Supplementary information on kriging approach

Kriging is an interpolation technique, used in this study to calculate mole fractions and wind speeds on a regular altitude-latitude grid downwind of London (e.g. Figs 6d, 6e and 6f) based on a weighted average of sparse measured data from the aircraft sampling (e.g. Figs. 6a, 6b and 6c). In simple cases where data vary over the same length scale in each dimension, the coefficients used in this weighted average would only depend on the geometric distance of each measurement point from a given grid point. However, in this case we expect the data to vary over a shorter length scale in the vertical dimension than the horizontal dimension (as is typical within the atmospheric boundary layer). In order to use kriging to interpolate this data, we first have to rescale distances in each dimension so that we can calculate a variogram which depends only on a single distance parameter.

We followed the same approach in all four cases: kriging of CO_2 mole fraction, CH_4 mole fraction, CO mole fraction and perpendicular wind speed. We used a modified version of the MATLAB-based software EasyKrig (© Dezhang Chu and Woods Hole Ocean Institution), which automatically normalises the distances in each dimension by the total distance range in that dimension as the default option. Starting with this default scaling, we plotted a 2D experimental variogram (i.e. using vector displacement rather than distance) and iteratively rescaled the vertical distances to reduce the anisotropy evident in this variogram.

Having set the relative distance scales in each dimension, we then plotted a 1D experimental variogram based on the (rescaled) distances between the measured data points. We selected a variogram model based on this experimental variogram, with a particular focus on good representation of the experimental data at small separation distances. The initial variogram parameters were determined by a non-linear least squares fit, using a Levenberg-Marquardt algorithm, to the experimental variogram. These parameters were then optimised by iteratively kriging the data, calculating the Q1 and Q2 validation criteria, and adjusting the parameters according to these criteria.

The kriging itself was performed using a moving neighbourhood approach. Rather than considering all measurements when calculating the kriged output for a given grid point, only measurements within a radius equal to 0.1 of the full length scale (i.e. the rescaled length of each axis) were considered, up to a maximum of the nearest 180 points. This moving neighbourhood approach was adopted due to the computational constraints associated with kriging a dataset containing over 7000 points (particularly for the calculation of the Q1 and Q2 validation criteria). The assumption that only local measurements exert a strong influence on each output grid point can be expected to be reasonable in the context of both in-plume and background data here.

The model variogram parameters for each of the four kriged datasets are given in Table S1. The two variogram models used here were:

Linear: $\gamma(h) = \theta.h + \gamma(0)$

Gaussian: $\gamma(h) = \theta\left[1 - exp\left(-\left(\frac{h}{L}\right)^2\right)\right] + \gamma(0)$

Here the lag h represents the rescaled distance between two points and γ is the normalised variogram:

$$\gamma(h) = 1 - \frac{R(h)}{R(0)}$$

where $R(h)$ is the covariance function for points separated by h.

	CO_2	CH_4	CO	U
Model type	Linear	Linear	Linear	Gaussian
$\gamma(0)$	0.02	0.008	0.11	0.036
θ	2.48	5.592	3.09	0.069
L	-	-	-	0.007

Table S1: Model variogram parameters for each dataset