0.06	33.1±0.2	32.93±0.06	33.1±0.1
Livestock	20.5±0.3	20.6±0.2	20.6±0.1	20.7±0.1	20.59±0.05
Oil & Gas	11.98±0.05	11.97±0.03	12.0±0.1	12.04±0.05	12.01±0.03
Waste	10.89±0.06	10.89±0.04	10.91±0.08	10.91±0.05	10.90±0.01
Coal mining	7.8±0.2	7.82±0.05	7.88±0.08	7.9±0.2	7.88±0.06
Rice	6.81±0.03	6.80±0.01	6.79±0.05	6.78±0.01	6.81±0.01
BB	2.99±0.01	2.98±0.01	2.94±0.04	2.92±0.03	2.96±0.03
Termites	2.26±0.01	2.27±0.01	2.27±0.07	2.27±0.01	2.27±0.01
Biofuel	2.17±0.01	2.17±0.01	2.18±0.02	2.18±0.01	2.17±0.01
Other AN*	1.30±0.01	1.31±0.01	1.31±0.01	1.31±0.01	1.31±0.01
Soil Absorption	-3.29±0.01	-3.29±0.01	-3.29±0.06	-3.31±0.02	-3.29±0.01
CO					
CH_4 oxidation	51±1	46.7±0.4	46±2	42±2	46±3
NMVOC	23±4	23.6±0.6	24±7	25±4	24.1±0.6
AN S America	3.9±0.3	6.7±0.5	7±1	6.6±0.6	6±1
BB Africa	3.5±0.2	4.6±0.5	7±3	8±5	6±2
BB Australia	6±4	6±11	3±13	3±6	4±2
AN Other	4±1	4.9±0.4	5±1	5±1	4.7±0.6
AN Africa	2.4±0.3	3.4±0.5	4.3±0.8	3.9±0.2	3.5±0.7
AN Australia	3±8	0.7±2.9	0.6±0.4	2±2	1.4±0.8
BB S America	1.7±0.3	2.01±0.07	2.1±0.3	4±3	2.4±0.9
BB Other	1.4±0.9	1.6±0.2	0.69±0.09	0.58±0.05	1.1±0.5
BB E Asia	0.07±0.01	0.079±0.006	0.4±0.1	0.3±0.1	0.2±0.1
BB Indonesia	0.07±0.02	0.076±0.005	0.1±0.3	0.27±0.07	0.13±0.08

*BB - Biomass Burning, AN - Anthropogenic. The contribution for the 4 measurements sections (e.g. NB) represent the meadian of the modelled data for the specific trips with the interquartile range (difference between the 75th and 25th percentile). The average column represents the mean contribution across the 4 sections with one standard deviation (σ). The order of the tracers is from largest to the smallest contribution. Positive tracers represent sources while negative contributions represent the sink of the specified gases.

Table S6. Measured (Meas) and modelled (Mod) enhancement ratios with the standard error, and the correlation coefficient (r) of the species during specific coincident enhancement events.

