Roadside air quality trends in London – identifying the outliers Part 1

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Summary

A large number of policy initiatives are being taken across London, the UK and the EU to improve air quality. These initiatives are expected to have a direct impact on the air quality in the whole of London but especially alongside busy roads. The objective of this study was to identify and characterise those roadside locations in London that have experienced the greatest changes in air pollution concentrations since 2010. To focus on the changes in ambient air pollutants due to local traffic emissions trends and to ensure that the regional scale changes did not confound the analysis, roadside increments above the urban background concentration were calculated. These concentrations were denoted by a Δ. Trends were examined at 65 London monitoring sites.

Between 2005 and 2009 there was an overall increase ΔNO_X and ΔNO_2. This is in sharp contrast to the decreasing trends predicted from the progressively tighter Euroclass emissions factors but concurs with a growing body of evidence that suggests that real-world emissions from diesel vehicles did not align with improved performance found in type approval tests. ΔPM_{10} showed a slight decrease overall during the same period.

Between 2010 and 2014 an improving picture was found with overall decreases in ΔNO_X, ΔNO_2 and $\Delta PM_{2.5}$. It appears that changes in $\Delta PM_{2.5}$ can be explained by changes in exhaust emissions of black carbon consistent with particle traps and other diesel emissions abatement. However, ΔPM_{10} concentrations showed no significant overall change suggesting an increase in coarse particles was offsetting decreases in tailpipe emissions; this was especially the case in outer London where some sites had increasing ΔPM_{10} trends. In most cases trends were similar at all times of the day, however, where differences in rates of change in ΔNO_X, ΔNO_2 and ΔPM_{10} were seen these showed lesser decreases or greater increases during the evening peak hours and weekends, when cars dominate traffic, when compared with morning peak periods that are dominated by heavier vehicles.

Between 2010 and 2014, sites that exhibited decreasing ΔNO_X and ΔNO_2 showed an approximate 1:1 ratio of ΔNO_X: ΔNO_2. However this ratio was not consistent at the monitoring sites that showed the greatest rate of improvement in ΔNO_2; Wandsworth - Putney High Street and Merton - Civic Centre showed $\Delta NO_X > \Delta NO_2$ but the reverse was true at Lambeth – Brixton Road and Camden – Euston Road showing that different technologies might be responsible for NO_X and NO_2 changes along London's roads or that traffic conditions may affect the performance of these technologies.

Westminster – Marylebone Rd is often cited as example for the roadside AQMS in inner London. Between 2010 and 2014 Marylebone Road experienced a slower decrease in NO_2 roadside concentrations (decrease of 1-6% annually from concentrations in 2010) compared to other sites in London, which raises the question why Marylebone Rd did not benefit from the NO_2 reduction observed in other locations. Despite a decrease in NO_2 levels in Marylebone Rd, NO_X concentrations increased by ~5% per year from 2010 to2014. This increase in NO_X concentrations was unusual but was observed elsewhere including City of London – Walbrook Wharf and Hackney – Old St and at Brent -Ikea. Conversely, Marylebone Road experienced one of the fastest reductions in ΔPM_{10} concentrations (10% annual decrease from levels in 2010). Only Camden – Swiss Cottage and Sutton – Wallington observed a faster PM_{10} annual decrease.

Changes in the ΔCO_2 did not match the downward predictions from improved fleet efficiency. ΔCO_2 showed an increase at Westminster - Marylebone Rd and non-significant changes at the two other roadside sites where change could be assessed.

Despite some sites exhibiting a downward trend in their roadside increment, the annual mean concentration of NO_2 exceeded the 2010 European Limit Value at around ¾ of road and kerbside AQMSs during 2014, with seven AQMSs measuring concentrations that were more than twice the limit.

This is the first time that London's roadside monitoring sites have been considered as a population rather than summarised as a mean behaviour only, allowing greater insight into the differential changes in air pollution abatement policies. The next stages of the project will characterise monitoring sites according to their vehicle activities and emissions with the aim of being able to indentify why air pollution continues to deteriorate alongside some roads and to replicate the decreases seen at the best performing locations.

1 Introduction

The air quality close to roads in large urban areas is usually affected by concentrations of nitrogen oxides (NO_X), particulate matter (PM) and black carbon (CBLK) among other pollutants from traffic. Some of these roads are located in central areas along high streets or popular streets used by pedestrians, therefore exposure to traffic-related pollutants can be very considerable. The main source of pollutants close to roads is exhaust emissions from traffic, especially diesel engines which emit nitrogen oxides (NO_X) in form of nitrogen monoxide (NO) and primary nitrogen dioxide (NO_2) and also elemental carbon and black carbon. However, other non-exhaust traffic-related emissions such as resuspension, tyre-wear and brake-wear could represent an important fraction of PM in roads. Diesel emissions are also known to be an important source of ultrafine particles (particles with <0.1 µm in diameter) which can be inhaled deeper in the lung system and therefore are thought to be more toxic than larger particles (HEI, 2013). Whilst the vast majority of roadside locations in London met the PM_{10} EU Annual Mean Limit Value of 40 µg m^{-3} in 2013 (Mittal et al., 2015; www.londonair.org.uk), the majority still exceeded the NO_2 EU Annual Mean Limit Value of 40 µg m^{-3} by a large margin. Additionally, meeting the EU $PM_{2.5}$ exposure reduction target remains challenging.

A large number of policy initiatives are being taken across London, the UK and the EU to improve air quality. Specific policies in London include the Mayor’s Air Quality Strategy (MAQS) in 2010 with the roll out of new hybrid buses or low-emission buses (Euro-IV) (GLA, 2010). Moreover, Transport for London (TfL) completed a bus retrofit program which fitted over 1000 buses with a Selective Catalytic Reduction (SCR) system to remove nitrogen oxide (NO_X) emissions from exhaust. The retrofit program was prioritized for those buses with routes along busy roads such as Elephant and Castle, Marylebone Road, Fulham Broadway, Oxford Street and Putney High Street (https://tfl.gov.uk/info-for/media/press-releases/2014/july/world-s-largest-bus-retrofit-programme-compjeted).

These initiatives were expected to have a direct impact on the air quality in the whole of London but especially alongside busy roads. Measurements from London’s roadside sites indicate that the pollution concentrations at some sites are improving much faster than others. For example, ambient concentrations of NO_2 and PM_{10} measured at Marylebone Road have reduced over the last four years but this tendency has not been replicated across other London roads. Conversely, NO_X concentrations at Marylebone Road have not decreased as expected.

The objective of this study is to identify those roadside locations in London that have experienced the greatest changes in air pollution concentrations since 2010. The air pollutants analyzed were: NO_X, NO_2, $PM_{10,}$ $PM_{2.5}$), CBLK, organic and elemental carbon (OC, EC), particle number (PN) and carbon dioxide (CO_2). Trends between 2010 and 2014 are compared with those between 2005 and 2009 to highlight those sites with a marked change in tendency.

2 Methods

2.1 Monitoring sites

In order to focus on the changes in ambient air pollution due to local traffic emissions (exhaust and non-exhaust), trends of the roadside increments above the urban background concentration were calculated. In this way changes over time due to processes at the regional scale did not confound the analysis. Roadside increments for a given pollutant were calculated for all the roadside and kerbside Air Quality Monitoring Sites (AQMSs) in London on an hourly basis by removing the background concentration from that measured at the roadside AQMS. Air Quality data was extracted from the UK Automatic Urban and Rural Network (AURN), DEFRA's Particle Composition and Number Network and Black Carbon Networks along with the London Air Quality Network (LAQN). In total these comprised 65 roadside AQMSs (Figure 1).

Measurements from Kensington and Chelsea - North Kensington (KC1, 51.521°N, -0.2135°E) were taken as background concentrations. The choice of KC1 as background was for three reasons:

- The use of a single background site allowed roadside increments to be directly compared between different roadside locations.
- It is the urban background AQMS with the longest complete time series for all pollutants.
- Trends observed at North Kensington were the same (within confidence interval) of the overall trends observed for all urban background sites in London (Supplementary Figure 1; Supplementary Figure 2), with the only exception of trends for NO_2 between 2005-2009 when a faster decrease in NO_2 concentrations was observed at this site compared with the other urban background sites in London. The rate for KC1 was -1.07 (-1.80, -0.81) µg m^{-3} y^{-1} and that for all London background sites was -0.37 (-0.67,-0.07) µg m^{-3} y^{-1}.

Roadside increments have been labelled with the prefix Δ.

2.2 Measurement methods

NOx (NO + NO_2) was measured by chemi-luminiscence and fortnightly calibrations enabled the traceability of measurements to national metrological standards. PM_{10} and $PM_{2.5}$ were measured by TEOM-FDMS (Tapered Element Oscillating Microbalance - Filter Dynamics Measurement System) and by TEOM. TEOM-FDMS measurements were reference equivalent. PM measurements made by TEOM were converted to reference equivalent using the Volatile Correction Model (VCM) (Green et al., 2009). CBLK in $PM_{2.5}$ was measured by the Magee Aethalometer AE22. Ultrafine particles were measured by Condensation Particle Counter (CPC) which counts particle number concentration (PNC; expressed as #N m^{-3}). Particles were first grown by condensation to a bigger size, thus allowing easy detection by laser scattering. The carbonaceous compounds in PM_{10} (organic and elemental carbon, abbreviated OC and EC, respectively) were measured by exposing a filter for a day followed by thermo-optical analysis. All instruments were subject to twice yearly audit tests by the National Physical Laboratory or Ricardo AEA.

Trends for roadside CO_2 concentrations have been also assessed. CO_2 is a greenhouse gas that it is found naturally in the atmosphere. However, CO_2 concentrations have been rising since industrial times due to the use of fossil fuels and landscape changes. Anthropogenic sources of CO_2 in urban areas are related to combustion processes such as burning of fossil fuels and electricity production. Roadside increments of CO_2 are taken as indication of direct exhaust emissions from traffic. CO_2 concentrations were measured using a LiCOR-820 Non-Dispersive IR analyzer. Two-point calibrations were carried out every 15 days with a zero-scrubber (soda lime) and a CO_2 span gas referenced to the International Scale (WMO-X2007).

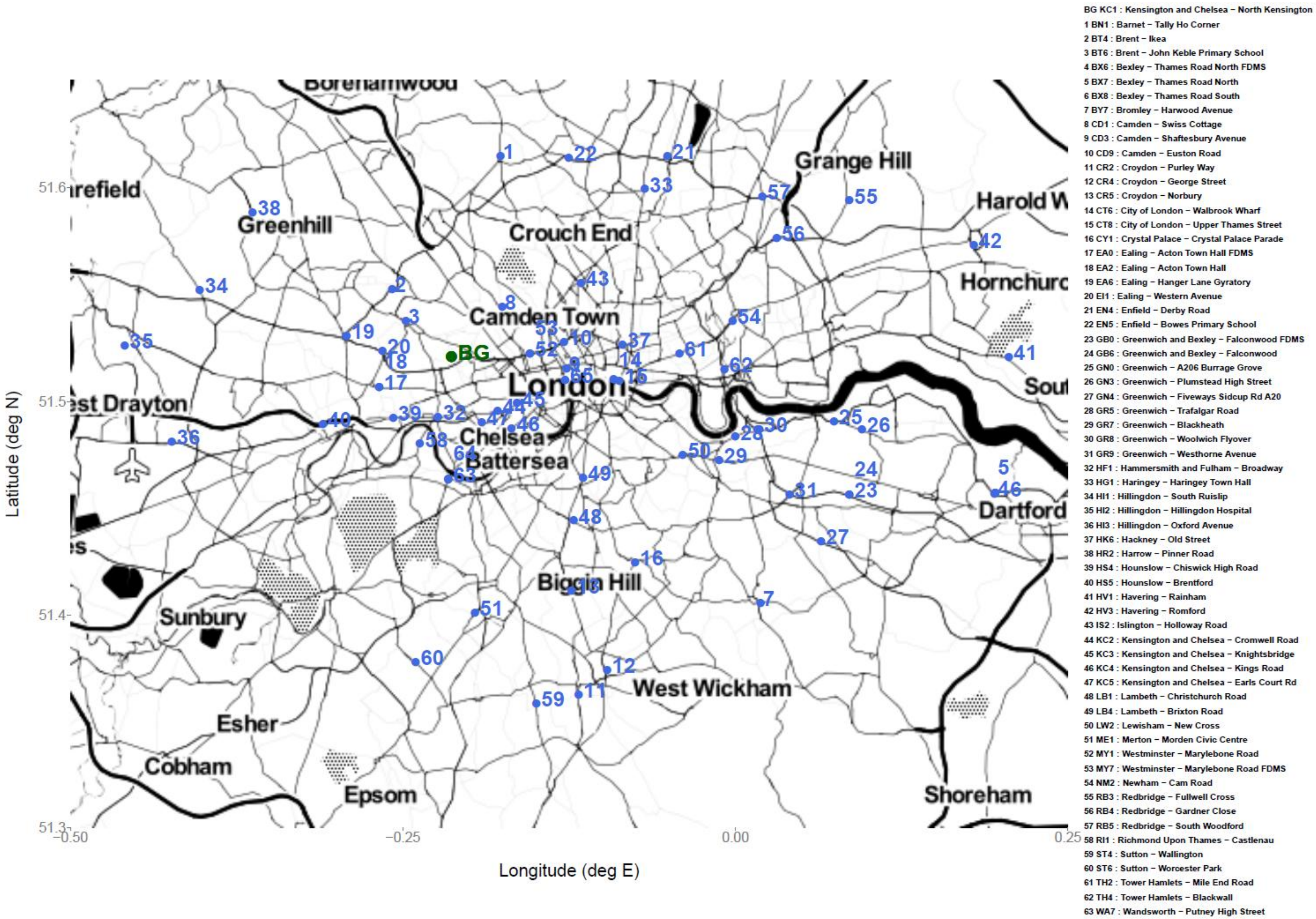

Figure 1. Map with the roadside sites and the background site (BG) used in this study.

