Use of emission ratios for the stationary Breathe London network

1. Introduction

The concept of emission ratio (ER) or emission index (EI) is extremely well embedded in the air quality monitoring and modelling frameworks. The bulk of this work has either used traditional reference instrumentation or has looked at tailpipe emissions, and there has been significant discussion of laboratory verses on road EI or ER estimates. There has been work, including by the partners of this project (Popoola et al., 2018), which has used low-cost sensors, essentially of the type used here, together with modelling to evaluate and compare ER estimates with those from models. In that sense the methodology itself, including the use of low-cost sensors, is well proven in the peer reviewed scientific literature.

Primary pollutant emissions (NO_2, NO_X, fine particles) are primarily associated with combustion sources and therefore with CO_2 emissions. The ratio of pollutant-to-CO_2 from an emissions source is the emission ratio (or emission index) and is generally distinct for each particular vehicle type or combustion source (traffic, domestic etc.). Knowledge of the emission ratio is thus a key additional test of an emission inventory, which is key information required for policy intervention decisions.

The inclusion of CO_2 measurements in data collected by each Breathe-London (BL) AQMesh pod (and, although not discussed here, the Google Street View cars) therefore enables the determination of emission ratios. However, this is only possible if the local emission signals can be separated from the non-local component (i.e. CO_2 signals of 10-50 ppm from a background of 400ppm, and the equivalent for each pollutant), which in turn is only possible if the network has high time resolution measurements (see network calibration appendix), as is the case in Breathe-London for both the static and mobile measurements. Emission ratios from stationary and mobile sources provide complementary information.

2. AQMesh Analysis

2.1 Illustration of methodology

This method for deriving emission ratios from stationary sensors has been previously used with low cost sensors for a study by the Cambridge Group at Heathrow Airport (Popoola et al., 2018.)

Figure 1., below, taken from Popoola et al. (2018) show how emission ratios can be used. The figures show NO and NO_2 emission indices derived from measurements which are using essentially the same technology as the AQMesh developed by the Cambridge group for Heathrow Airport. The ratios derived in that project were obtained in exactly the same way as for the Breathe-London project, which was first to apply scale separation algorithms across the network

to separate local from non-local pollutant concentrations, and then to take the local components of NO, NO_2, and CO_2 mixing ratios to derive the scatterplots typical of those shown below. In each figure the gradient represents the average emission ratio for that period and location in appropriate units.

Also shown in each figure are the equivalent emission ratios calculated using the ADMS model (also used in Breathe London) for comparison. In this case the aircraft type mix was included explicitly, leading to a range of emission ratios for NO evident in both measurements and model, but with a higher degree of uniformity for the NO_2 emission ratio (again in both measurements and model).

Measuring the emission ratios is a key additional and direct test of the emission inventory which is used in the ADMS model over and above simple knowledge of pollutant concentrations and is significantly less influenced by model dependent features such as atmospheric dispersion and treatment of the planetary boundary layer and, of course, is essentially independent of meteorological variations.