	ΔCH_4:ΔCO (ppb·ppb^{-1})		ΔCH_4:ΔCO_2 (ppb·ppm^{-1})		ΔCO:ΔCO_2 (ppb·ppm^{-1})	
Event	Ratio Meas/Mod	r Meas/Mod	Ratio Meas/Mod	r Meas/Mod	Ratio Meas/Mod	r Meas/Mod
1	1.7±0.3/1.83±0.02	0.93/1.00	15±3/9.1±0.5	0.94/0.99	8±1/5.0±0.3	0.94/0.99
3	0.8±0.3/3.93±0.06	0.66/1.00	6±1/10.3±0.2	0.82/1.00	4.6±0.8/2.62±0.05	0.83/1.00
4	1.6±0.3/2.42±0.09	0.95/1.00	8±2/12±7	0.86/0.66	4±2/5±3	0.74/0.62
5	1.4±0.3/5.0±0.7	0.89/0.95	3.5±0.3/9±3	0.98/0.81	2.2±0.4/1.5±0.7	0.93/0.70
6	1.2±0.4/1.4±0.2	0.83/0.93	2.7±0.9/9±5	0.80/0.68	2.1±0.5/5±3	0.88/0.61
7	3.5±0.6/1.9±0.6	0.90/0.77	3.8±0.4/12±2	0.96/0.92	0.9±0.2/4±1	0.84/0.83
9	4.2±0.6/3.6±0.1	0.89/1.00	15±3/14.1±0.3	0.83/1.00	3.3±0.6/3.88±0.09	0.84/1.00
10	1.0±0.1/-0.3±0.1	0.93/-0.61	4.7±0.6/-6±4	0.91/-0.37	4.4±0.4/24±5	0.95/0.76
12	0.8±0.3/1.7±0.6	0.72/0.74	2.0±0.6/4.0±0.9	0.78/0.86	1.4±0.6/1.2±0.6	0.63/0.58
17	0.16±0.05/0.15±0.05	0.76/0.72	6.0±2/5±2	0.79/0.65	20±10/24±8	0.55/0.73
8	1.0±0.1/2.5±0.3	0.95/0.95				
11	1.0±0.1/1.6±0.5	0.87/0.64				
13	0.27±0.06/0.069±0.003	0.80/0.99				
14	2.2±0.5/2.4±0.7	0.82/0.74				
15	1.24±0.07/1.86±0.08	1.00/1.00				
16			1.9±0.3/10.1±0.3	0.94/1.00		
2					2.3±0.4/0.21±0.08	0.87/0.64

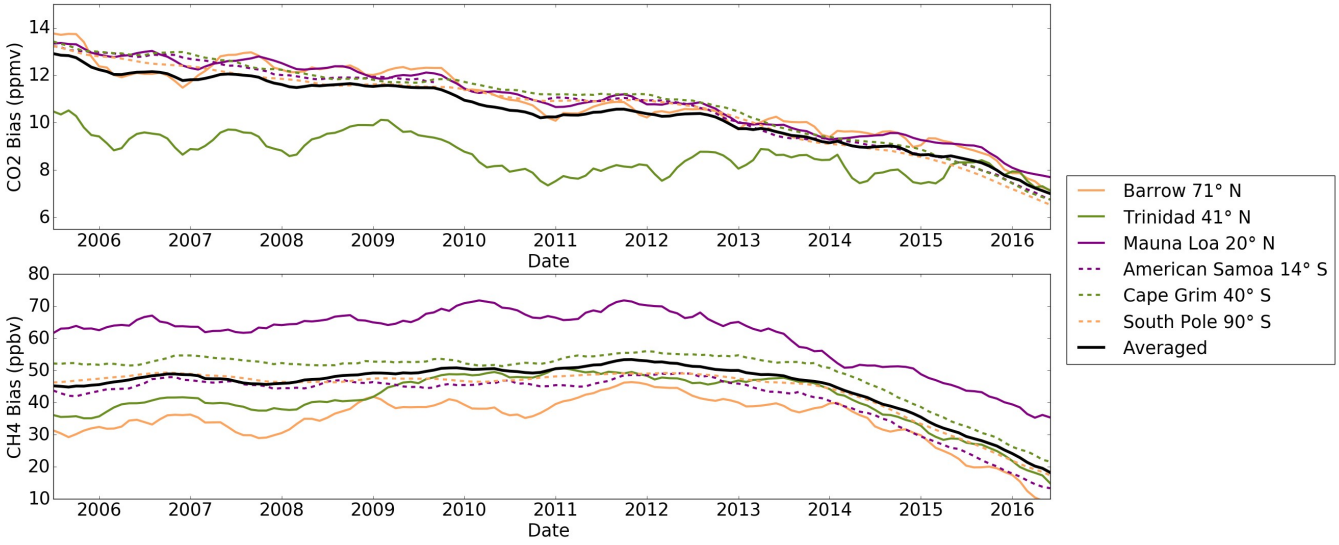

Figure S1. The model - measurement offset used for the bias correction method for CO_2 and CH_4. This offset was calculated by a globally averaged 13 point running mean on the difference between the modelled and measured data at the specified background sites. Solid lines represent background sites in the northern hemisphere while the dashed lines represent the offsets for the southern hemisphere background stations.