2.3 Calculating trends

Trends in roadside increments were calculated for two periods: 1st January 2005 to 31st December 2009; and 1st January 2010 to 31st December 2014. Linear trends over the 5-year-periods were calculated using the ‘TheilSen’ function from the R/openair package (Carslaw and Ropkins, 2012; 2015). Briefly, the function works as follows. Given a set *n x,y* pairs, the slopes between all pairs of points are calculated and the median is given as the most probable slope (trend). This method is robust to outliers and can be used in non-normal and heteroscedastic data series (e.g. ambient air quality data). Confidence intervals were calculated at the 95% interval and estimates of the p-value were calculated by bootstrap sampling. A sensitivity test in the trend calculations was carried out and details are given in Supplementary Material (SM. 2).

Trends on roadside increments were calculated from monthly means which were first calculated from hourly roadside increments. Monthly means were calculated when data capture was greater than 75%. Missing monthly data was linearly interpolated. Time series were de-seasonalized by applying a LOESS smoothing function (Cleveland et al., 1990). Only sites with at least 45 months of available data for each of the 5-year-periods were reported.

The overall trend for all roadside and kerbside AQMSs in London for each time period was calculated by fitting the linear Random-Effects Model “DerSimonian-Laird estimator” (from the R/metafor package; Viechtbauer, 2010). The Random-Effects (RE) fit assumes that there are two sources of variation in the data set: the within-site estimation variance (variability in the trend calculated for one site as expressed by the confidence intervals) and between studies (variability of trends among sites) (Borenstein et al., 2010).

Trends for the morning and evening rush hour peaks and weekends were estimated using the same methodology. Morning and evening peaks were calculated from weekday data, excluding Public and Bank Holidays in the UK, between 6h and 10h, and between 16h and 20h local time, respectively.

Expressing the trend as percentage change presented some difficulties where the increment was small. At these locations small trends can become inflated when expressed as percentage changes which might not be realistic given intrinsic uncertainties of the measurements and in the trend assessment. This was especially the case at roadside sites in outer London. Instead of calculating a crude percentage, this metric was visualised by plotting trend against increment in the year of the period for all sites assessed.

Distance to London’s city centre for each AQMS was calculated, setting the centre at Charing Cross (51.508°N, 0.125°W). Sites <10 km from Charing Cross were flagged as inner London; sites > 10 km away from Charing Cross were flagged as outer London.

3 Results

3.1 Trends for the regulated pollutants: NO_2 (and NO_X) and PM

3.1.1 Trends between 2005 and 2009

The trends for the NO_X, NO_2 and PM_{10} roadside increments (namely ΔNO_X, ΔNO_2 and ΔPM_{10}, respectively) for the period 2005-2009 for both individual roads and for the whole of London are summarized in Figure 2. Trends in ΔNO_X were not statistically significant for a great range of sites in London; however the number of sites with a positive trend exceeded those with a negative one leading to an overall increase of 0.87 (0.07, 1.68) µg m^{-3} y^{-1}. For ΔNO_2, the majority of sites experienced an increase over time and the overall trend was 1.63 (1.25, 2.01) µg m^{-3} y^{-1}. The picture for the trends in ΔPM_{10} was more mixed with the majority of sites with a zero trend. Overall, the roadside AQMSs in London displayed a slight decreasing trend in ΔPM_{10}: -0.19 (-0.34, -0.03) µg m^{-3} y^{-1}.

Relative to their roadside increment in 2005 the majority of sites with a positive and statistically significant trend had ΔNO_X and ΔNO_2 rates greater than 5% (Figure 3 A, B and Figure 4A, B). ΔNO_X increased between 5 and 20%; ΔNO_2, between 20 and 50%. Sites with the greatest upward trends (>50%) in ΔNO_2 were located in outer London. That was due to very small roadside increments in the first year. Sites which experienced a decrease in their ΔNO_2 concentrations did it at a rate of ~1-5% and those were Islington – Holloway Road (IS2), Lambeth – Brixton Road (LB4) in inner London; and Redbridge – Fullwell Cross (RB3) in outer London.

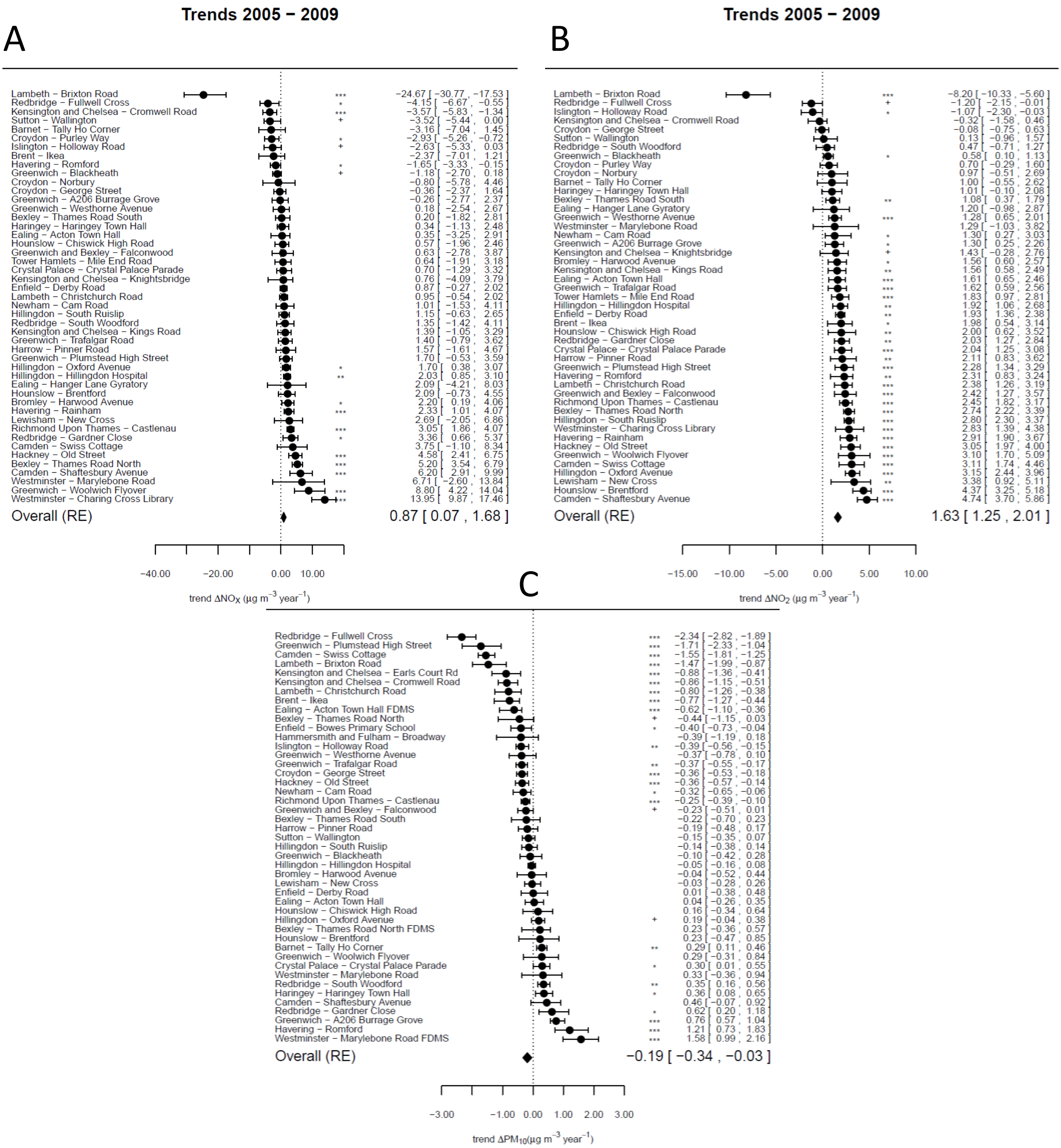

*Figure 2. Forest plots for the trends over time (expressed in µg m^{-3} y^{-1}) for the roadside increments of NO_X (A), NO_2 (B) and PM_{10} (C) for the 2005-2009 period. *** trend significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.*

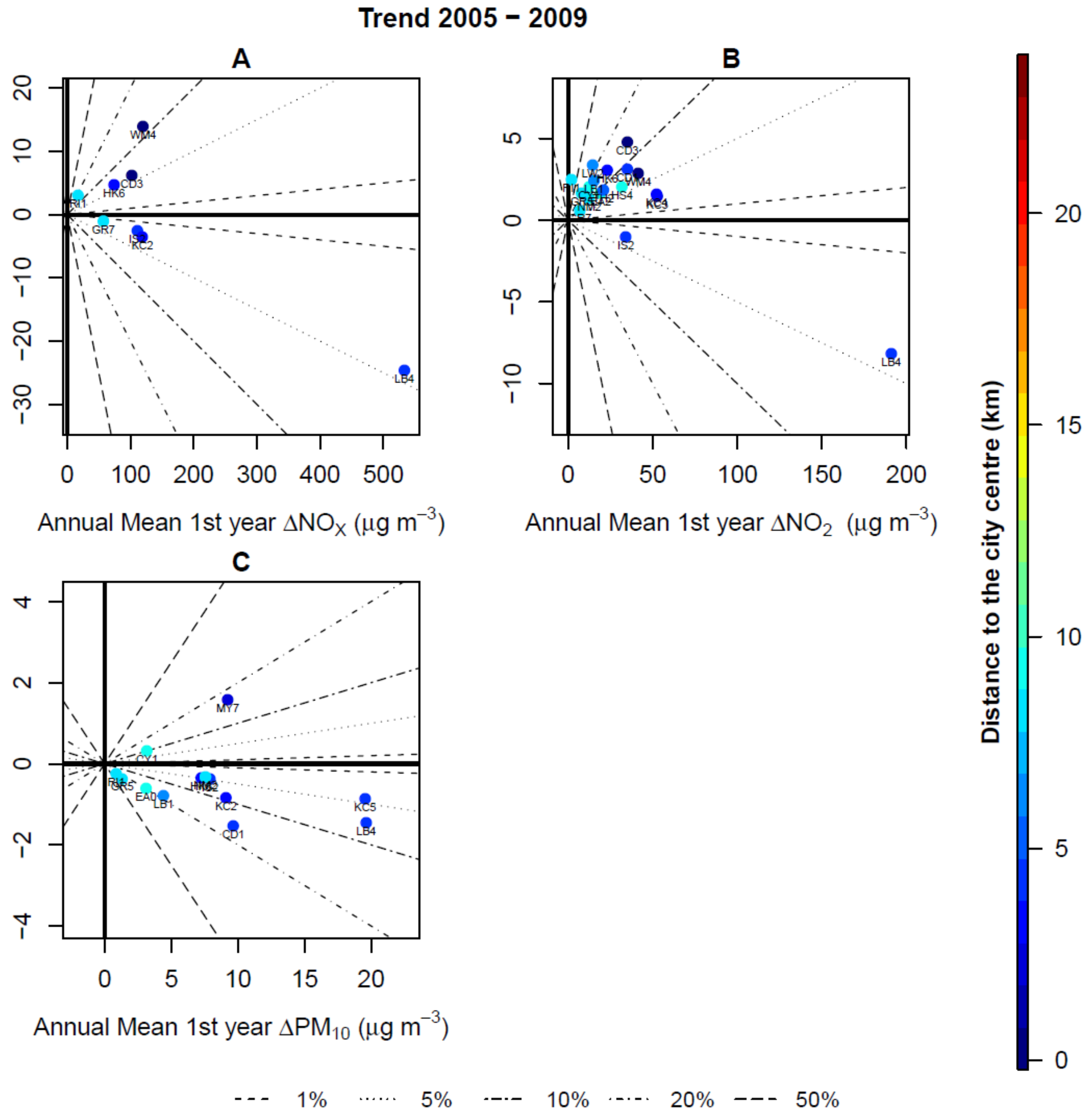

Figure 3. Median trend for the roadside increments between 2005 and 2009 against mean roadside concentration in 2005 for NO_X (A), NO_2 (B) and PM_{10} (C). Only sites in inner London (< 10 km from the city centre) with statistical significant trend (p<0.1) are shown.

Most sites in inner London with a statistically significant trend in ΔPM_{10}, decreased at a ~5% rate or faster (Figure 3C). Only Crystal Palace – Crystal Palace Parade (CY1) and Westminster - Marylebone Road FDMS (MY7) showed a positive trend (>10%). The picture was more mixed in sites in outer London (Figure 4C). Some sites increased at a much faster rate such as 50% at Haringey – Town Hall (HG1); Greenwich – A206 Burrage Grove (GN0); and Havering – Romford (HV3). Others exhibited a fast downward trend such as Greenwich – Plumstead High Street (GN3) and Redbridge – Fullwell Cross (RB3).

