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The CAMS interim Reanalysis of Carbon Monoxide, Ozone and Aerosol for 2003–2015

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1 Totals and variability of the emissions of CR and CAMSIRA

In the paper, we focus on 7 regions (Europe, North America, East Asia, South America, North Africa Tropical Africa and Maritime South East Asia) as well as the tropics, the Arctic and the Antarctic. The areas are shown in Figure S1 and the coordinates of the bounding boxes are given in Table 3 of the paper. Table S1 list the annual mean, maximum and minimum emissions for different source regions for CO, NO, isoprene (C_5H_8,) and all aerosol components (desert dust, sea salt, black carbon, organic matter and SO_2 tracer). An estimated linear trend, if significant with 95% confidence interval, is also listed. Time series of the anthropogenic and biomass burning emissions are shown in Figure S2.

The anthropogenic MACCity emissions for CO and NO decrease over Europe and North America in the range of 1 to 5% per year, whereas they increase over South East Asia by a similar amount. The global trend is almost zero for CO and slightly positive for NO.

The observed emissions from biomass burning as well as modelled emission of biogenic VOC, desert dust and sea salt depend strongly on the inter-annual variability of the meteorological situation but changes in land use also play a role for long term trends. Besides a large year-to-year variability, the biomass burning emissions for CO show no significant trend over the whole 2003–2015 period. However, a significant reduction occurred until 2013 when global CO biomass burning emissions decreased from 423Tg in 2003 to 288 Tg in 2013. The decreasing trend was most pronounced in South America (-7.5%/yr) where it occurred over the whole period.

The variability of biomass burning in Maritime South East Asia (MSEA) is dominated by the occurrence of El-Nino related dry conditions (Field et al., 2009), which led to extreme emissions in 2006, 2009, 2014 and most importantly in 2015. The MSEA emissions in 2015 reached 140 Tg, which was about 31% (447 Tg) of the global biomass burning emissions in that year, whereas in earlier years the emissions from this region constituted 5–15% of the global value. The Indonesian fires in September and October of 2015 caused by far the highest annual wildfire emissions as well as the highest total monthly CO emissions in the whole period (Huijnen et al., 2016).

Tropical Africa, which consists of two distinct fire regions with different seasonality north and south of the Equator, is usually the largest source of CO biomass burning emissions because it contributes about 30% of the global value. There was little inter-annual variation in the annual emission from this region between 2003 and 2015 although a small negative, but not significant, trend was detected.

As the biomass burning emissions of black carbon and organic matter are mostly proportional to the CO emissions the discussion above is also valid for these aerosol species.

Month-specific isoprene (C_5H_8) emissions from vegetation were only simulated for the years 2003–2010 and a climatology was used for the later years (see section 2). They have decreased slightly in the 2003-2010 period, in particular in South America, where biomass burning emission decreased also significantly. The lowest global annual isoprene emissions were about 85% of the highest annual emissions in this period.

For methane, which also leads to CO formation during its oxidation, no emissions were used but instead surface concentrations were nudged to constrain atmospheric concentrations. An observations based trend of 1.5–3.5 ppm per year was applied to these surface concentrations and consequently the global CH_4 burden increased from 4840 Tg in 2003 to 4915 Tg (0.1%/yr) in 2015.

The emissions of sea salt and desert dust were calculated on-line in C-IFS (Morcrette et al., 2009). The average total amount of emitted desert dust was 1489 Tg/yr, which is within the range of 1000–3000 Tg/yr given in a review by Boucher (2015). Schutgens et al. (2012) suggest larger desert dust emissions of 3244 Tg/yr using a data assimilation approach based on AERONET and MODIS observations. The median of the dust emissions for the year 2000 of the models compared in the AeroCom is 1640 Tg/yr (Textor et al., 2006) and they ranged between 500 and 4400 Tg/yr (Huneeus et al., 2011).

