Supplement of

Atmospheric measurements of ratios between CO_2 and co-emitted species from traffic: a tunnel study in the Paris megacity

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Supplementary material:

<u>Mean diurnal cycles:</u>

Mole fractions vary significantly over the course of the day. The mean diurnal cycle (± 1-σ standard deviation) amplitudes are summarized in Table 1.

	Amplitude minimum	Associated standard deviation	Amplitude maximum	Associated standard deviation
CO_2	486.81	14.95	750.21	166.26
CO	429.30	144.27	2670.52	1595.44
NO	377.51	86.12	1284.46	732.97
NO_2	82.32	16.09	319.82	208.02
Benzene	0.38	0.18	2.67	2.89
Toluene	1.07	0.58	7.53	6.36
m&p-xylenes	0.23	0.12	1.72	1.84
o-xylene	0.14	0.08	1.25	1.45
n-propylbenzene	0.06	0.05	0.84	1.15
m&p-ethyltoluene	0.08	0.07	0.53	0.52
Ethylbenzene	0.14	0.08	1.02	1.18
Acetylene	0.73	0.39	5.96	6.36
Propene	0.63	0.49	4.63	4.67
Ethylene	4.15	2.82	8.55	8.70
i-pentane	0.82	0.67	7.96	9.77
n-pentane	0.51	0.33	4.84	16.05

5   **Table 1.** Mean diurnal cycle amplitudes and associated standard deviations. Amplitudes for CO2 are reported in ppm, all others are reported in ppb.

<u>Background discussion:</u>

Two options were considered to represent background levels: (i) nighttime concentrations or (ii) daily
10   concentrations out of the traffic peaks. Table 2 shows the averaged background concentrations considering these two options. Associated standard deviations are also mentioned. According to this table, we notice that background concentrations in the option (ii) are more variable because the standard deviations are higher than the ones of the option (i). During daytime out of the traffic peaks, vehicle density is high but vehicle speed is also high. Vehicles may bring outside air in the
15   tunnel, diluting the air in the tunnel.

	Option (i)		Option (ii)	
	Nighttime averaged concentrations	Associated standard deviation	Daytime averaged concentrations outside rush hours	Associated standard deviation
CO_2	495.92	23.46	555.85	26.64
CO	516.24	213.80	776.11	232.61
NO	445.42	91.35	839.46	119.82
NO_2	88.11	17.78	160.52	28.03

Benzene	0.48	0.24	0.67	0.21
Toluene	1.33	0.84	1.65	0.75
m&p-xylenes	0.25	0.15	0.38	0.19
o-xylene	0.16	0.10	0.25	0.12
n-propylbenzene	0.09	0.07	0.20	0.07
m&p-ethyltoluene	0.08	0.06	0.11	0.03
Ethylbenzene	0.15	0.09	0.22	0.09
Acetylene	0.97	0.75	1.32	0.90
Propene	0.74	0.49	1.79	1.61
Ethylene	6.1	3.91	7.79	4.55
Propane	1.71	0.19	3.00	2.98
i-pentane	0.94	0.77	1.59	1.51
n-pentane	0.57	0.33	1.13	1.56
n-butane	0.98	0.85	0.99	1.17
i-butane	0.82	0.92	1.22	1.85

Table 2. Averaged background concentrations and associated standard deviations considering the two options. Mole fractions for CO2 are reported in ppm, all others are reported in ppb.

20   Nighttime concentrations may be less representative of daytime background levels because vehicle density is very low, but they are less affected by the traffic source that we want to isolate. Furthermore, they look more stable.

The ratios between co-emitted species and CO_2 are evaluated correcting compound concentrations with background concentrations of option (i) (in the main paper) and of option (ii). We obtained exactly the same results for coefficients of determination and ratios $\Delta Species/\Delta CO_2$.