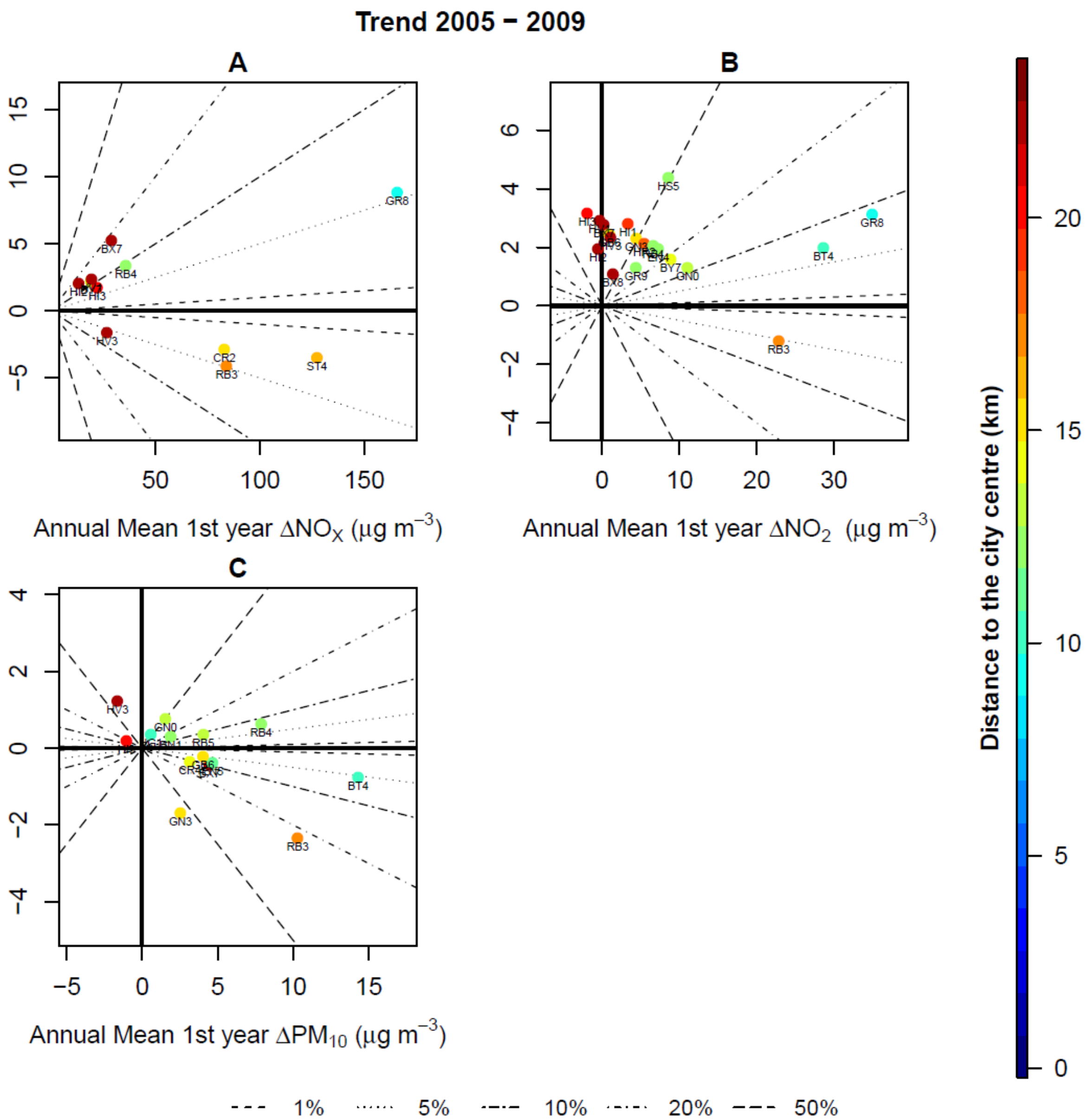

Figure 4. Median trend for the roadside increments between 2005 and 2009 against mean roadside concentration in 2005 for NO_X (A), NO_2 (B) and PM_{10} (C). Only sites in outer London (> 10 km from the city centre) with statistical significant trend (p<0.1) are shown.

3.1.2 Trends between 2010 and 2014

Overall trends in ΔNO_X and ΔNO_2 changed sign for the period between 2010 and 2014 when compared with 2005 to 2009. ΔNO_X and ΔNO_2 experienced a general downward trend for this period: -1.11(-2.27, -0.04) and -1.65 (-2.27,-1.03) µg m^{-3} y^{-1}, respectively (Figure 5A, B). The downward trend for ΔNO_2 was exhibited at more sites in London than for ΔNO_X (Figure 5A, B). The majority of sites in both in inner and outer London decreased their ΔNO_2 concentrations at a rate <10% of the annual mean increment in 2010 (Figure 6B and Figure 7C) with the excepption of Merton – Civic Centre (ME1) which decreased at 18% per year. In inner London, City of London – Walbrook Wharf (CT6), Ealing – Westbourne Avenue (EI1) and Hackney – Old Street (HK6) showed positive trends in ΔNO_2 at a significant rate (5% -10% per year); in outer London, Greenwich – Sidcup Road (GN4) and Haringey – Town Hall (HG1) showed positive trends in ΔNO_2 (20% increase per year).

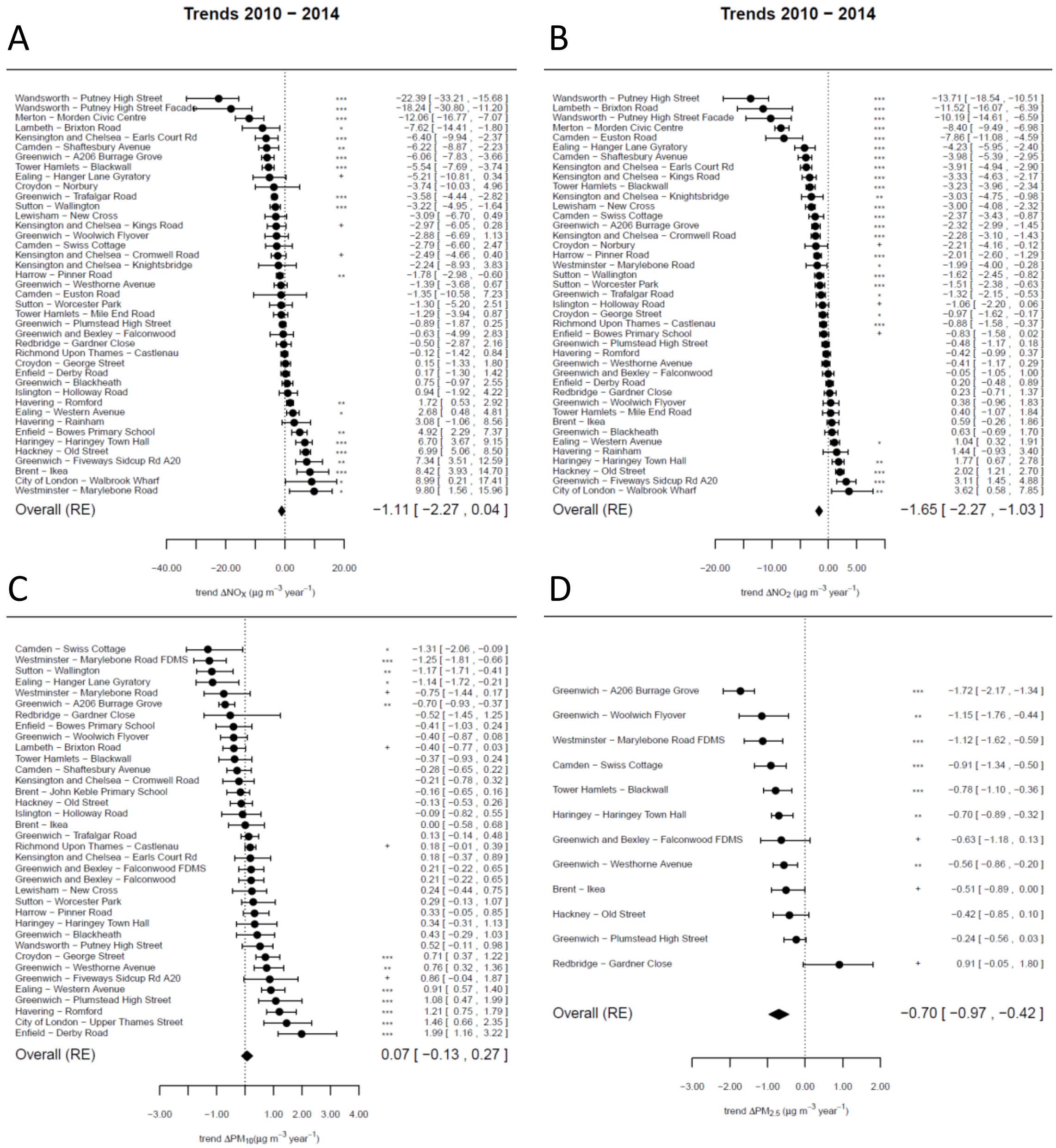

Figure 5. Forest plots for the trends over time (expressed in μg m^{-3} $year^{-1}$) for the roadside increments of NO_X (A), NO_2 (B), PM_{10} (C) and $PM_{2.5}$ (D) for the 2010-2014 period. *** trend significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.

The picture was again more mixed for PM_{10} and the overall trend for the ensemble of sites in London was fairly stable: 0.07 (-0.13, 0.27) μg m^{-3} y^{-1} (Figure 5C). In inner London, the sites closer to the centre (< 5 km) exhibited a significant downward trend (between 1% and 20% per year) with the only exception of City – Upper Thames Street (CT8) where ΔPM_{10} increased at 10% rate. Sites such as Ealing – Western Avenue (El1) and Richmond – Castlenau (RI1), located a bit further away from the centre (~8 km) exhibited a positive trend in ΔPM_{10} at a 10% per year rate. Most of the AQMSs in outer London experienced a significant upward trend (greater than 20% per year of the annual mean increment in 2010), with the exception of Ealing –Hanger Lane Gyratory (EA6), Greenwich – Burrage Grove (GN0) and Sutton – Wallington (ST4) (Figure 7C) where ΔPM_{10} decreased at a rate of 10-20% per year.

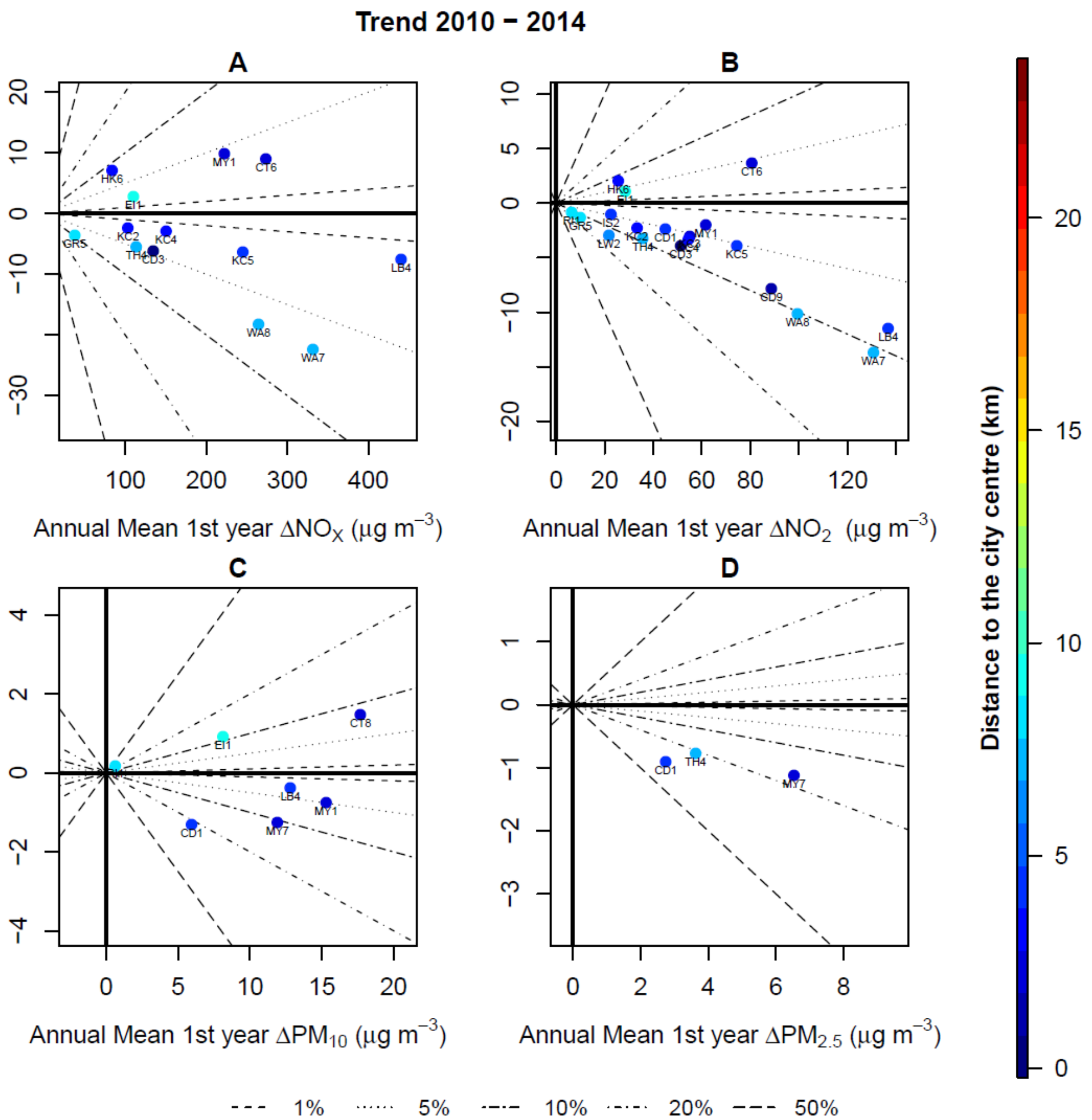

Figure 6. Median trend for the roadside increments between 2010 and 2014 against mean roadside concentration in 2010 for NO_X (A), NO_2 (B), PM_{10} (C) and $PM_{2.5}$ (D). Only sites in inner London (< 10 km from the city centre) with a statistically significant trend (p<0.1) are shown.

For this second period (2010-2014) trends for the roadside increments in $PM_{2.5}$ were also available for a number of sites. Overall, $\Delta PM_{2.5}$ decreased over time with a significant downward trend of -0.7 (-0.97, -0.42) µg m^{-3} y^{-1} (Figure 5D). The decrease in $\Delta PM_{2.5}$ was larger than 20% per year for the majority of sites in both inner and outer London (Figure 6D and Figure 7D) with a maximum of 50% per year decrease of the increment in 2010. Only one site (Redbridge – Gardner Close, RB4) exhibited increased $\Delta PM_{2.5}$ at a rate greater than 50% per year (Figure 7D).