The total annual emitted sea salt amounted to 6038 Tg/yr, which is at the upper end of the 1000–6000 Tg/yr range given in Boucher (2015). The Aerocom models have a large spread of the sea salt emissions ranging from 3000 to 18000 Tg/yr with a median value of 6280 Tg/yr. More, recently Spada et al. (2013) report a range of 3888 Tg/yr to 8114 Tg/yr, when comparing different sea salt emission schemes. The observation based estimate by Schutgens et al. (2012) is 9073 Tg/yr which is the same value as the optimal estimate for seas salt with a dry diameter up to 10 µm by Grythe et al. (2014).

The mainly wind driven sea salt and desert dust annual emissions in C-IFS did not have a large year-to-year differences on the global scale. However, a significant positive trend of

0.4%/yr was detected in the sea salt emissions, which may require a more detailed investigation.

The average annual emissions of black carbon agreed well with the median value of the AeroCom models. The C-IFS emissions of organic matter and of the sulfate precursor, which includes SO_2 and DMS, were about 20% higher than the respective AeroCom medians but they were both well within the range of the individual models. The black carbon emissions decreased by -0.8%/yr which is consistent with the decrease in CO biomass burning emissions.

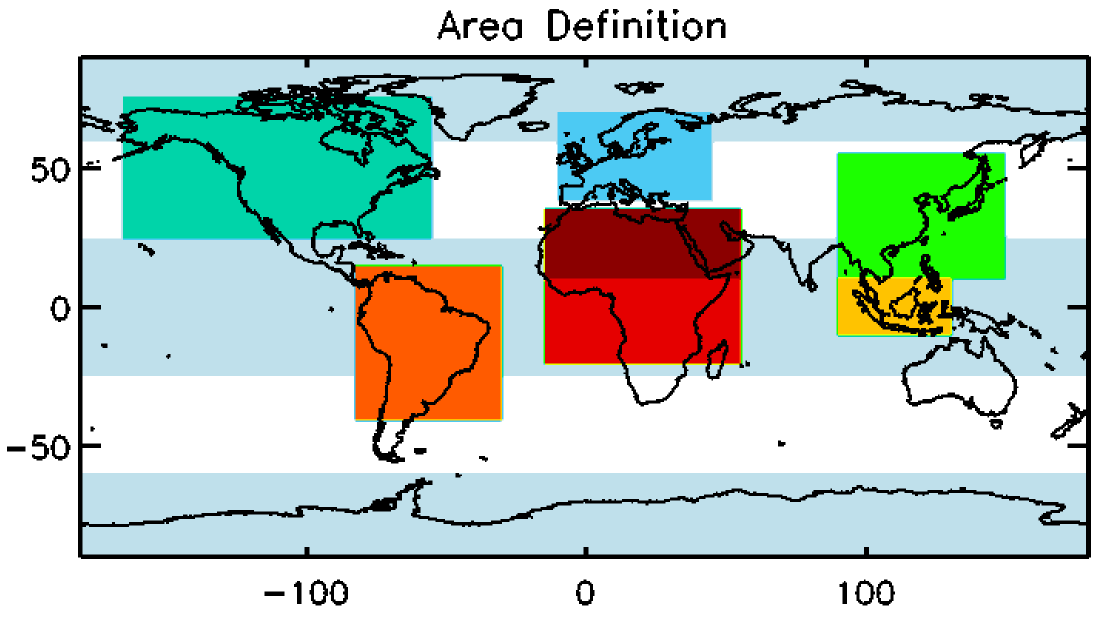

Figure S1 Area definition for North America (turquoise), Europe (blue), East Asia (green), South America (orange), Tropical Africa (dark red) and Maritime South East Asia (yellow) as well as Tropics, Antarctica and the Arctic (all light blue)(see Table 3 of the paper for coordinates).