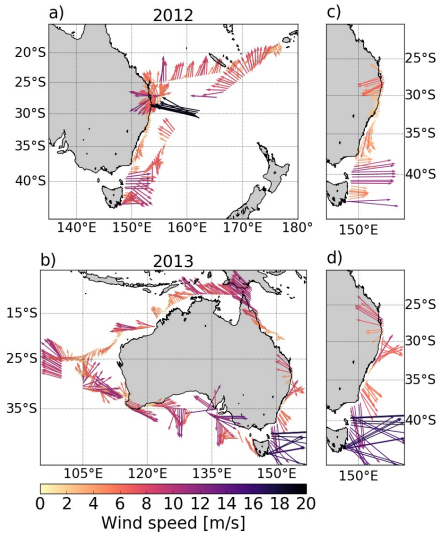

Figure S2. Wind speed and directions along the measurement tracks in 2012 (a) and 2013 (b) from MERRA2 meteorology. Plots c) and d) are zoomed in versions of the winds along the east coast for both years.

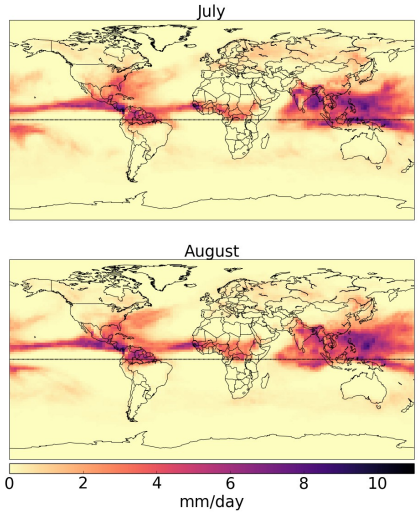

Figure S3. Position of the ITCZ and SPCZ during July and August 2013 based on total convective precipitation at the ground from MERRA2 meteorology processed with the GEOS-Chem model.