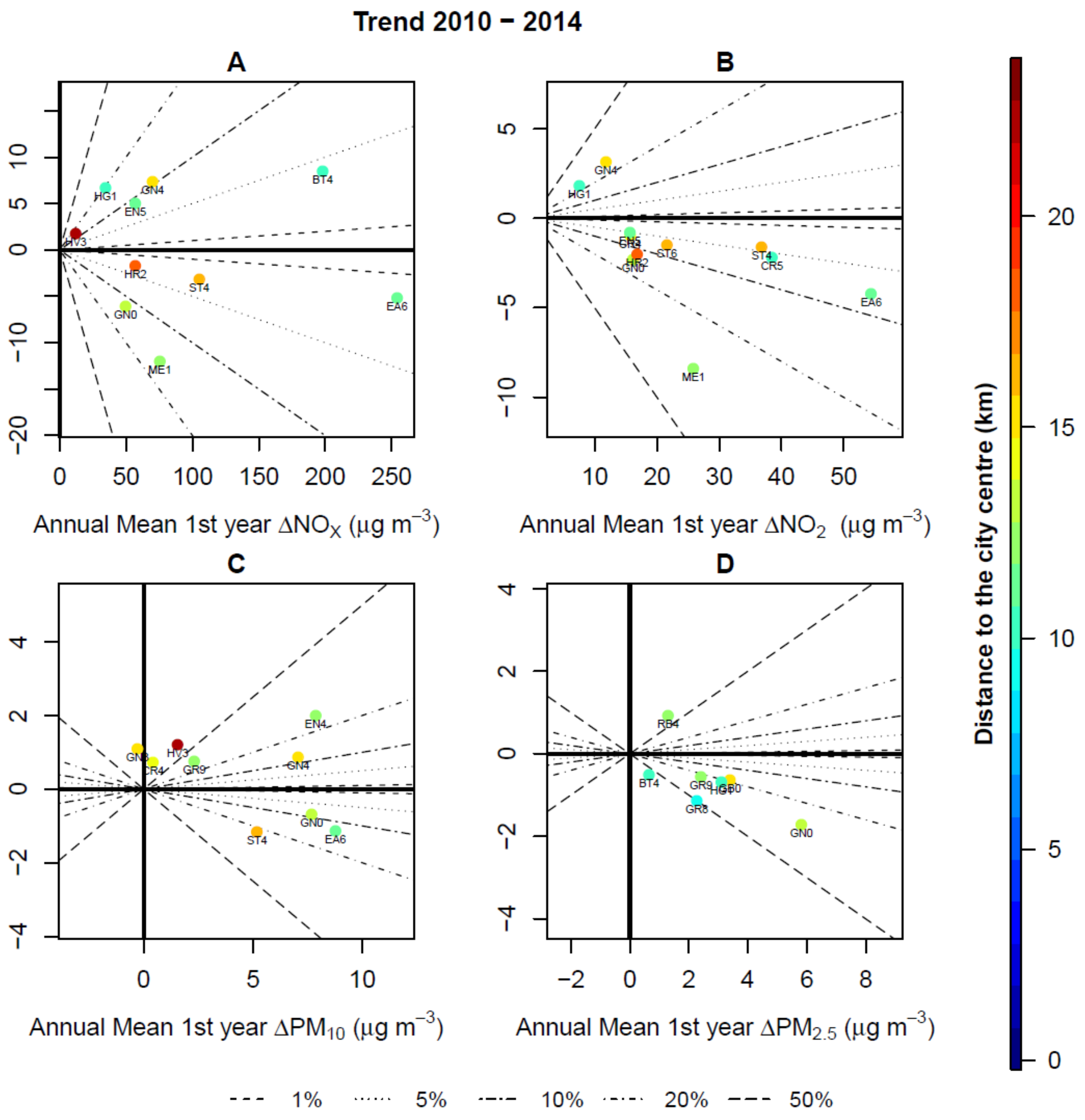

Figure 7. Median trend for the roadside increments between 2010 and 2014 against mean roadside concentration in 2010 for NO_X (A), NO_2 (B), PM_{10} (C) and $PM_{2.5}$ (D). Only sites in inner London (< 10 km from the city centre) with statistically significant trend (p<0.1).

To explain the extent to which trends were dominated by different composition of the traffic fleet, trends for roadside increments for weekday morning and evening rush hours; and for weekends were calculated (Supplementary Figure 3-Supplementary Figure 9). Note than only sites with a statistically significant overall trend are shown. Generally, monitoring sites exhibited similar trends for rush hour peaks and weekends for all the pollutants. However some interesting features are worth highlighting. The positive trend in ΔNO_X and ΔNO_2 at Hackney – Old St (HK6) (about 10% per year of the mean roadside concentration in 2010) was dominated by the trends during the weekday evening rush hour and weekends (~10% per year) while the trend during morning rush hour was <5% per year (Supplementary Figure 3, Supplementary Figure 5). The trend in ΔNO_X at Brent – Ikea (BT4) (~5% per year increase) was mostly due to the weekend concentrations that increased at a similar rate while trends were around ±1% per year during morning and evening peaks (Supplementary Figure 4). The positive trends in ΔPM_{10} observed in sites in outer London were attributed to an increase of their concentration in the evening rush hour and weekends. Notable were Richmond – Castelnau (RI1) (Supplementary Figure 7), Greenwich – Westbourne Av (GR9) and Enfield – Derby Rd (EN4) (Supplementary Figure 8) where trends during evening and weekends were ~50% per year while morning trends were <20% per year of the roadside PM_{10} concentrations in 2010. At these sites, the overall positive trend observed in 2010-2014 might have been led by passenger cars and not by HGVs that dominate the morning traffic peaks.

The comparison of trends for the different pollutants measured at the same AQMS offers an insight into the changes in the sources that might have taken place. The rate of reduction in ΔNO_2 and ΔNO_x were similar in most AQMSs between 2010 and 2014 (Figure 8A) with most the sites aligned on the 1:1 line.

However, some sites observed a much faster decrease in ΔNO_x concentrations than ΔNO_2: Wandsworth - Putney High Street (WA7, WA8) and Merton - Modern Civic Centre (ME1) and all sites located >10 km from the city centre. Conversely, at Lambeth – Brixton Road (LB4) and Camden – Euston Road (CD9) (sites in inner London) the downward trend in ΔNO_2 was faster than the downward trend for ΔNO_x. Other sites experienced a downward trend for ΔNO_2 concentrations whilst ΔNO_x increased (sites located in the right bottom quadrant in Figure 8A): Westminster – Marylebone Road (MY1), Enfield – Bowes Primary School (EN5), among others. Sites that experienced increases in ΔNO_x trends but decreases in ΔNO_2 were: City of London – Walbrook Wharf (CT6), Greenwich – Fiveways Sidcup Rd (GN4), Hackney – Old Street (HK6), Haringey – Town Hall (HG1).

The comparison between the trends in $\Delta PM_{2.5}$ and trends in ΔPM_{10} indicated that the majority of sites in inner London (distance from the city centre < 10 km) experienced a downward trend in both PM fractions at similar rates. Therefore the downward trend for ΔPM_{10} roadside concentrations could be attributed to the decrease in the fine fraction. With the exception of Greenwich – A206 Burrage Grove (GN0), sites in outer London experienced an increase in ΔPM_{10} while $\Delta PM_{2.5}$ decreased (sites in the right bottom quadrant in Figure 8B); implying an increase in coarse PM fraction whilst the levels in fine fraction went down. Only Redbridge – Gardner Close (RB4) experienced an increase in $\Delta PM_{2.5}$ while ΔPM_{10} levels decreased.

Direct comparison of absolute trends of ΔPM and trends of ΔNO_2 should be done cautiously since a change in the intensity of sources would respond differently in the absolute rate for the two pollutants. Some AQMSs observed a decreasing trend for both ΔNO_2 and ΔPM (Figure 8C, D, bottom left quadrant). Notably, some sites experienced a decrease in both PM fractions: Greenwich – Burrage Grove (GN0), Tower Hamlets - Blackwall (TH4) and Camden – Swiss Cottage (CD1). These might be the result of a decrease in the traffic flow, possibly accompanied by a decrease in traffic exhaust emissions. Other sites exhibited a decrease in both ΔNO_2 and $\Delta PM_{2.5}$ concentrations while ΔPM_{10} increased: Greenwich – Westbourne Avenue (GR9) and Plumstead High Street (GN3). That might be due to a decrease in the exhaust emissions while non-exhaust traffic emissions (such as resuspension or tyre-wear) increased with an increase in the traffic flows. Notably, there are a group of sites where ΔNO_2 and ΔPM_{10} roadside concentrations increased over time but these sites were located mostly in outer London: Haringey - Town Hall (HG1), Ealing – Western Avenue (EI1), Greenwich – Blackheath (GR7) and Sidcup Road (GN4).

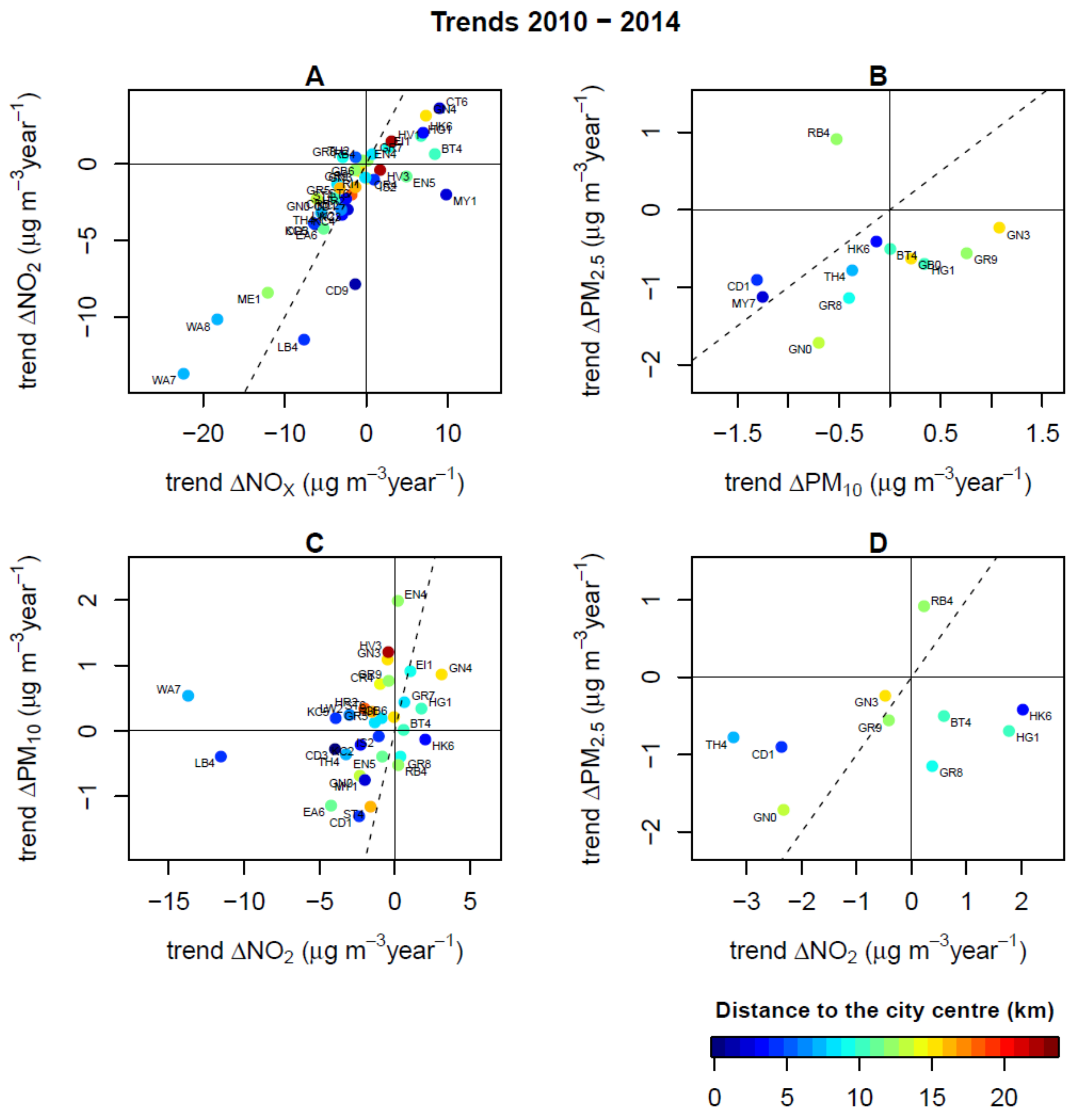

Figure 8. Comparison of trends in ΔNO_2 vs trends in ΔNO_X (A), trends in $\Delta PM_{2.5}$ vs trends in ΔPM_{10} (B), trends in ΔPM_{10} vs trends in ΔNO_2 (C) and trends in $\Delta PM_{2.5}$ vs trends in ΔNO_X. Dashed line indicates the 1:1 line.

It should be mentioned that some of London's roadside AQMSs dominated by buses in their traffic fleet observed a much faster change in ΔNO_2 compared to ΔPM_{10} (Figure 8C). Most notable were Wandsworth – Putney High Street and Lambeth – Brixton Rd (LB4) (sites) where ΔNO_2 concentrations decreased at a significant rate (10% per year) while PM_{10} remained fairly constant (with a median trend of 0.5 μg m^{-3} $year^{-1}$ and -0.5 μg m^{-3} $year^{-1}$, respectively, representing about 2% annual change).

3.2 Comparison of trends 2005-09 with trends 2010-14

Most of the roadside and kerbside AQMSs in London observed an improvement in concentrations of ΔNO_X and ΔNO_2 during the second period while between 2005 and 2009 ΔNO_X and ΔNO_2 increased (see sites in bottom right corner in Figure 9A, B and Figure 10A, B). However, some sites exhibited an upward trend in both periods: Hackney – Old Street (HK6), Haringey – Town Hall (HG1), Havering – Rainham (HV1) and Westminster – Marylebone Road (MY1) (the latter only for ΔNO_X). Some sites observed deterioration in ΔNO_X in the second period having benefited from a decrease in the first one: Islington -Holloway Road (IS2), Brent – Ikea (BT4), Havering – Romford Road (HV3) and Greenwich – Blackheath (GR7). No AQMS in London observed this pattern in terms of ΔNO_2. Sites with an increasing trend in ΔNO_2 for both periods of time showed a much slower trend in the second period. It is also worth mentioning the trend exhibited at Lambeth – Brixton Road (LB4) that showed a downward trend during both periods for both ΔNO_X and ΔNO_2 which was faster during 2010-2014 compared with the period before.

The majority of sites close to the city centre (<10 km) experienced a downward trend in ΔPM_{10} during 2010-2014 (Figure 9C) along with Sutton – Wallington (ST4), Greenwich – Woolwich Flyover (GR8) and Burrage Grove (GN0) and Redbrige – Fullwell Close (RB4) which were located further from the centre (Figure 10C). The AQMSs that observed an upward trend in the period 2010-2014 were mostly those further away from the centre, and some of them showed a downward trend in the first period: Greenwich – Plumstead High Street (GN3), Greenwich – Westhorne Avenue (GR9), Greenwich and Bexley – Falconwood (GB6), Harrow – Pinner Road (HR2) and Croydon – George Street (CR4).