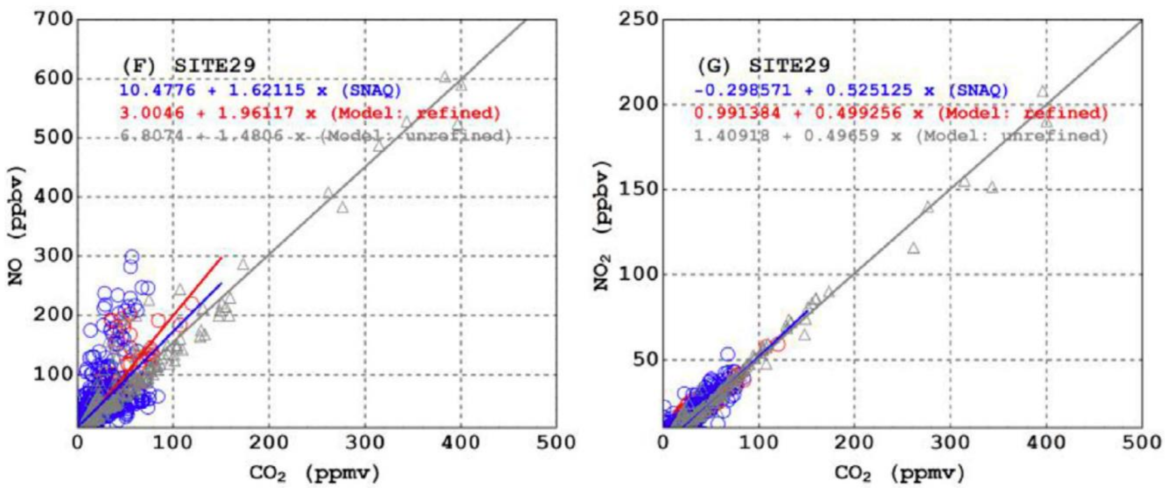

Figure 1. *Scatterplots of local components of NO and NO_2 against CO_2 used for emission ratio determination, at a selected site at Heathrow Airport. Also shown are model calculations (From Popoola et al., 2018).*

2.2 Preliminary analysis of Breathe-London static network results.

Analysis of emission ratios from the Breathe-London project remains at a relatively preliminary stage, in part because of the need to first apply the experimental calibration approach to the CO_2 sensors in the network, but also because issues such as the impacts of the O_3 cross interference on NO_2 sensors are still in the process of being promulgated through the data analysis methodology. All data is, however, archived for future analysis and interpretation. Nevertheless, some preliminary results are shown below, for the periods April to August 2019 and for the entirety of 2019.

Following the approach described above, emission ratios were derived for 88 sites across the network which satisfied QA/QC requirements.

In **Figure 2** are shown (left panel) monthly average network average NO_x values for the BL and LAQN (London Air Quality Network) networks (88 and 86 sites respectively). A clear and largely expected annual pattern is seen, with higher values in the winter months as expected due to meteorological conditions leading to generally lower boundary layer heights. There is generally good correspondence between the two networks, although as the individual sites of the two networks are not co-located and represent different mixes of roadside, kerbside and urban background sites, some differences are to be expected. **Figure 2** (right panel) shown the equivalent emission indices calculated from the Breathe-London static network data, in this case aggregated by ULEZ (21 pods) and non-ULEZ (66 pods) locations. Note, LAQN network do not have CO_2 and are reported as hourly averages so cannot be used for ER analysis.

The key inferences that can be made from this figure are that while there are some fluctuations, the NO_x emission index remains largely unchanged throughout the year, certainly with no systematic wintertime perturbation, which would be consistent with little seasonal change in the traffic mix, and a very clear indication that the use of emission indices removes the effects of meteorologically in induced fluctuations in pollutant levels. There are some indications that the non-ULEZ sites appear to have emission ratios systematically lower than ULEZ sites, indicative of differences in traffic mix in the two regions. Overall, it is clear that the use of emission indices provides an extra dimension of interpretation of air quality levels compared to information from the traditional network.

