Supplement

Figure S1: AGAGE/ NOAA stations

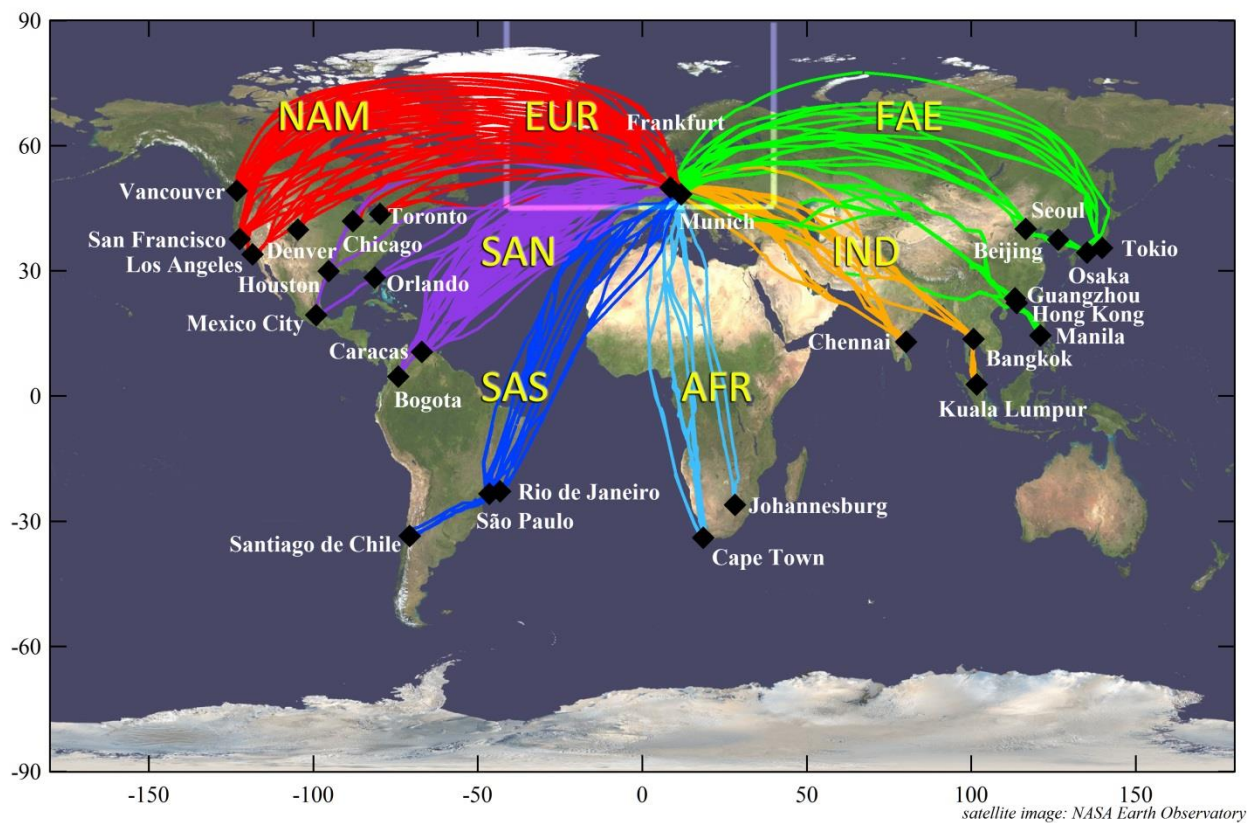