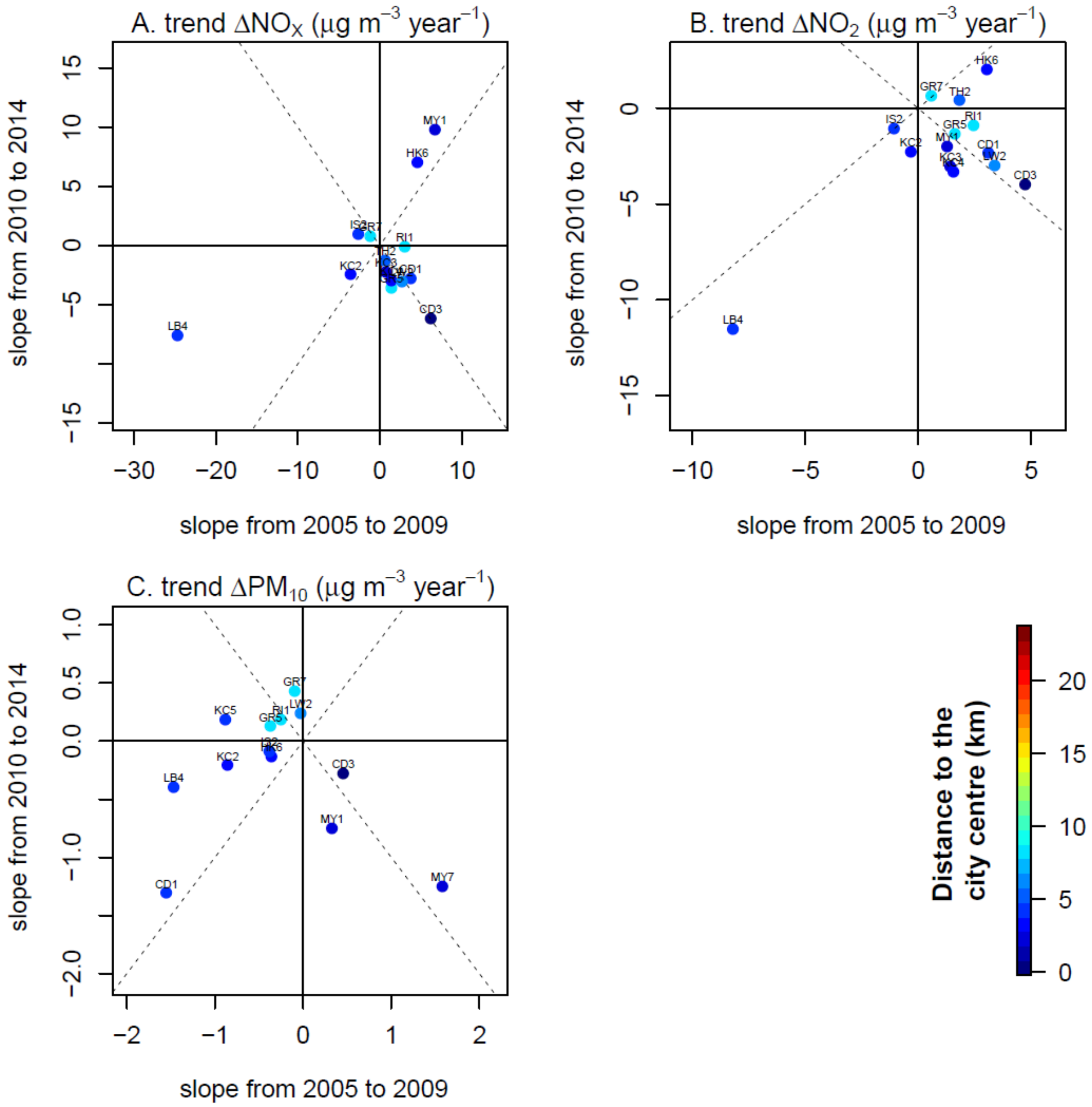

Figure 9. Comparison of the trends for ΔNO_X (A), ΔNO_2 (B) and ΔPM_{10} (C) calculated for the two time periods for sites in Inner London. Dashed line indicates the 1:1 line.

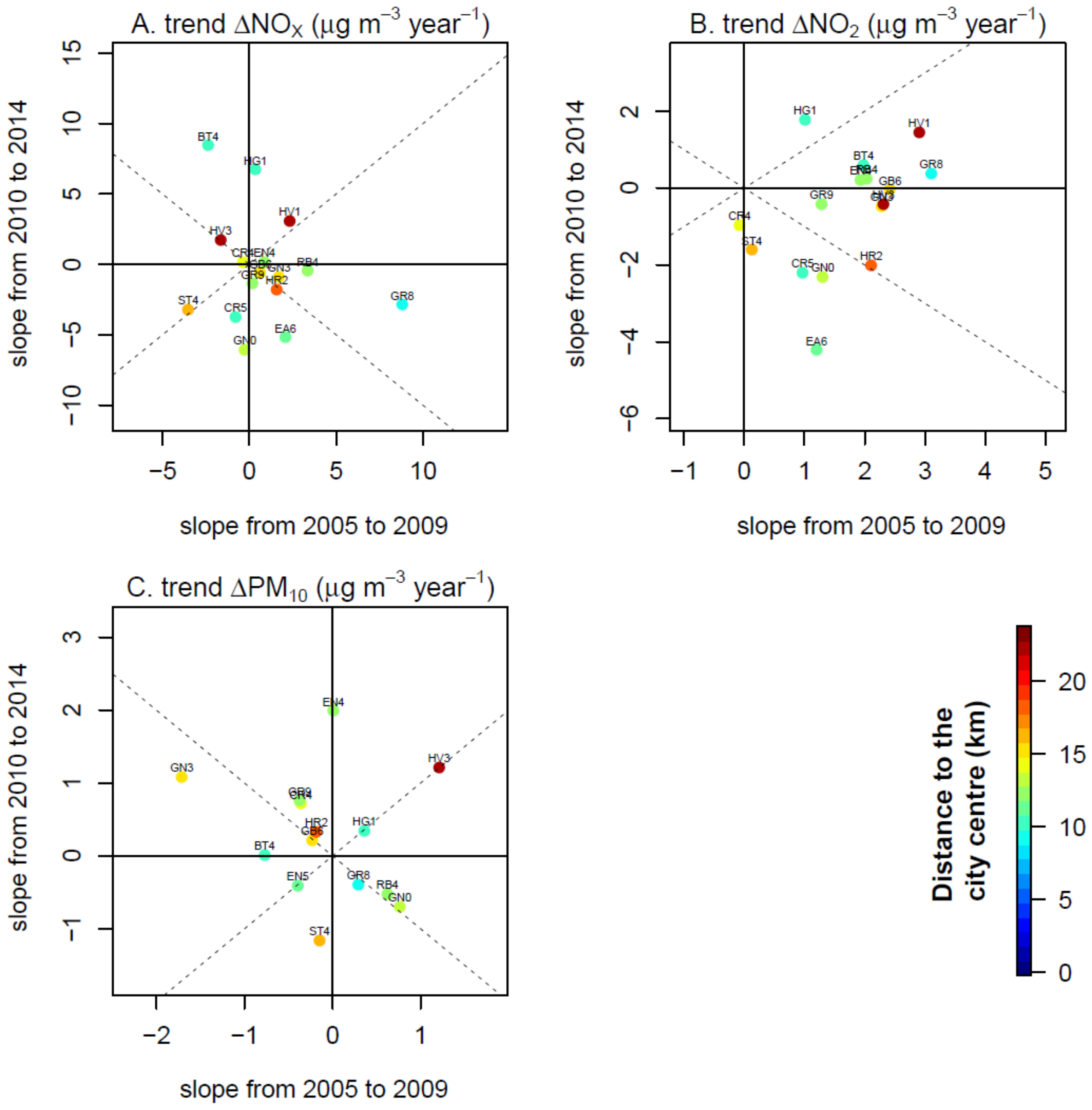

Figure 10. Comparison of the trends for ΔNO_X (A), ΔNO_2 (B) and ΔPM_{10} (C) calculated for the two time periods for sites in Outer London. Dashed line indicates the 1:1 line.

3.3 Trends for other air pollutants

Three roadside AQMSs in the London network measured carbon dioxide (CO_2) and black carbon (CBLK) for the period 2010-2014: Westminster – Marylebone Road (MY1 and MY7 for TEOM and TEOM-FDMS measurements, respectively); Brent – Ikea (BT4) and Tower Hamlets – Blackwall (TH4). Marylebone Rd measured particle number (PN) along with organic and elemental carbon (OC, EC, respectively). The trends for these pollutants are summarized in Table 1.

ΔCO_2 concentrations showed a significant upward trend at Marylebone Road. Tower Hamlets also experienced an upward trend although not statistically significant. Trends in ΔCO_2 did not match those of ΔNO_X, ΔNO_2 or ΔCBLK (Figure 11 C, D, I). ΔCO_2 showed the opposite trend to ΔPM_{10} and $\Delta PM_{2.5}$ (while ΔPM concentrations decreased over time, ΔCO_2 increased; Figure 11A, B). However, a significant upward trend was also observed for ΔOC measurements at Marylebone Road for the same period of time (Table 1).

Table 1. Results for the trends calculated for the period between 2010 and 2014 for ΔCO_2, Δblack carbon (ΔCBLK), Δparticle number (ΔPN) and Δorganic and Δelemental carbon (ΔOC, ΔEC).

	Westminster Marylebone Rd	Brent Ikea	Tower Hamlets Blackwall
ΔCO_2 (ppm y^{-1})	1.08 (0.14, 2.29)*	-0.48 (-1.42, 0.64)	0.44 (-0.20, 1.08)
ΔCBLK (µg m^{-3} y^{-1})	-1.04 (-1.32, -0.74)***	-0.53 (-0.70, -0.34)***	-0.28 (-0.39, -0.18)***
ΔPN (N m^{-3} y^{-1})	-2.25 (-30.49, -11.41)·10^{3}***	---	---
ΔOC (µg m^{-3} y^{-1})[a]	0.22 (0.03, 0.45)*	---	---
ΔEC (µg m^{-3} y^{-1})[a]	-0.65 (-0.95, -0.37)***	---	---

**** trend significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. [a] trend calculated for the period between 2010 and 2013.*

Black carbon measurements showed a significant decrease in their roadside increment at the three AQMSs where it was measured with an overall decrease estimated at -0.59 (-0.96, -0.23) µg m^{-3} y^{-1}. The decrease was faster at Marylebone Rd, followed by Brent and Tower Hamlets. The decrease in ΔCBLK was consistent with the decrease in ΔPM_{10} and $\Delta PM_{2.5}$ (Figure 11E, F) with trends aligned in the 1:1 line. Roadside increments in PN and EC also showed a significant downward trend for 2010-2014 although this was only measured at Marylebone Road (Table 1).

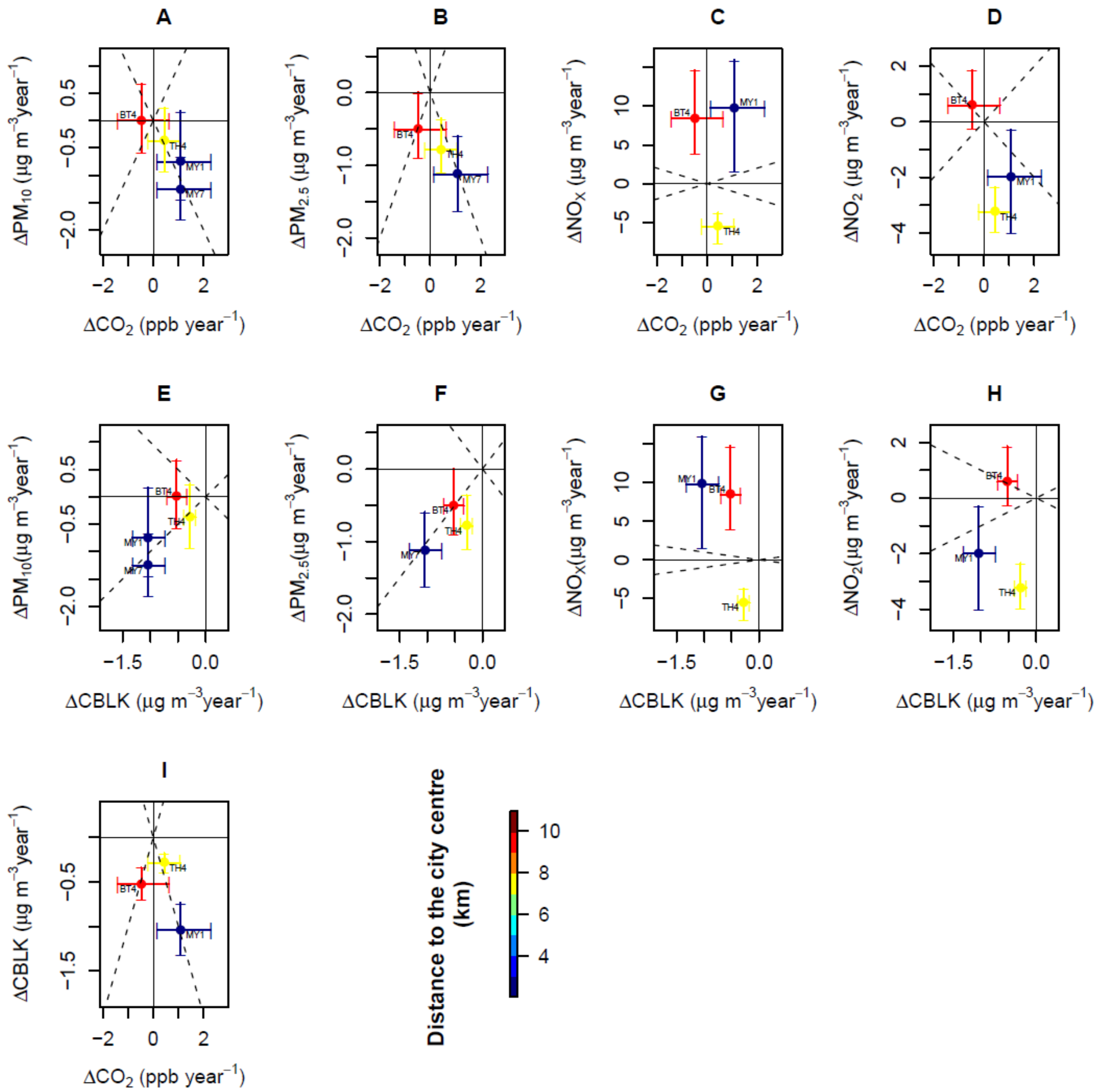

Figure 11.Trends of roadside CO_2 increments (A-D) and trends of roadside black carbon increments (E-H) against trends of regulated pollutants for the period between 2010 and 2014; and comparison of trends of black carbon against trends of CO_2 (I) at Marylebone Road, Brent – Ikea and Tower Hamlets – Blackwall.