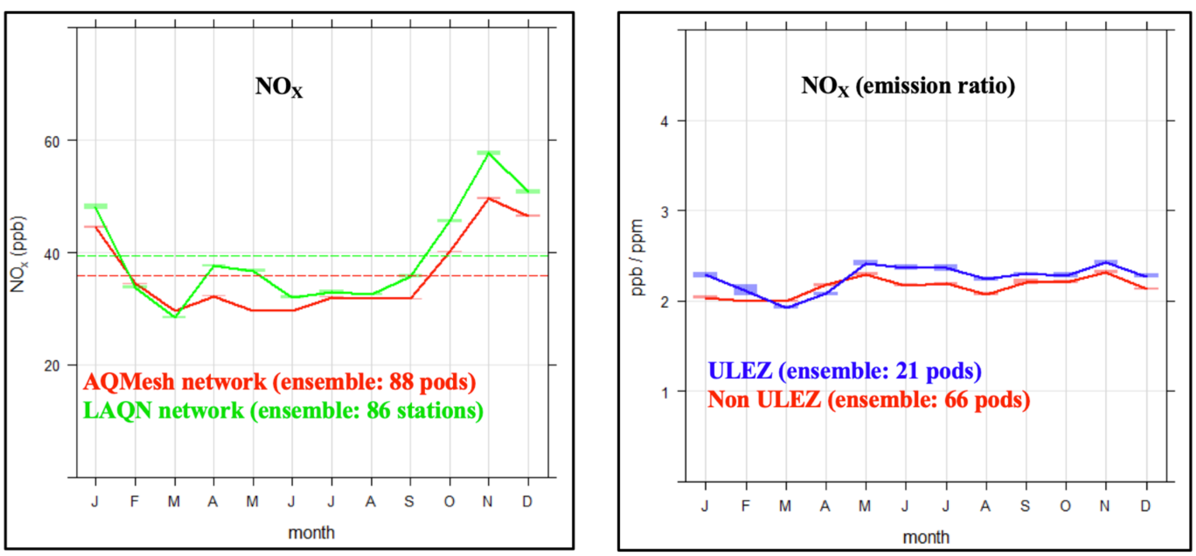

Figure 2. *Left: Network average statistics derived from the BL and LAQN London networks by month for 2019. Right: emission indices from the BL network split by ULEZ and non-ULEZ by month for 2019. Taken from the BL-AQEG/Defra submission. See text for more details.*

In **Figure 3** below are shown how the emission indices vary both as time of day and day of week across all BL static sites (top row), urban background sites (middle row) and taking only kerbside and roadside sites (bottom row). A clear diurnal pattern is seen in all cases, with differences evident (lower EI ratios) on weekends, and between all three categories of sites. The data are also disaggregated by ULEZ/non-ULEZ, with significant differences observed particularly for the urban background sites, and interestingly a significant morning enhancement in the non-ULEZ kerbside and roadside sites (bottom row) suggestive of high emission vehicles entering the ULEZ from outside London at these times.

A thorough evaluation of these results requires detailed data on the vehicle fleet mix by time of day at monitor locations, however, this was beyond the scope of this phase of the BL project. Overall, however, our expectation is that the variability in NO_x:CO_2 ratio will reflect changes in the traffic mix (different vehicle types have different emission ratios, as shown in the table (Table 1) of factors used in the CERC ADMS model).