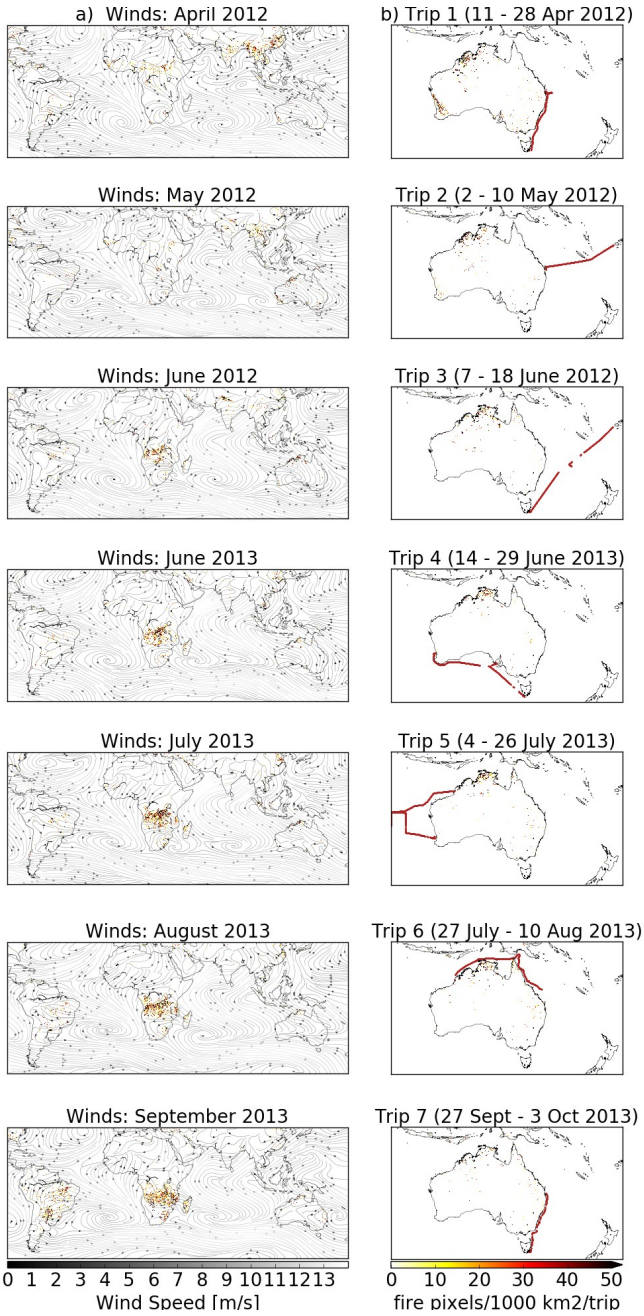

Figure S4. Total active burning fire detection with the MODIS instrument during the seven ship cruises in 2012 and 2013 globally (a) and for Australia (b) with the measurement tracks (red line) during the specific trips. Plots under a) show the total active fires 3 to 1 week prior to the trips, and monthly wind fields from the GEOS-Chem model to identify biomass burning emissions from other regions and their transport. Plots under b) show the total active fires during the duration of the specific trips. The active burning fires are represented with the count of the number of fires observed within a 1,000 km^2 area.