4 Discussion and conclusions

Policies that aimed to reduce ambient air pollution levels by regulating traffic emissions in London had a clear impact from 2010 onwards. The majority of roadside and kerbside sites in London had a significant downward trend in their NO_2 and $PM_{2.5}$ roadside increment. NO_2 levels decreased annually by between 5 and 20% of the mean roadside NO_2 increment in 2010; $PM_{2.5}$ decreased annually by 10-50% of the mean roadside increment in 2010. Those policies had a clear impact compared to the trend observed for the 5-year-period before (2005-2009) when a wider upward tendency was observed in ΔNO_2 concentrations. No data is available to compare trends between the two periods of time for $\Delta PM_{2.5}$. ΔNO_X also registered a general downward trend (1-5% per year) between 2010 and 2014.

Despite the general behaviour of ambient air pollutants at roadside sites in London there was clear intra-city variability in trends. This might be explained by two causes. First, different policies might have been applied locally. Second, due to different composition of the local vehicle fleet, air pollution trends might respond differently to fleet technology changes (e.g. introduction of Euro-classes, alternative-fuelled vehicles, etc.), introduction of emission abatement technologies to diesel heavy-vehicles or to behavioural changes with people and businesses changing their use of specific vehicle types (e.g. increase of the fleet age during the economic downturn).

In the UK, the share of diesel vehicles achieved a record volume in registrations in 2013 (SSMT, 2013). Previous studies have indicated that diesel emissions make important contributions to NO_X and primary NO_2 emissions in urban areas (Sundvor et al., 2012 and references within). In London some central routes are dominated by buses powered by diesel, contributing 33% of the total road transport NO_2 emissions in central London as estimated by the LAEI (GLA, 2013). During the second analysis period, Transport for London (TfL) and the Department for Transport invested in a retrofit programme for Euro-III buses with a low-NO_2 Selective Catalytic Reduction Trap (SCRT) system that combines a CRT (Continuously Regenerating Trap) to reduce particle emissions; and a SCR (Selective Catalyst Reduction) to reduce NO_X emissions. The programme was completed in March 2014 (Carslaw et al., 2015). SCRT systems have been shown to reduce primary emissions of NO_2 by 61% and NO_X by 45% under real-driving conditions in London compared to buses only fitted with a CRT (Carslaw et al., 2015). Retrofitting buses along Putney High Street led to a decrease in local NO_X and NO_2, with the concentration of ΔNO_X decreasing to a greater extent than that of ΔNO_2 (Barratt and Carslaw, 2014) consistent with the patterns seen in the trend analysis here. Merton- Civic Centre also experienced a greater decrease in ΔNO_X when compared to ΔNO_2 however this is in contrast to Camden –Euston Road and Lambeth- Brixton High Road that experienced a faster change in ΔNO_2 than ΔNO_X suggesting that different technologies might be responsible for NO_X and NO_2 changes along London's roads.

Despite the observed general decreasing trend in ΔNO_X, some sites experienced a worsening in their ΔNO_X concentrations (e.g. sites in central London such as Westminster- Marylebone Road; Hackney –Old Street; City of London – Walbrook Wharf, with increases of up to 10% per year; at other sites in outer London such as Brent – Ikea, Enfield – Bowes Primary School and Haringey – Town Hall their ΔNO_X levels increased by up to 20% annually compared to 2010 concentrations).

Although $\Delta PM_{2.5}$ decreased in the period between 2010 and 2014, ΔPM_{10} roadside levels remained constant or decreasing at a similar annual rate as $\Delta PM_{2.5}$ indicating a general increase/stabilization in the coarse fraction. PM coarse is associated with non-exhaust traffic emissions such as resuspension from the road, brake and tyre-wear. This increase in the coarse fraction was seen mainly alongside roads in outer London. The collocation of other measurements such as black carbon at some sites in the network confirms that the decrease in $\Delta PM_{2.5}$ was largely explained by the decrease in traffic exhaust emissions.

Some roadside sites in London experienced a reduction in their ΔNO_2 and $\Delta PM_{2.5}$ concentrations while ΔPM_{10} remained constant. That would be the signature of sites with diesel NO_2 emissions control devices where exhaust emissions have been reduced. Assuming that traffic levels remained constant over time, non-exhaust traffic emissions such as resuspension, tyre-wear and brake-wear have not been tackled therefore not reduced.

There was, however, a general decrease in ΔPM_{10} in AQMSs located in inner London (~10% decrease per year on increments in 2010); however, a tendency to increase (10% per year) was observed in sites in outer London. The increment in ΔPM_{10} in AQMSs in outer London was mainly driven by an increase in roadside increments during the weekday evening rush hours and the weekends when traffic is generally dominated by passenger cars, though road speeds could also be a factor.

A number of sites, mainly in outer London, had increased ΔNO_2 and ΔPM_{10} during the second period of time indicating that traffic emissions (both exhaust and non-exhaust) increased.

The trend in ΔCO_2 shows an unexpected behaviour. Whilst traffic-exhaust related pollutants (Δblack carbon and $\Delta PM_{2.5}$) decreased between 2010 and 2014, ΔCO_2 increased. The trend in ΔCO_2 did not have any relation to trends in both ΔNO_X and ΔNO_2. Since ΔCO_2 roadside concentrations are related to fuel use, ΔCO_2 was expected to decrease with improving fleet efficiency and due to the increase of alternatively-fuelled vehicles in the traffic fleet in recent years, although some of these changes would have been offset by growth in light commercial vehicles and higher payload vans (SMMT, 2013). Overall the London Atmospheric Emissions Inventory (LAEI) predicted ~ 1% annual decrease in road transport CO_2 emissions between 2010 and 2015 which was not borne out by roadside measurements which showed an increase at Westminster - Marylebone Road and non-significant changes at the two other roadside sites where change could be assessed.

In the analysis we can clearly identify so-called outlier sites that experienced different trends compared to the overall tendency. Westminster – Marylebone Rd is often cited as example for the roadside AQMS in inner London. Between 2010 and 2014 Marylebone Rd experienced a slower decrease in NO_2 roadside concentrations (decrease of 1-6% annually from concentrations in 2010) compared to other sites in London: Wandsworth – Putney High St and Camden – Euston Rd in inner London. That raises the question why Marylebone Rd did not benefit from the NO_2 reduction observed in other locations. Despite a decrease in NO_2 levels in Marylebone Rd, NO_X concentrations increased by ~5% per year in the second period of time (2010-2014). This increase in NO_X concentrations was unusual and only observed but not unique. Conversely, Marylebone Road experienced one of the fastest reductions in PM_{10} concentrations (10% decrease from roadside PM_{10} levels in 2010) in the second period. Only Camden – Swiss Cottage and Sutton – Wallington observed a faster PM_{10} annual decrease (~20% per year).

It should be highlighted that despite some sites exhibiting a downward trend in their roadside increment, the annual mean concentration of NO_2 exceeded the 2010 European annual mean Limit Value at around ¾ of road and kerbside AQMSs during 2014, with seven AQMSs measuring concentrations that were more than twice the limit.

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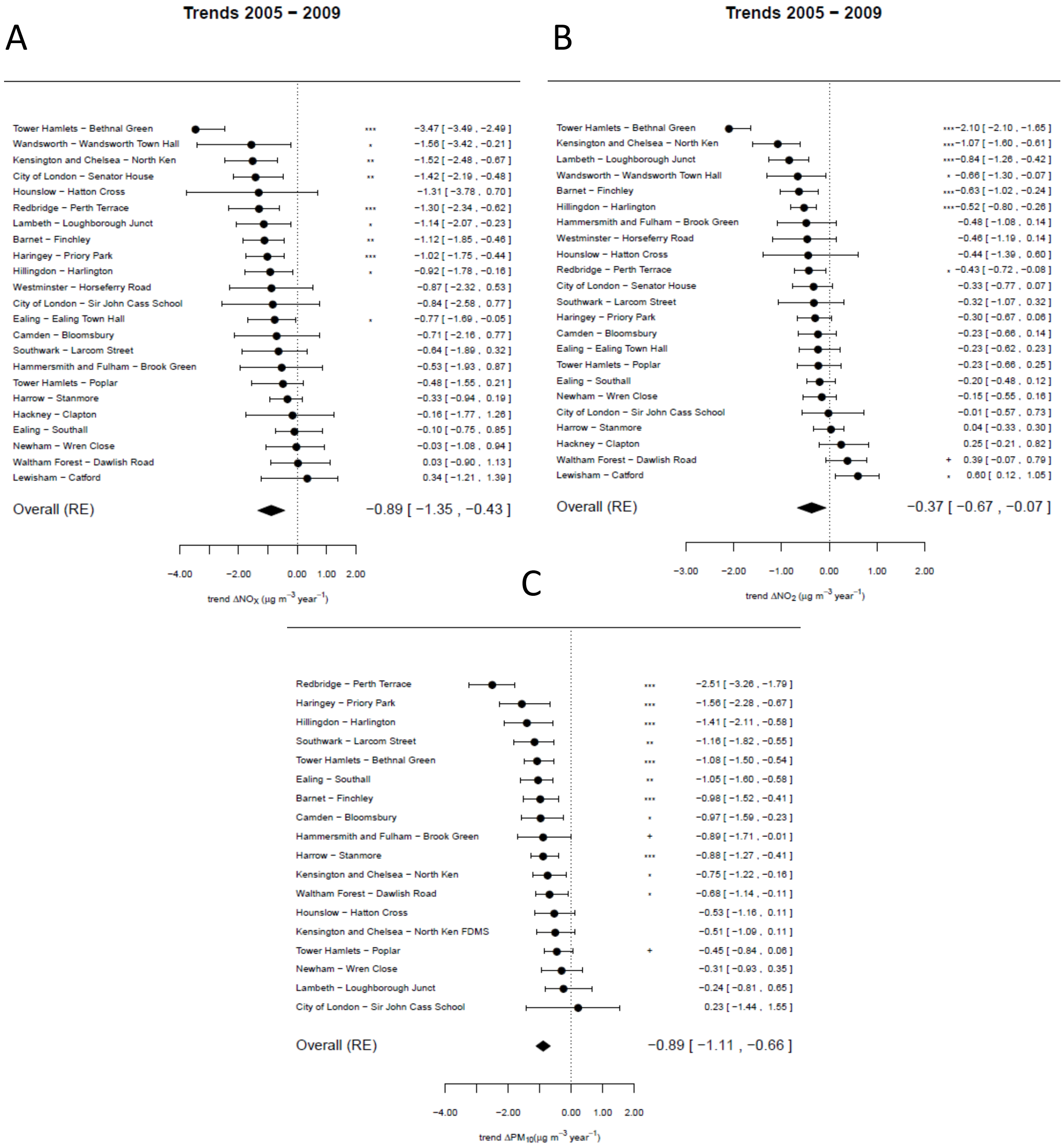
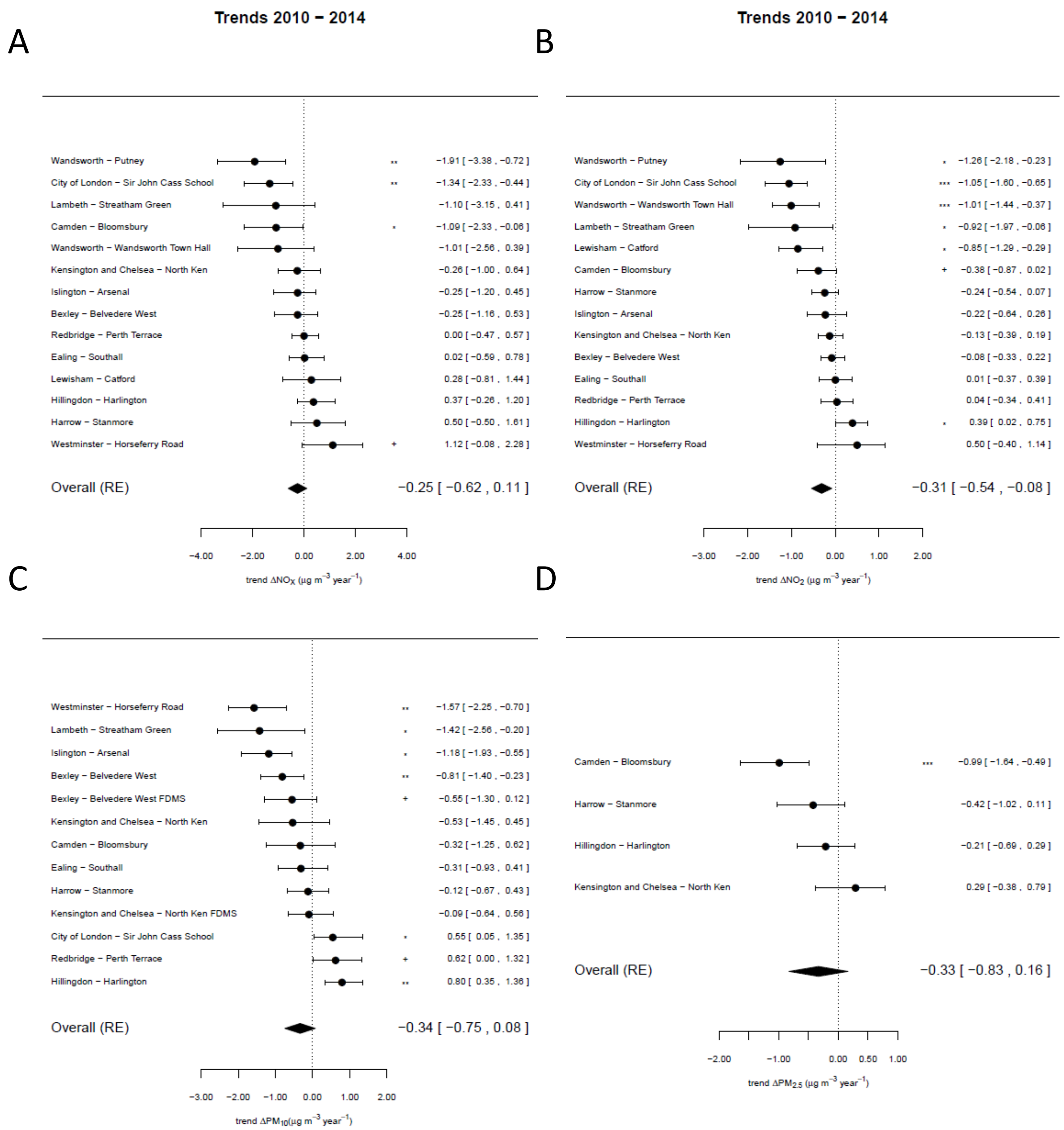
Supplementary Material

SM1. Trends of air pollutants at the London urban background sites



Supplementary Figure 1. Forest plots for the trends over time (expressed in µg m-3 year-1) for the urban background concentrations of NO_X (A), NO_2 (B) and PM_{10} (C) for the 2005-2009 period. *** trend significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.