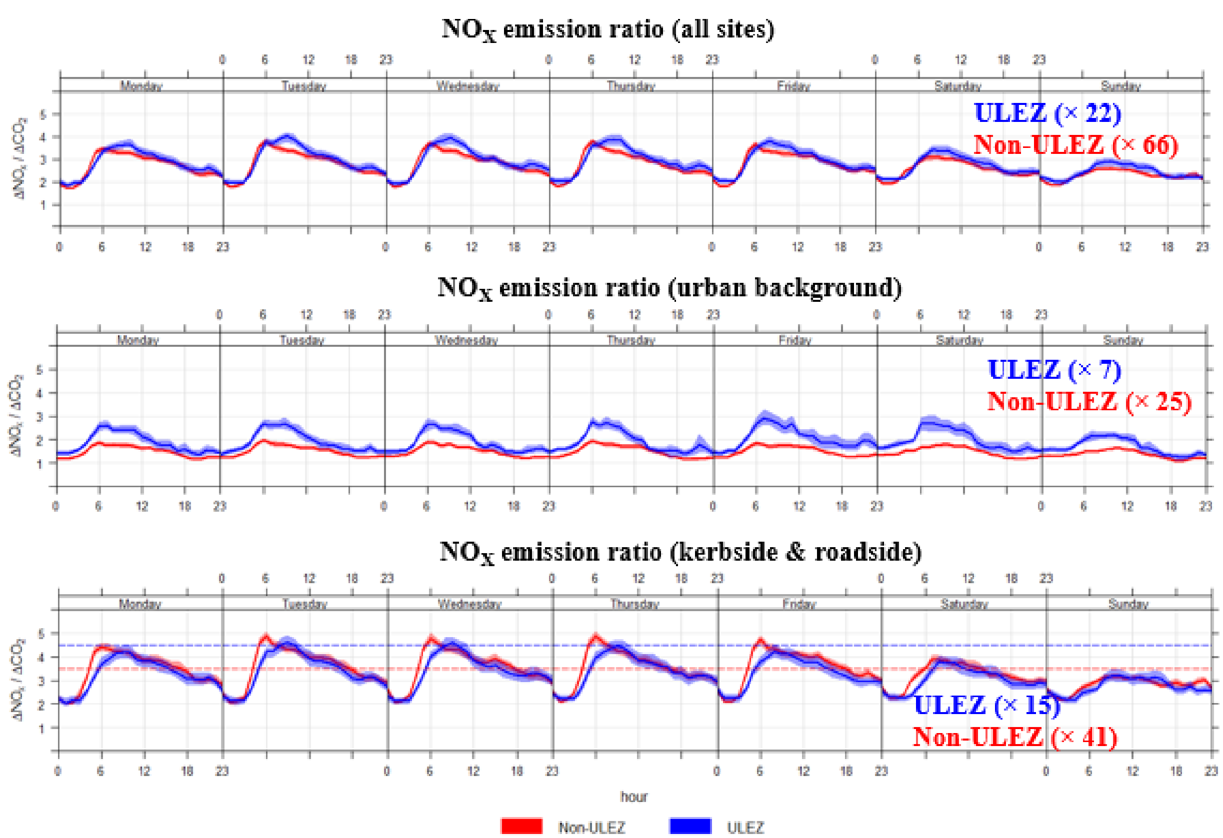

Figure 3. *Time series of NOx emission indices across the Breathe London network as a function of time of day and day of week. The data are disaggregated by ULEZ and non-ULEZ, and by site type (see text for further details).*

Emission source (vehicle type)	NOx EI (g/kg)
Car (Petrol)	0.51
Car (Diesel)	3.4
Taxi	3.2
LGV	3.3
Rigid HGV	3.5
Articulated HGV	3.2
Bus	4.1
Motorbike	0.58

Table 1. *Emission factors from CERC ADMS model, showing variations in emission ratios by vehicle type.*

3. Conclusions and caveats

Firstly, we note that, albeit at a relatively preliminary phase of analysis at this stage of the BL project, the estimation of emission indices or emission ratios across an air quality network represents an important step change in the capability of determining pollutant sources from measurements and models, in defining suitable policy interventions, and in quantifying the effectiveness of those interventions, accounting for meteorological effects in essentially real time.

Secondly, we note that while this study relates to Greater London, as with the concept of a low cost sensor network, and subject to the caveats raised elsewhere in this document, the methodology shown in preliminary form here is readily replicable elsewhere and therefore has relevance beyond this project.

This work is, however, incomplete, and a key component going forward will be to provide an in depth evaluation of the emission ratio approach, extending it to PM, and including an assessment of the likely performance of the method in air quality environments which may differ significantly from those found in London.

There are some specific issues which, although they require caveats at this stage do not represent fundamental limitations on the approaches outlined above:

1) As has been discussed elsewhere, the NO_2 sensors specific to this project have shown themselves to have a time-dependent O_3 cross interference which requires correction prior to EI evaluation. This work is in progress, although since NO_x is predominantly due to NO, at least in this dataset, the inclusion of this effect is expected to be relatively minor.
2) In the configuration used in this project, the CO_2 sensors have shown periodic baseline shifts which affect the absolute CO_2 measurements. The evidence is that the local contributions are only affected in a minimal way, so that the emission index calculations which rely on the local concentrations are not influenced significantly. Some additional work is needed to minimise this effect if absolute CO_2 levels are to be derived reliably.

Finally, we note that significant additional benefit from the measurement of emission indices can be obtained by integrating the measured emission indices with traffic information (vehicle type, age and mix). This additional step has not been attempted within this project.

References

1. Popoola, O. A., Carruthers, D., Lad, C., Bright, V. B., Mead, I.M., Stettler, M., Saffell, J. and Jones, R.L. (2018). The use of networks of low cost air quality sensors to quantify air quality in urban settings. Atmospheric Environment 194, 58-70. https://doi.org/10.1016/j.atmosenv.2018.09.030