Supplementary Information

Assessing the Optimized Precision of the Aircraft Mass Balance Method for Measurement of Urban Greenhouse Gas Emission Rates Through Averaging

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Text S1: Uncertainties for a single MBE

Two different types of uncertainties are identified here (Heimburger et al., 2013): *i*) uncertainties associated to concentrations, wind speed, temperature and pressure measurements (δER_{cal}), and *ii*) variability of the calculated emission rates due the growth of the convective boundary layer (CBL) (δER_{exp}). The absolute overall uncertainty of a single emission rate (ΔER) can thus be divided in two distinct terms (Eq. 3): the "calculation" uncertainty (δER_{cal}) and the "experimental" uncertainty (δER_{exp}), respectively.

$$\Delta ER = k * \sqrt{\delta ER_{cal}{}^2 + \delta ER_{exp}{}^2} \quad \text{Eq.3}$$

where k is the coverage factor set to 2 to obtain an expanded uncertainty representing a confidence level of 95% (Feinberg, 2009).

δER_{cal} accounts for the uncertainties of the terms appearing in Eq.1, i.e. uncertainties of the enhancement (E), perpendicular wind speed (U), pressure (P) and temperature (T). Because E, U, P and T are independent from each other, the general uncertainty equation for this term can thus be written as follows:

$$\delta ER_{cal} = F * \sqrt{\left(\frac{\delta E}{E}\right)^2 + \left(\frac{\delta U}{U}\right)^2 + \left(\frac{\delta P}{P}\right)^2 + \left(\frac{\delta T}{T}\right)^2} \quad \text{Eq.4}$$

The relative uncertainty of enhancement of ($\delta E/E$) depends on *i*) the uncertainty of the linear fit of the in-flight calibration curve calibration curve (σ_{calib}) for CH_4 and CO_2 (see manuscript section 2.2), and *ii*) the uncertainty of the linear function used to define the background (σ_{bg}).

As described in the manuscript (section 2.2), CO was not calibrated in-flight but before and after the field campaign at NOAA/ESLR. However, the three standard gas tanks used for in-flight calibrations for CH_4 and CO_2 did contain constant, albeit unknown, concentrations of CO, thereby allowing us to measure the precision of our CO measurements in-flight. During each in-flight calibration the variance in the average CO concentration (one standard deviation of the average) was found for each calibration tank. Although the average CO concentration differed between tanks, the *variance* across all three tanks was the same (at the $\alpha = 0.01$ level), with an average of 1.2 ppb (~1% relative variance for CO measurements made herein). This variance, then is used for σ_{calib} for CO in the determination of ($\delta E/E$) for CO.

σ_{bg} does not account for the uncertainties introduced by background determination, but only the standard deviation of residuals between the observed background concentrations (recorded on the edges of the downwind transects) and the modeled background concentrations (linear fit) used as background. The standard errors of σ_{calib} and σ_{bg}, correspond to the standard deviations of the residuals from the linear fit function:

$$\sigma = \sqrt{\frac{\sum r^2}{N-P}} \qquad \text{Eq.5}$$

where r is the residual (error of prediction, r = measured data subtracted by predicted data), N is the number of measurements used to build the linear regression, and P = 2 corresponds to the number of estimated parameters (slope and y-intercept) used to estimate the sum of squares. For each flight, δER_{cal} was found to range from 0.01% to 0.30% for CO_2, CH_4 and CO (Table S3).

The relative uncertainty of perpendicular wind speed ($\delta U_/U$) is defined as the propagation of *i*) the standard deviation of the mean wind speed from measurements recorded on the downwind transects only (corresponding to data only used for the emission rate calculation) and *ii*) the measurement precision of the BAT probe for horizontal wind speed measurements estimated at 0.4 m/s (Garman, 2009), divided by the averaged value of wind speed for the considered flight. $\delta U_/U$ varies from 11% to 19% depending on the flight (Table S3). The relative uncertainty of temperature ($\delta T_/T$) is equal to the averaged relative error between the temperature recorded by the microbead thermistor (used in the emission rate calculation) and the temperature from the thermocouple for each flight. This uncertainty varies from 1.7% to 2.5% (Table S3). The relative uncertainty of pressure ($\delta P_/P$) is defined as the averaged relative error between observed pressure and theoretical barometric pressure calculated for each data point recorded during the downwind transects of a flight. $\delta P_/P$ varies from 0.04% to 1.7%. Uncertainties in the enhancement, pressure and temperature are small compared to uncertainties in windspeed, which ultimately drive the calculation uncertainty (δER_{cal}) on the emission rates.

For our nine MBEs, the top of the CBL (z_i) grows from the beginning to the end of the experiment. VPs are performed before and after flying horizontal transects (upwind and downwind of the city) in order to quantify this growth during the experiment. LIDAR observations are used when only one or no VP was performed. To estimate the sensitivity of the CBL growth on the final emission rates (ER, Eq.2), we integrate all the measurements of each

MBE (F_{ijk}, Eq.1) from the surface up to the lowest, and up to the highest z_i observed for each experiment using the kriging approach. The relative difference between the two emission rates calculated with the two z_i limits (lowest and highest values) is taken as the experimental uncertainty (Table S3). This uncertainty (δER_{exp}) and "calculation" uncertainty (δER_{cal}) are then propagated following Eq.3 to obtain an estimate of the overall emission rate uncertainty for a single MBE (ΔER) (result on Table 1, main text). We are aware that the overall uncertainty does not take into account all the factors which might influence the emission rate values and highlighted in Cambaliza et al. (2014), such as the entrainment of the free troposphere, which might occur on the edges of the city (where backgrounds are defined). This uncertainty calculation also doesn't take into account variability in the emission rates depending on the background determination and the choice of the kriging method, which can represent a significant part of the uncertainty. This overall uncertainty is considered as the minimum emission rate uncertainty for a single flight.

References:

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