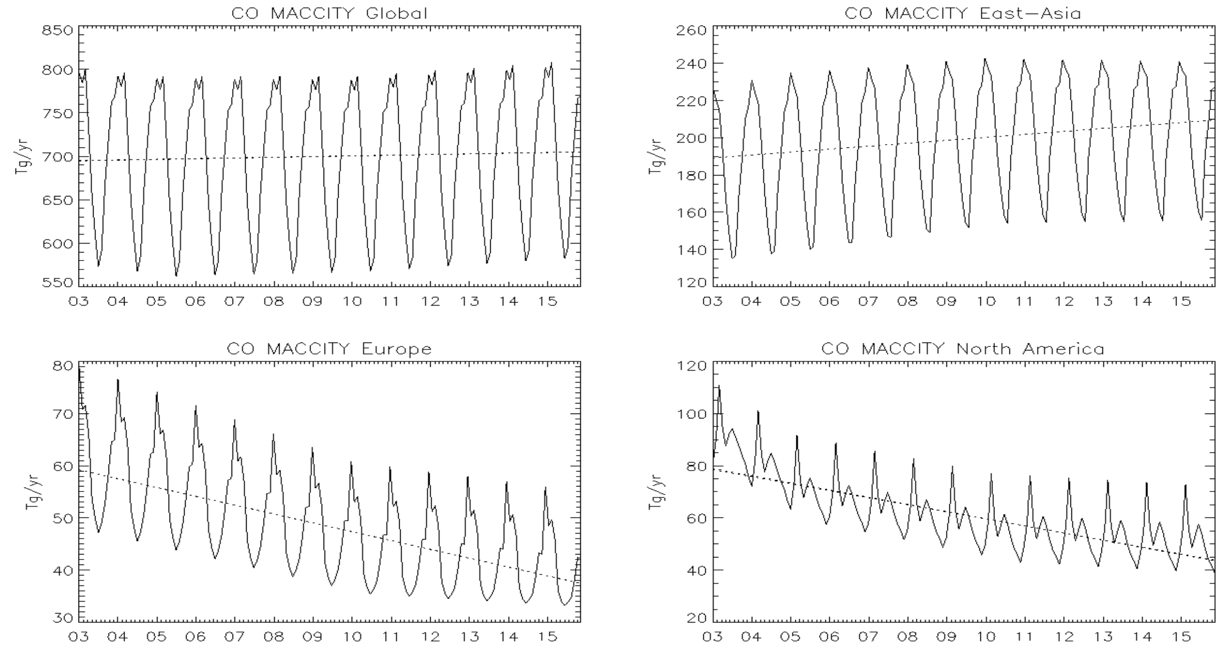

Figure S2 Monthly CO emissions from anthropogenic sources (MACCITY) for the whole globe and main source regions for the period 2003–2015.