25   The ambient air outside of the tunnel was also sampled during the campaign but is not regularly available for most of the species. These data is available every 4h for CO and CO_2. We linearly interpolated this ambient data every 4h to obtain a 1 minute step ambient dataset. Fig. 1 displays this time series.
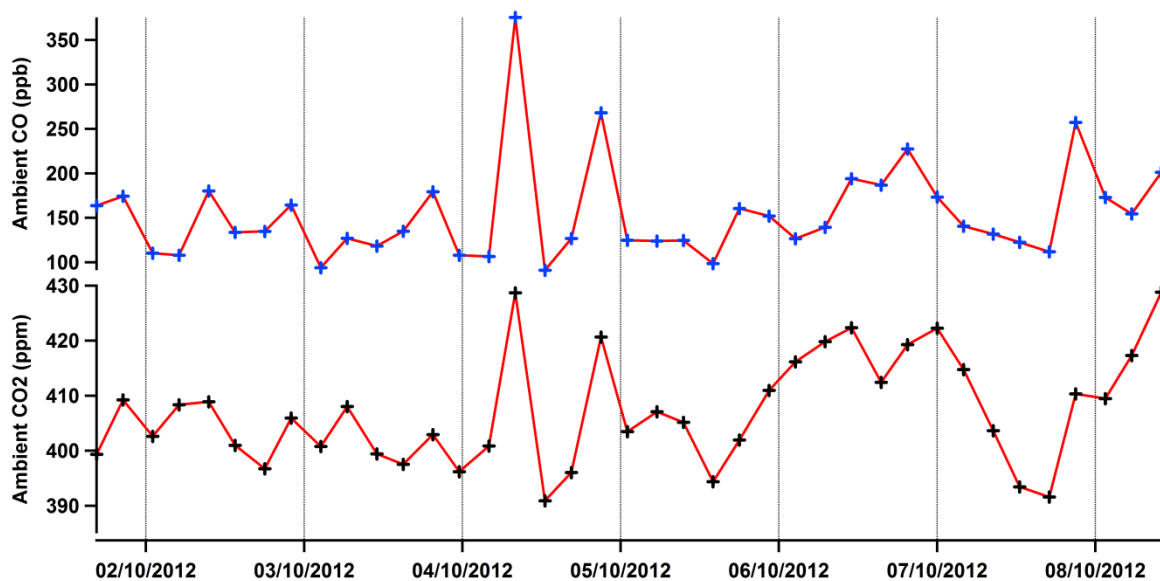
30 **Fig. 1.** (i) Temporal variation of the CO ambient concentration (outside of the tunnel). Blue points represent real measurements and red lines the linear interpolation. (ii) Temporal variation of the CO_2 ambient concentration (outside of the tunnel). Black points represent real measurements and red lines the linear interpolation.

35 Then, we corrected CO and CO_2 concentrations in the tunnel during traffic peaks using the ambient interpolated concentrations as background level. As in the main paper, we evaluate the correlation between ΔCO and $ΔCO_2$ and the ratio in this case. Fig. 2 shows this scatterplot. We notice that the coefficient of determination is the same than in the two other options and the ratio ΔCO to $ΔCO_2$ is nearly the same (0.2% of difference).
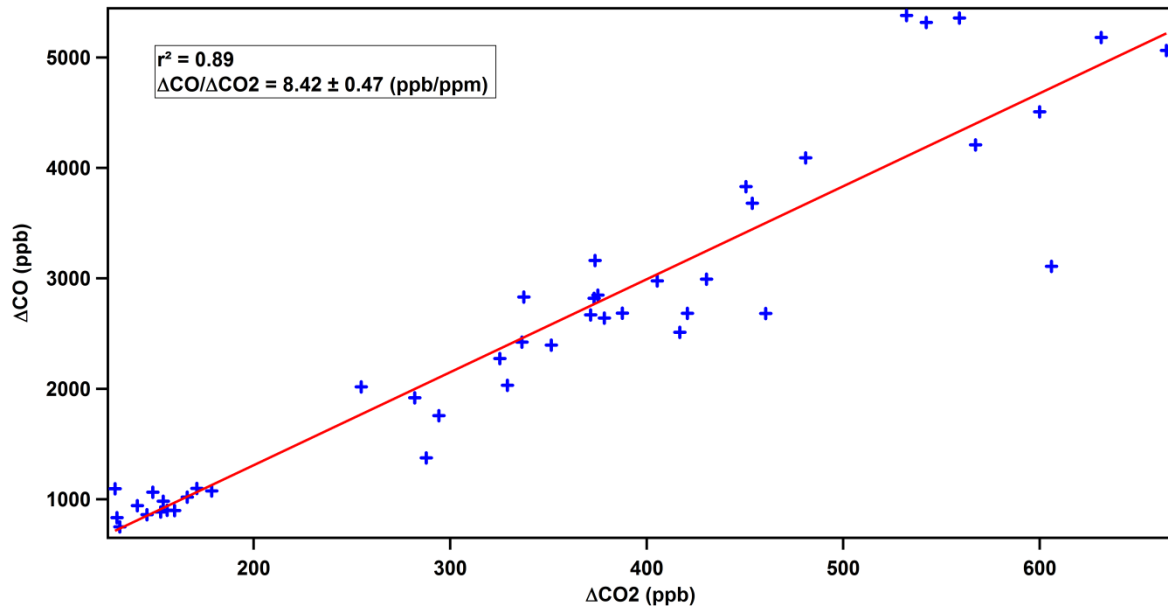
40 **Fig. 2.** Correlation between ΔCO and $ΔCO_2$ using ambient concentrations as background levels. Red line represents the linear regression fit between ΔCO and $ΔCO_2$.