Figure S2: CARIBIC flights and destinations

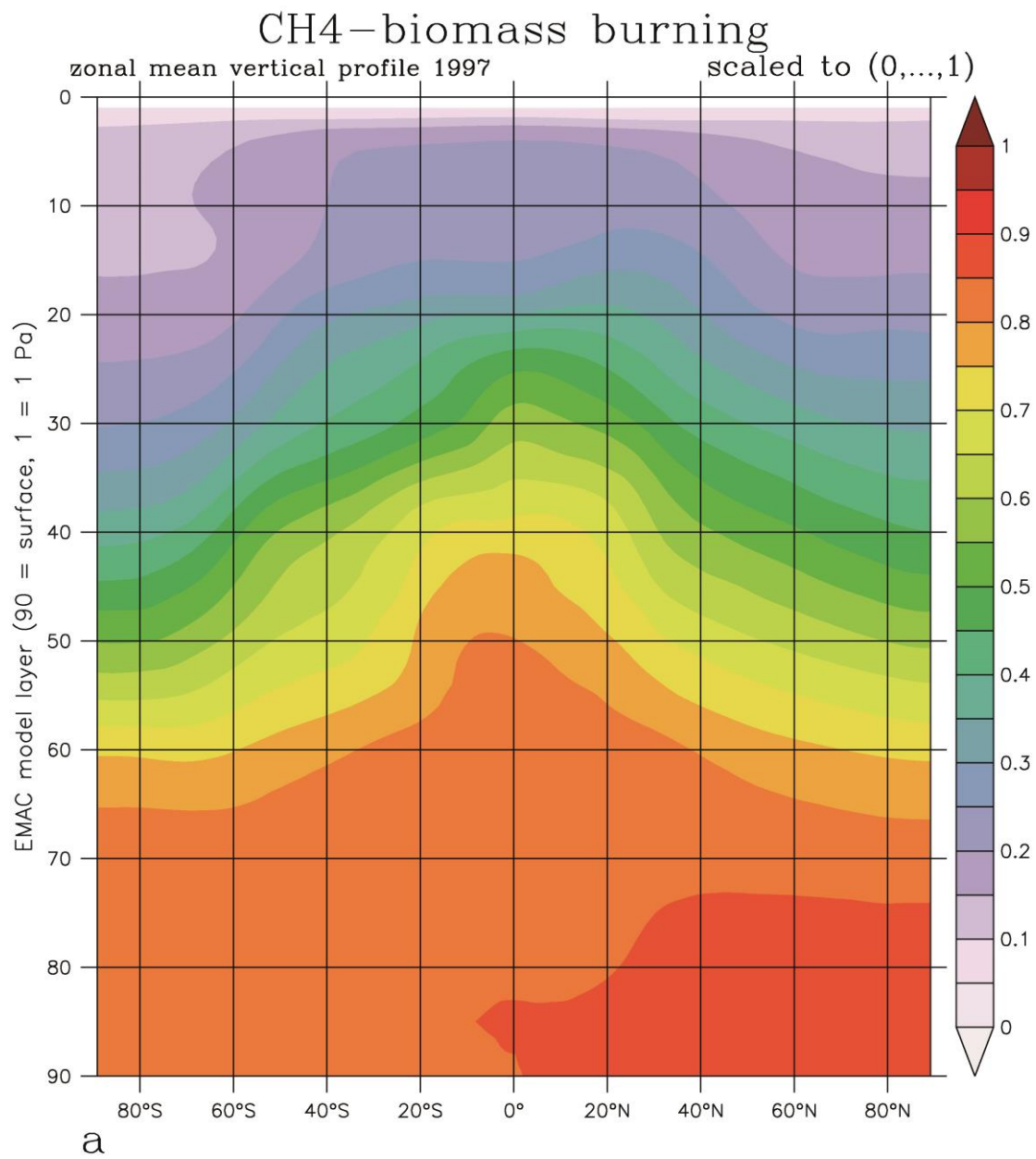



Figure S3:

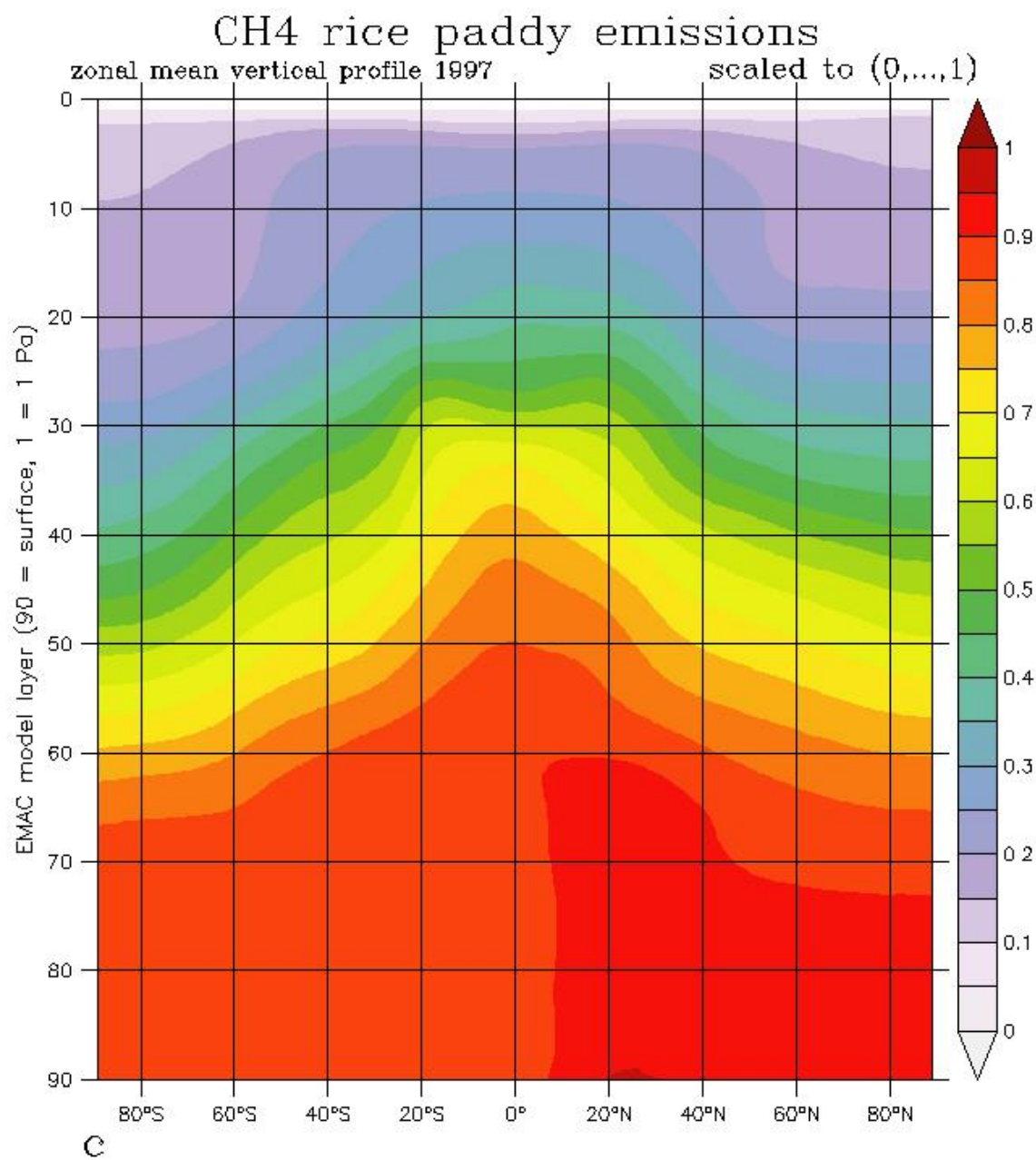
a: Zonal mean biomass burning CH_4 mixing-ratios in 1997 (scaled to 0 … 1).

b: same, but for tropical rice paddy released CH$_4$ mixing-ratios.

Figure S4:

EMAC CH_4 calculations (red) and CARIBIC-1/2 observations (blue) from 1997 through 2006 separately by sampling area.