Supplementary Figure 2. Forest plots for the trends over time (expressed in µg m^{-3} $year^{-1}$) for the urban background concentrations of NO_X (A), NO_2 (B), PM_{10} (C) and $PM_{2.5}$ (D) for the 2010-2014 period. *** trend significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.

SM2. Sensitivity test in trends calculations

Sensitivity tests were carried out to evaluate how robust the method and report findings were to different assumptions in the trend calculations. The overall trend, as estimated by the Random-Effect (RE) model, was calculated for all the roadside pollutants for the two periods of time using different assumptions.

- First, the effect of the monthly data capture threshold was assessed. The 75% data capture threshold was compared with a 95% threshold.

- Second, the influence of possible autocorrelation in the time series in the uncertainty estimates was assessed. Autocorrelation can exist in time series where one measurement is dependent on the previous one.

Results from the sensitivity test are summarized in Supplementary Table 1.

The different assumptions used in the trend calculations did not affect either the sign or the significance of the overall trend for any of the pollutants and time periods. Although changes were found in the overall estimates these changes were within the uncertainty estimates from the results of the main analysis.

The 75% data capture was therefore favoured for the main analysis reducing the degree to which interpolation was used. This data capture threshold was consistent with that used for EU reporting when calculating hourly and daily mean concentrations.

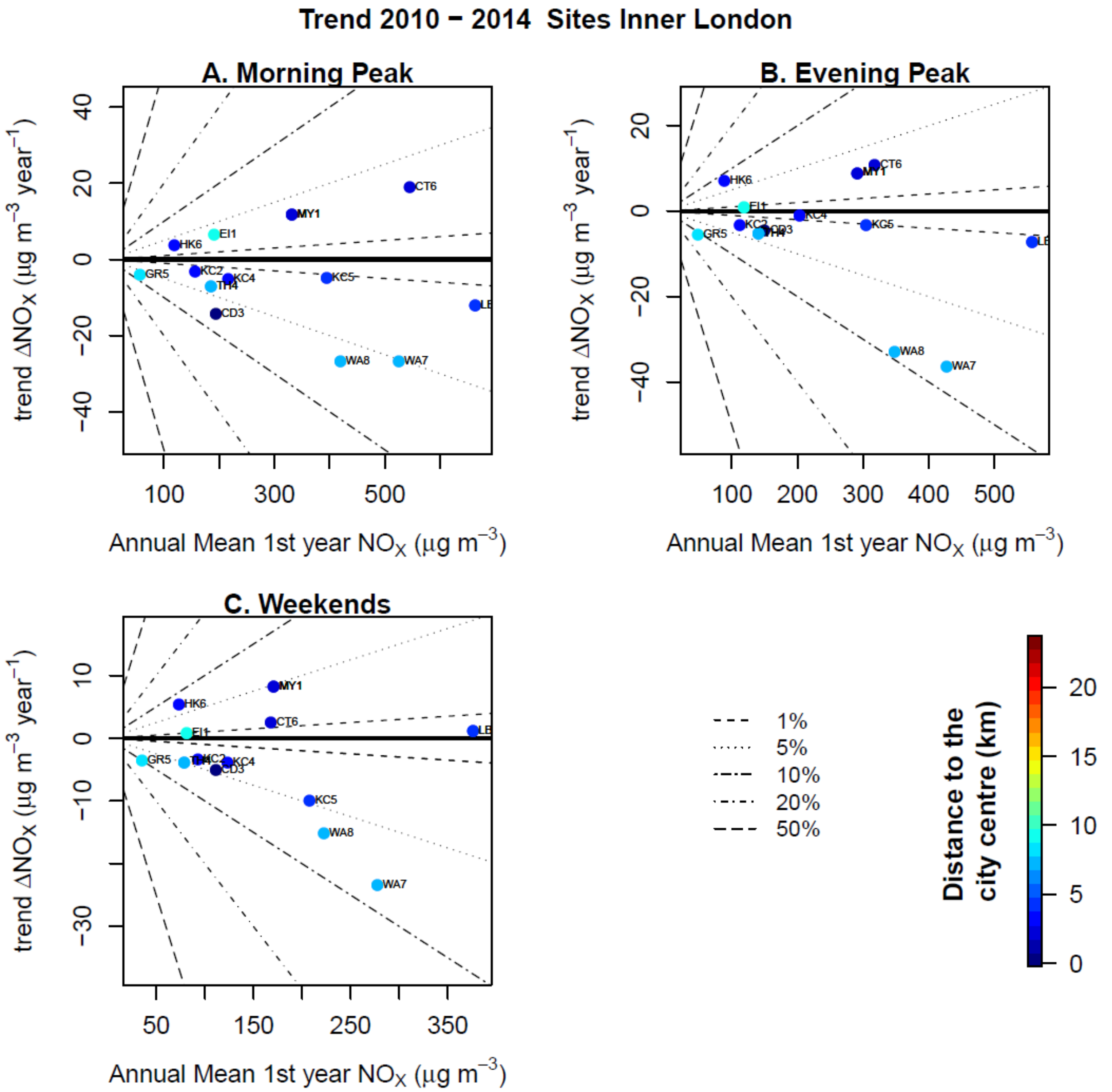
Allowing for auto-correlation led to contradictory results. As expected the inclusion of autocorrelation led to increased uncertainty in trends at the majority of sites, when compared with the main analysis. However, despite the increased uncertainty at individual sites the RE calculations of overall change had less uncertainty having severely down weighted those monitoring sites with largest trends and largest uncertainly.

Supplementary Table 1.Overall estimate for trends for roadside pollutants as calculated by the Random-Effects model using different parameters (data capture, autocorrelation) in the calculation of the trends.

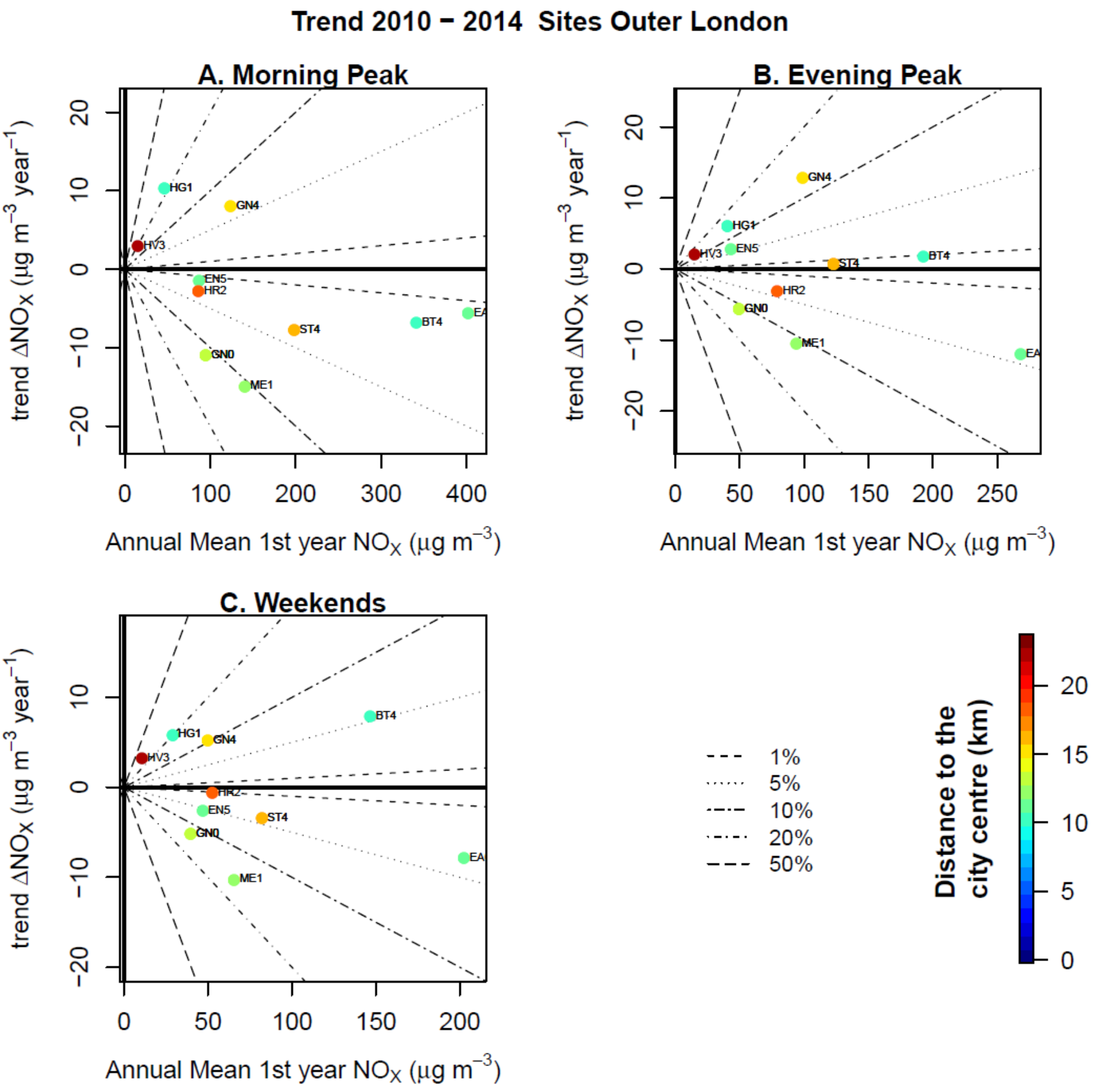
Period	Pollutant	75% data capture, no autocorrelation	75%data capture, corrected for autocorrelation	95% data capture, no autocorrelation	95%data capture, corrected for autocorrelation
2005-2009	ΔNO_X	0.87 [0.07, 1.68]*	0.94 [0.19, 1.69]*	1.02 [0.20, 1.84]*	1.07 [0.24, 1.91]*
	ΔNO_2	1.63 [1.25, 2.01]***	1.67 [1.28, 2.07]***	1.53 [1.05, 2.00]***	1.64 [1.16, 2.11]***
	ΔPM_{10}	-0.19 [-0.34, -0.03]*	-0.19 [-0.33, -0.04]*	-0.18 [-0.33, -0.03]*	-0.16 [-0.31,-0.02]*
2010-2014	ΔNO_X	-1.11 [-2.27, 0.04]+	-0.89 [-2.12, 0.34]	-0.90 [-1.96, 0.16]+	-0.58 [-1.75, 0.59]
	ΔNO_2	-1.65 [-2.27, -1.03]***	-1.48 [-2.13, -0.82]***	-1.29 [-1.82, -0.77]***	-0.97 [-1.50, -0.43]***
	ΔPM_{10}	0.07 [-0.13, 0.27]	0.05 [-0.13, 0.23]	0.03 [-0.22, 0.27]	0.06 [-0.19, 0.32]
	$\Delta PM_{2.5}$	-0.70 [-0.97, -0.42]***	-0.73 [-1.00, -0.46]***	-0.71 [-1.01, -0.41]***	-0.76 [-1.12, -0.40]***