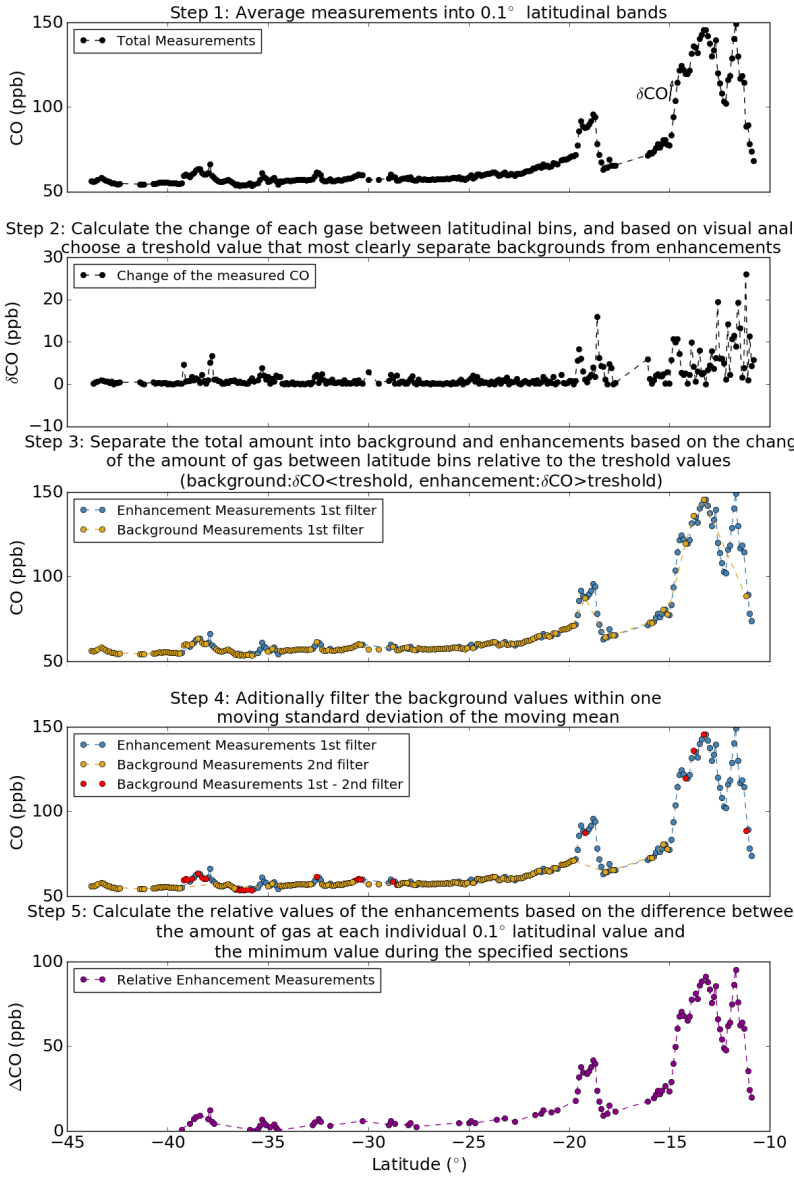

Figure S5. The separation process of the measured and modelled data into background values and enhancements. We use CO from the 2013 NB section as an example. The threshold values for each gas used during the separation can be found in Table S3.

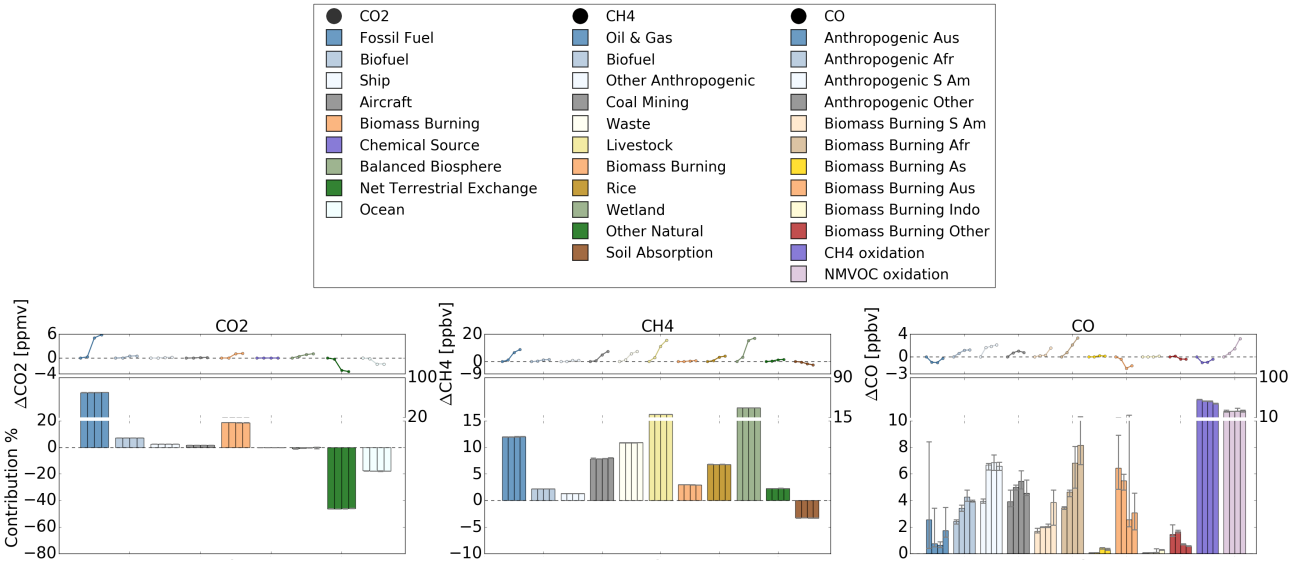

Figure S6. CO_2, CH_4 and CO tracer contribution across the 4 measurement section in 2012 and 2013 (2012 NB, 2012 2B, 2013 NB, 2013 SB, from left to right) for the total amount of each gas.