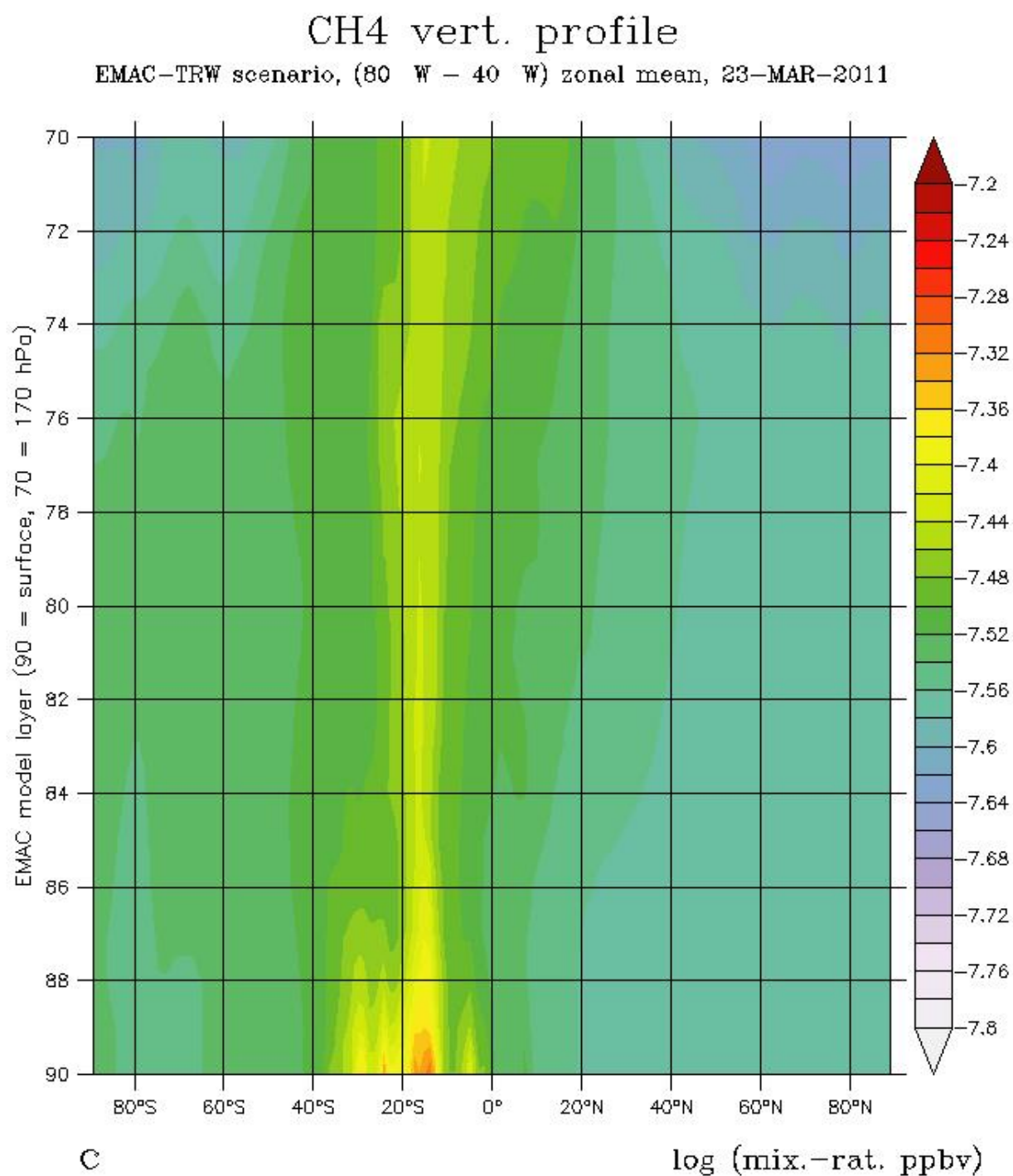



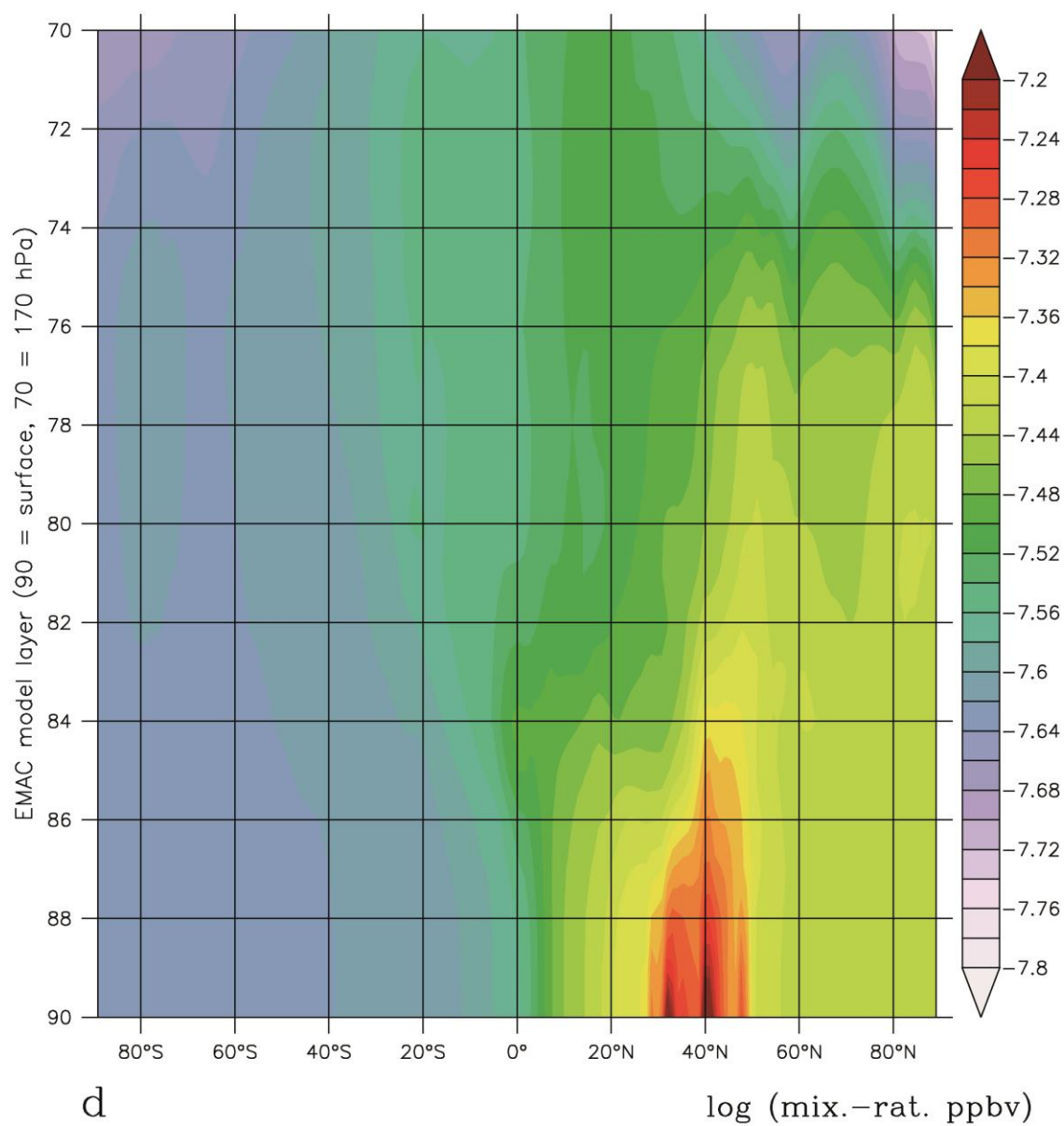
Figure S5:

a: Vertical methane mixing ratio distribution of tagged TRO emissions.