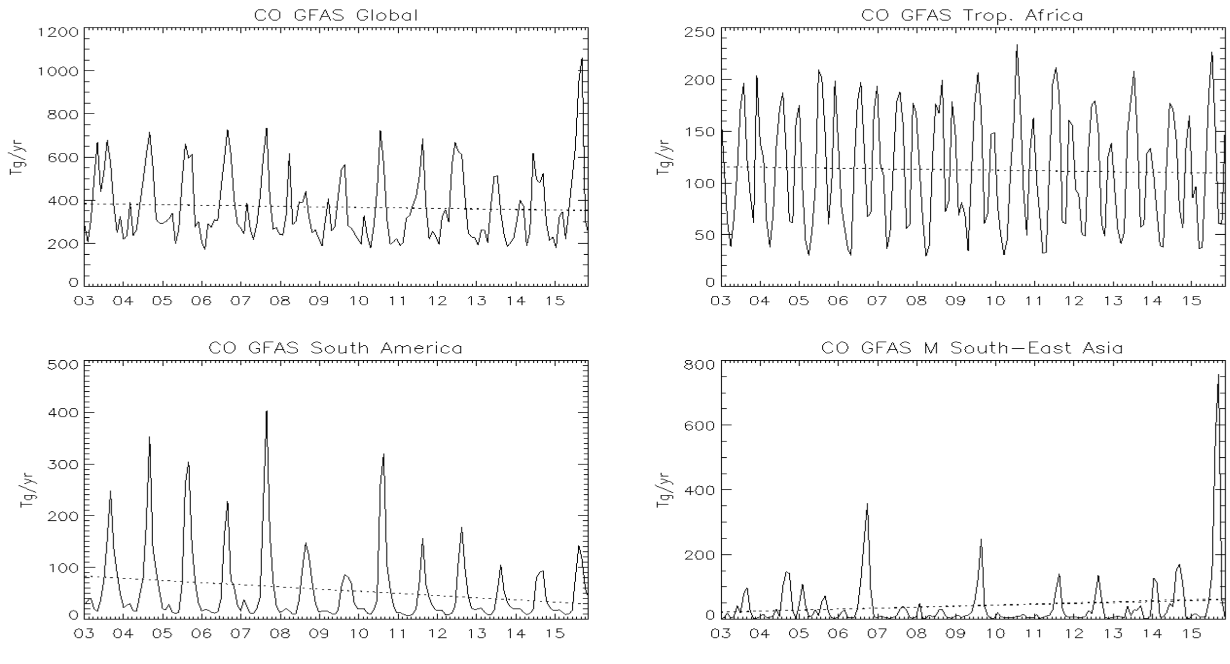

Figure S3 Monthly CO emissions from biomass burning (GFAS) for the whole globe and main source regions for the period 2003–2015.

Species	Area	Source	Mean (Tg)	Min (Tg)	Year of Min	Max (Tg)	Year of max	Significant (95%) linear trend (%/yr)
CO	Global	BB	368.9	293.1	2013	447.5	2015	
CO	Trop. Africa	BB	112.6	105.5	2014	119.7	2007	
CO	South America	BB	57.7	31.7	2013	88.4	2004	-7.4
CO	MSE Asia	BB	41.2	7.9	2010	136.2	2015	
CO	tropics	BB	267.4	204.4	2013	347.9	2015	
CO	Global	A	699.9	694.8	2005	710.7	2015	0.1
CO	East-Asia	A	198.7	185.6	2003	204.3	2015	0.8
CO	Europe	A	48.9	41.2	2015	60.6	2003	-3.5
CO	North America	A	62.2	51.2	2015	89.2	2003	-4.5
CO	tropics	A	316.4	301.4	2003	329.2	2015	0.7
NO	Global	A	81.5	79.6	2003	84.1	2015	0.4
NO	East Asia	A	20.7	17.0	2003	23.2	2015	2.5
NO	Europe	A	10.7	9.9	2015	12.0	2003	-1.7
NO	North America	A	13.5	11.9	2015	16.1	2003	-2.5
NO	tropics	A	24.3	22.5	2003	25.9	2015	1.1
C5H8	Global	B	574.6	518.0	2008	607.8	2004	
C5H8	East-Asia	B	32.6	29.7	2008	38.1	2010	
C5H8	Europe	B	6.8	6.2	2007	8.4	2010	
C5H8	North America	B	21.7	18.1	2009	24.5	2006	
C5H8	tropics	B	461.7	411.1	2008	493.3	2004	
SS1	Global	T	32	32	2003	34	2010	0.5
SS2	Global	T	2711	2696	2003	2883	2010	0.5
SS3	Global	T	3295	3277	2003	3504	2010	0.5
DD1	Global	T	114.9	113.1	2010	121.7	2015	
DD2	Global	T	459.5	452.2	2010	486.9	2015	
DD3	Global	T	919.1	904.4	2010	973.8	2015	
OM	Global	T	123.0	112.0	2013	150.3	2003	
BC	Global	T	11.4	10.8	2009	12.9	2003	-0.8
ASO2	Global	T	137.1	139.9	2003	141.4	2010	0.05

Table S1 Mean, minimum and maximum annual emissions (Tg), year of extreme and significant (95%) linear trend (%/yr) for different species and regions for the period 2003–2015. Sources categories are A: anthropogenic, BB: biomass burning, T: total, B: biogenic)