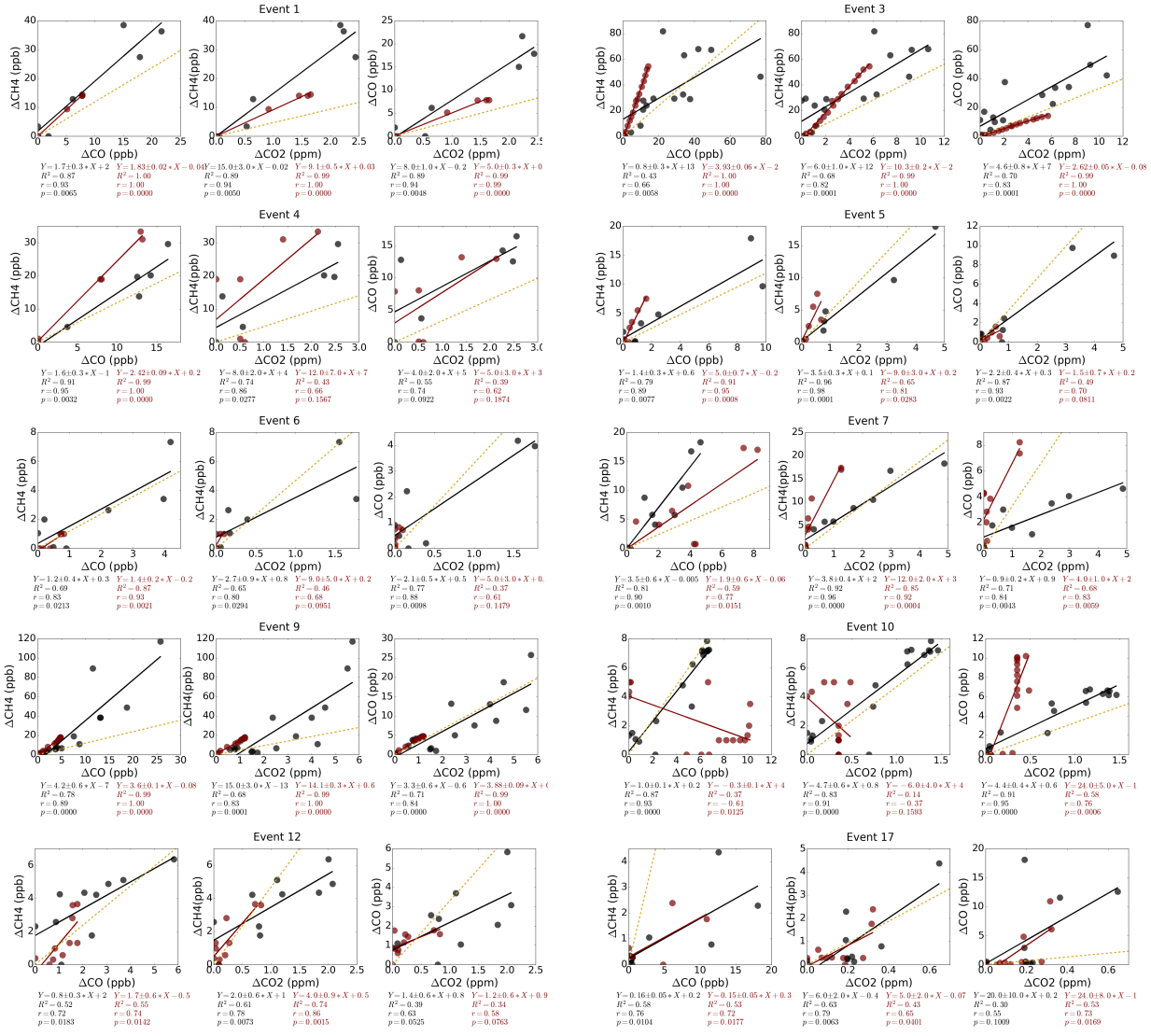

Figure S7. Correlations between observed (black) and modelled (red) ΔCO_2, ΔCH_4 and ΔCO with a linear regression fit for events when all three gases showed co-enhancements. The yellow dashed line represents the median measured enhancement ratio (slope) of all the events during the specific combination of the gases (ΔCH_4:ΔCO = 1.2 ppb·ppb^{-1}; ΔCH_4:ΔCO_2 = 4.7 ppb·ppm^{-1}; ΔCO:ΔCO_2 = 3.3 ppb·ppm^{-1}) and it is added as a reference line for the individual enhancement ratios.