CH4 vert. profile

EMAC-SHA scenario, (120 W – 70 W) zonal mean, 23–MAR–2011

d

log (mix.–rat. ppbv)

b: Same as c, but for SHA emissions.

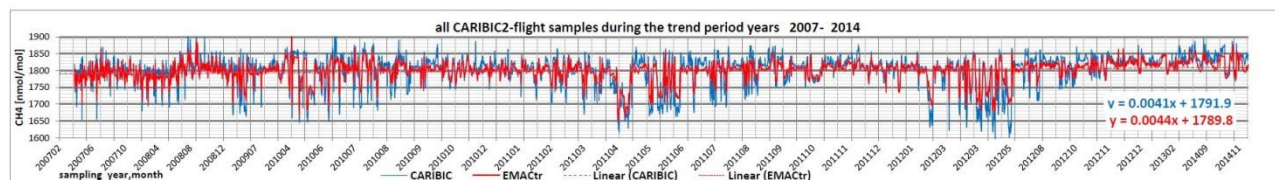
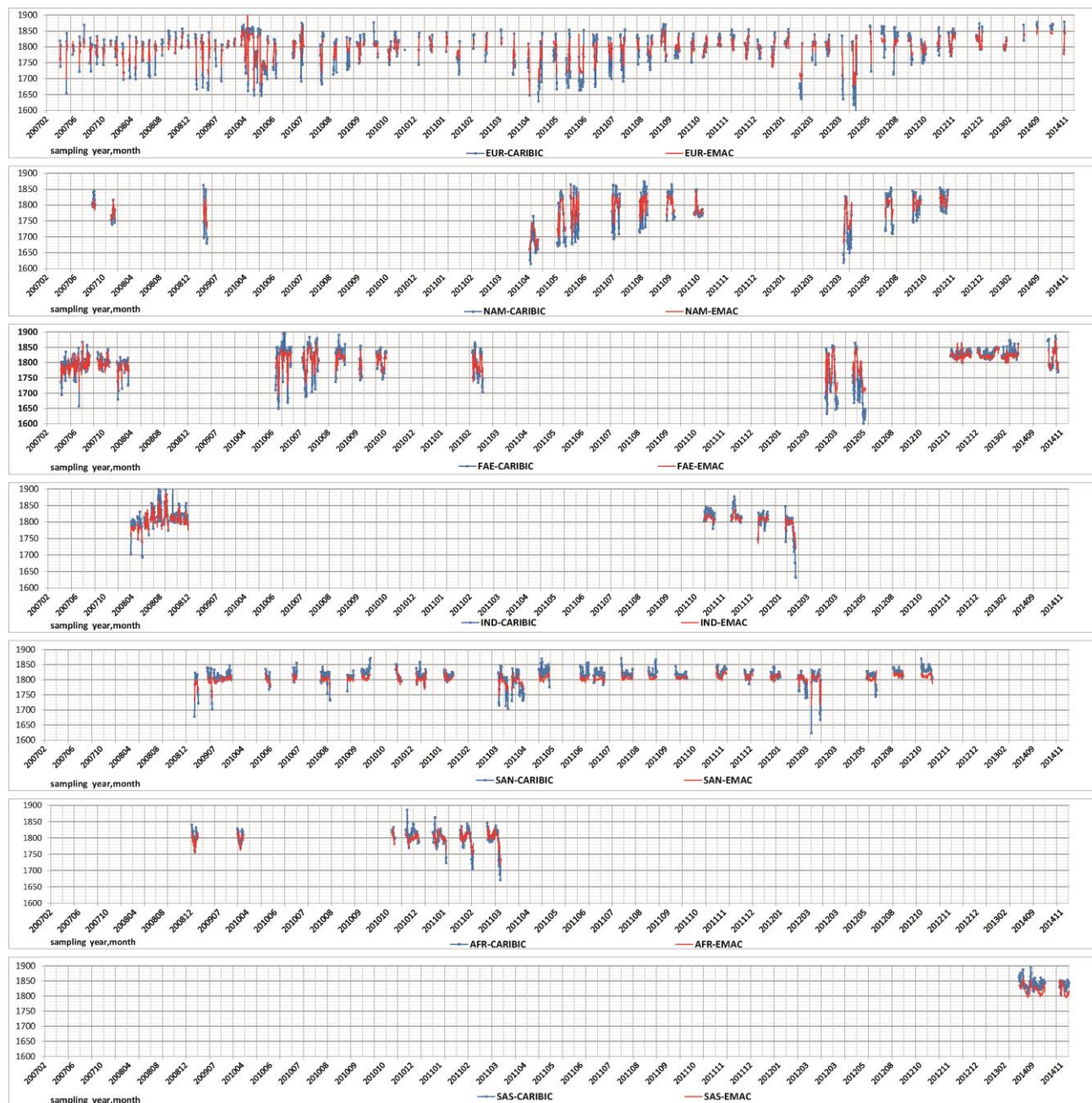