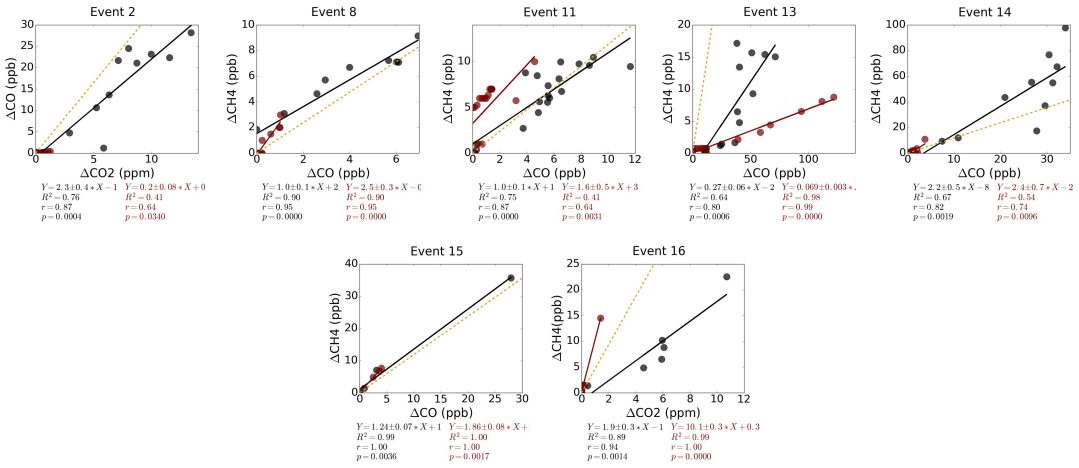

Figure S8. Same as Figure S7 but for events when only two gases showed co-enhancements.

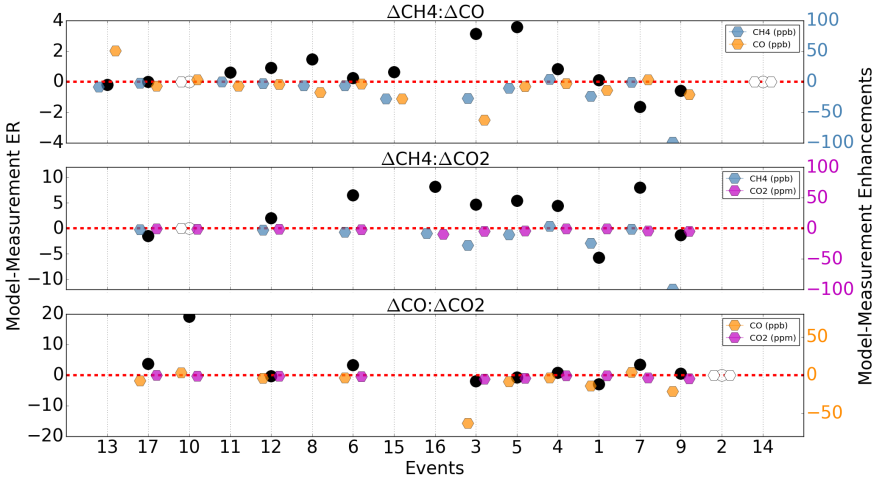

Figure S9. Model and measurement enhancement ratio (ER, black dots) and enhancement (coloured markers) differences for the species that showed coincident enhancements. The white markers represent enhancements and ERs where the model failed to capture the measured values. Units for the ERs are same as in Table S6.