Figure S6

a: EMAC calculated CH$_4$, including trend, and CARIBIC-2 observations 2007 through 2014.

b: same as Fig. 6a but resolved by flight regions.

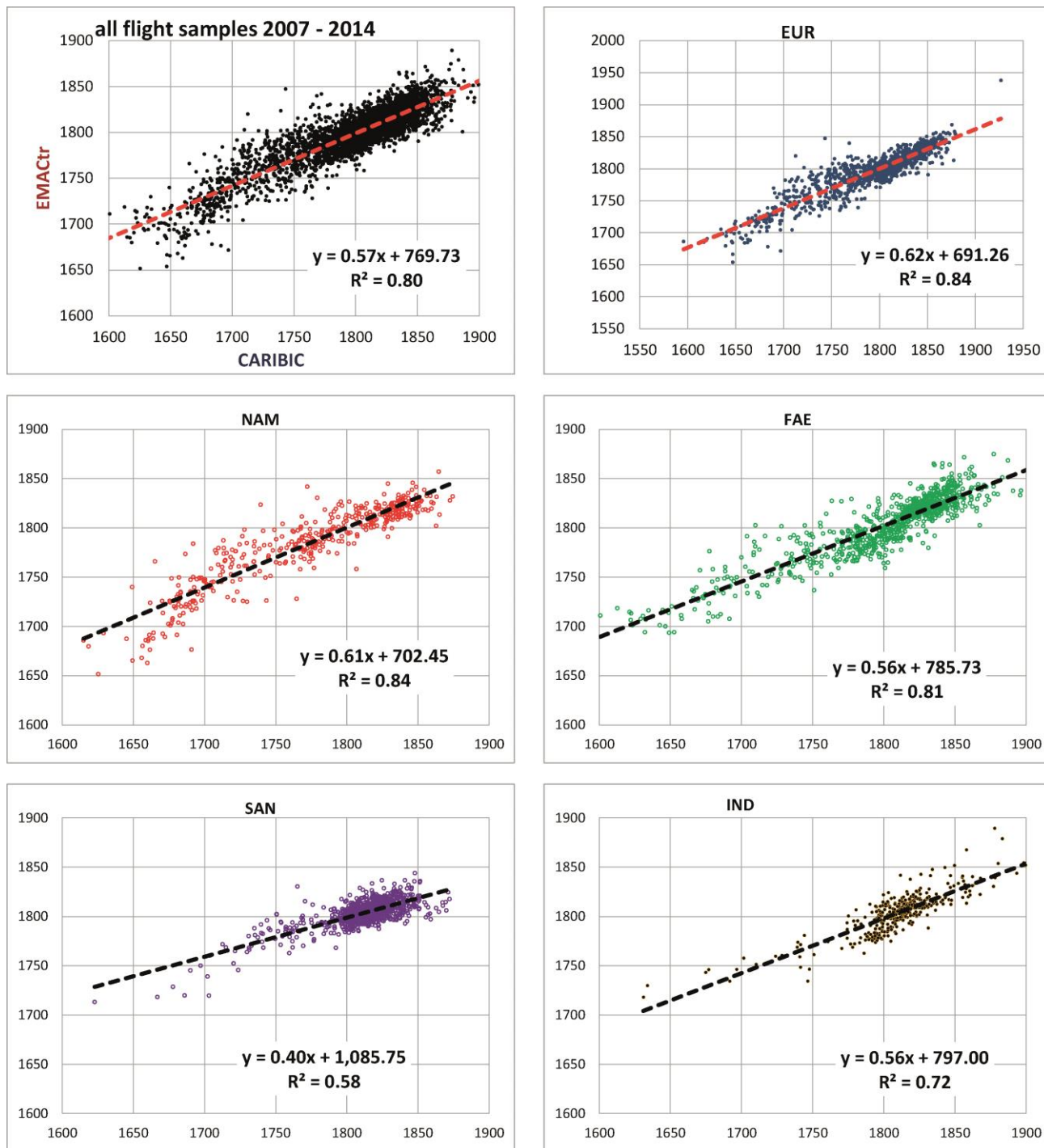



Figure S7: Linear regression between CARIBIC-2 samples and EMAC calculations for all trend period flights (2007 - 2014) and for flight regions with more than 300 samples.

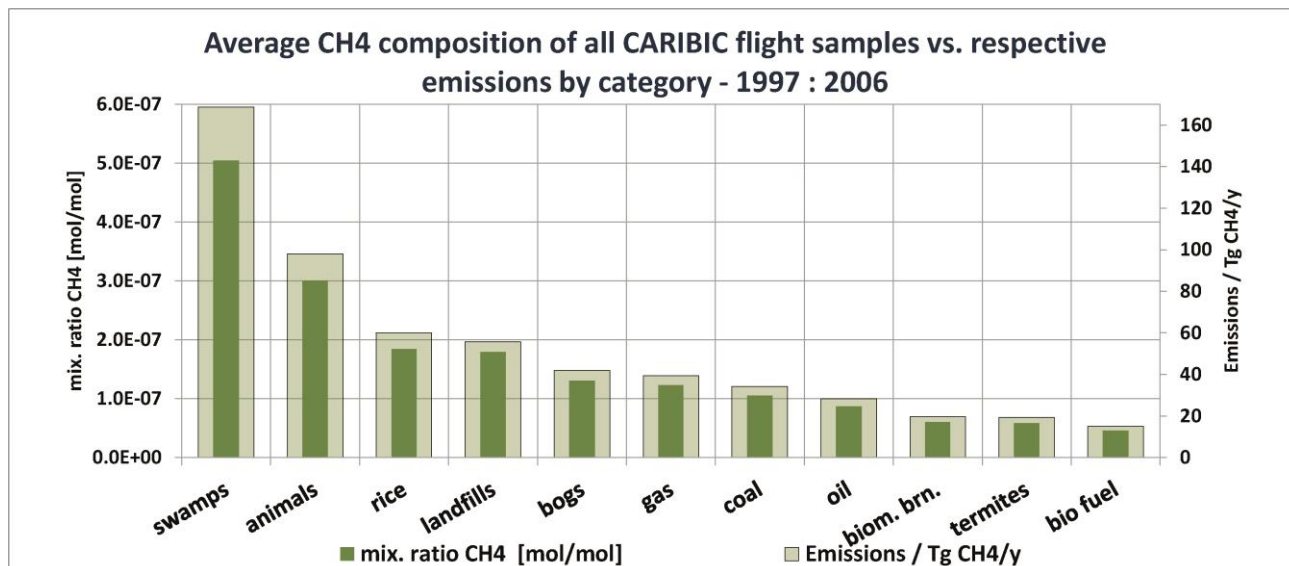



Figure S8. Average CH$_4$ composition of all CARIBIC flight samples 1997 - 2006, compared to respective emissions.