****** trend significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant.***

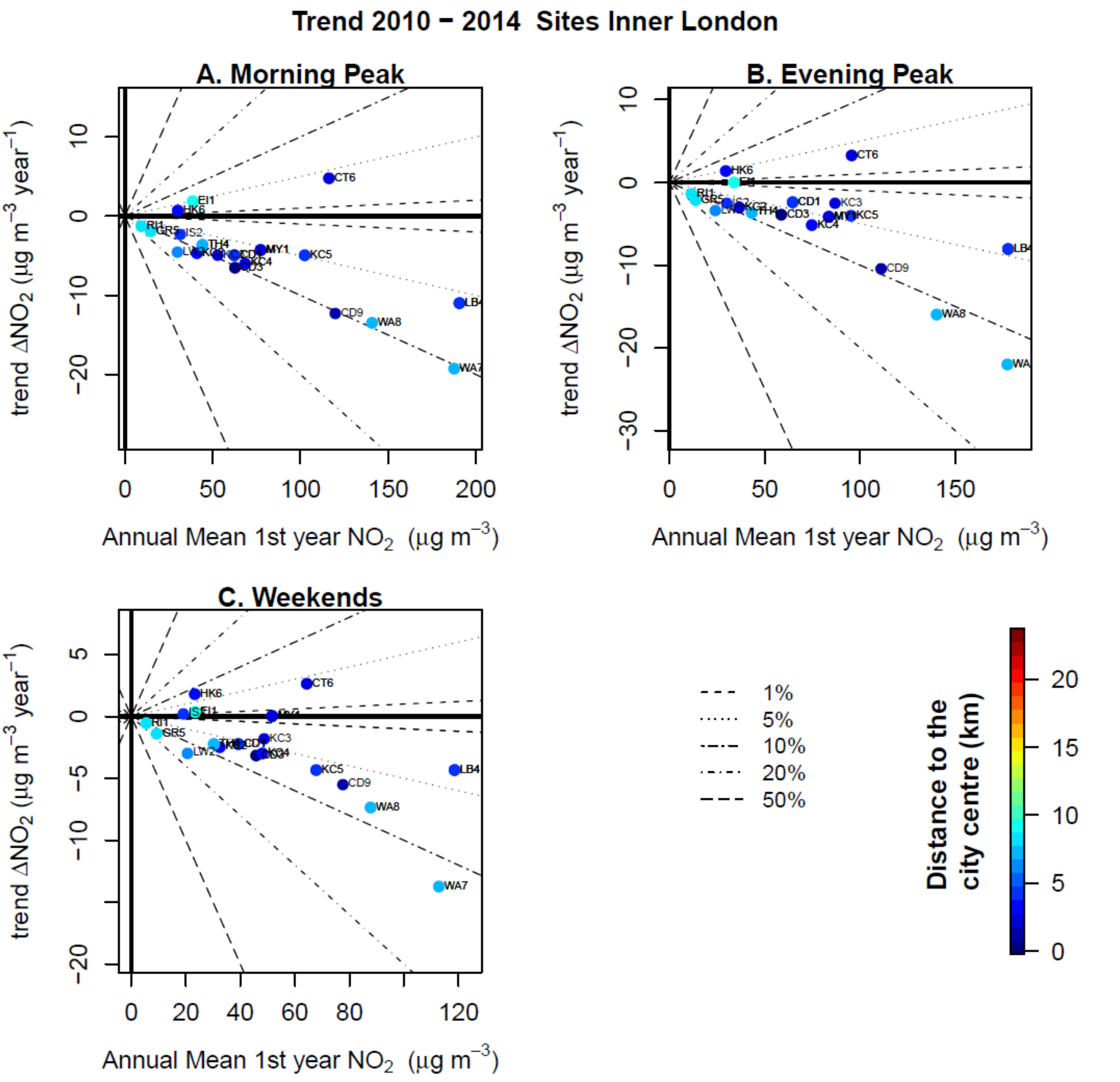
SM3. Trends in air pollutants for morning and evening weekday rush hour and weekends



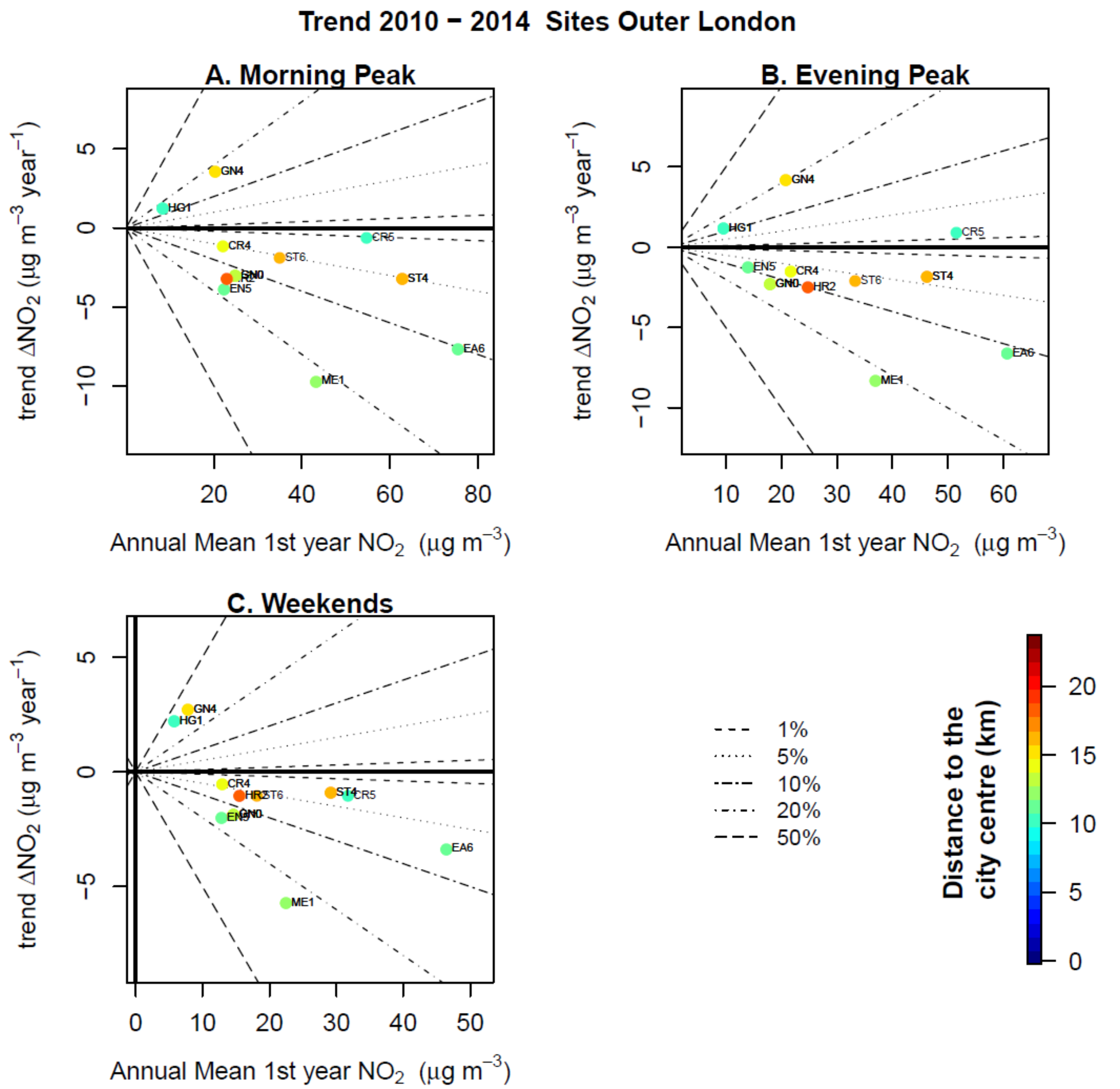
Supplementary Figure 3. Median trend for NO_X roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for inner London sites (<10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.



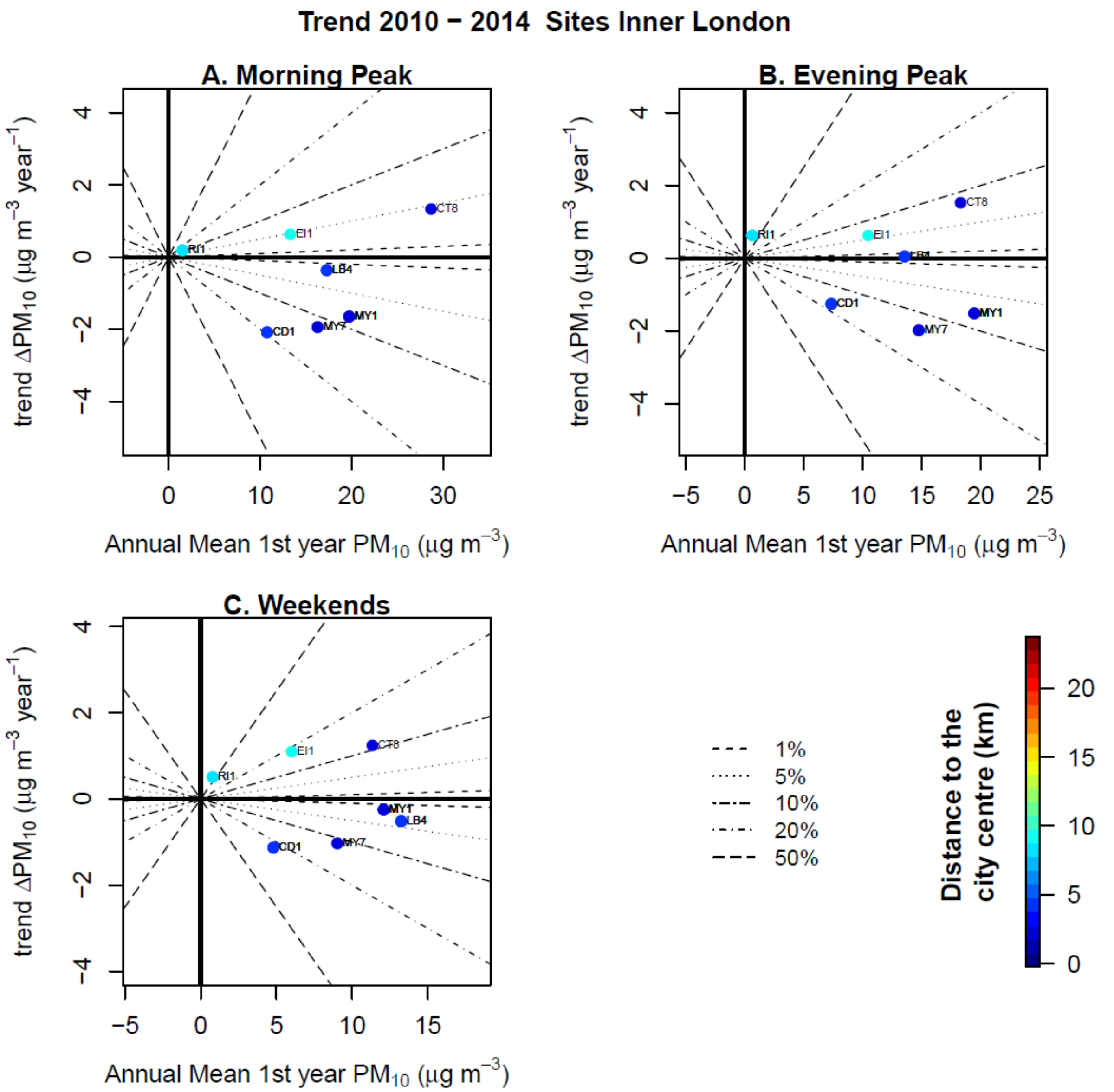
Supplementary Figure 4. Median trend for NO_X roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for outer London sites (>10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.



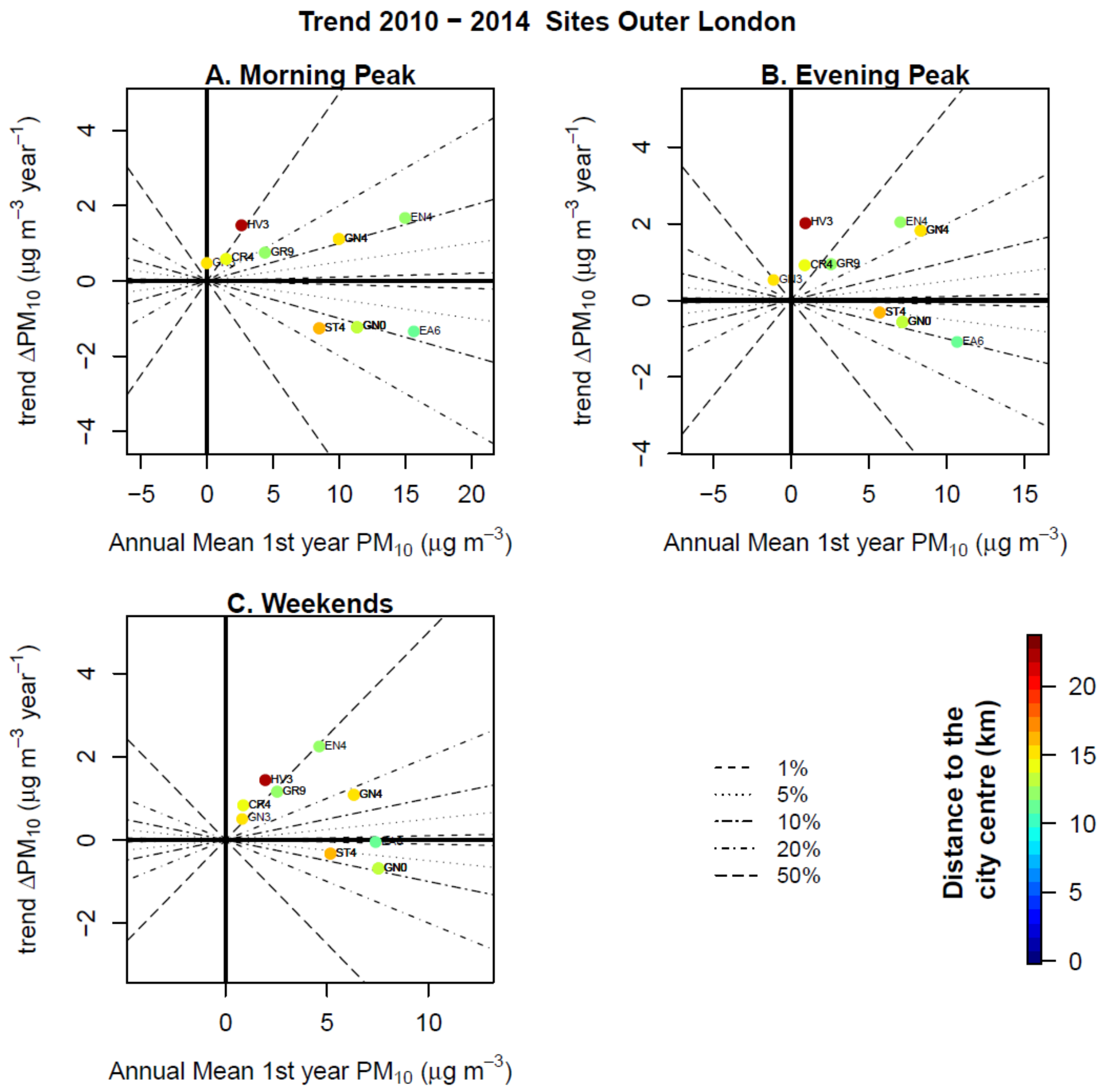
Supplementary Figure 5. Median trend for NO_2 roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for inner London sites (<10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.



Supplementary Figure 6. Median trend for NO_2 roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for outer London sites (>10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.



Supplementary Figure 7. Median trend for PM_{10} roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for inner London sites (<10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.



Supplementary Figure 8. Median trend for PM_{10} roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for outer London sites (>10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.

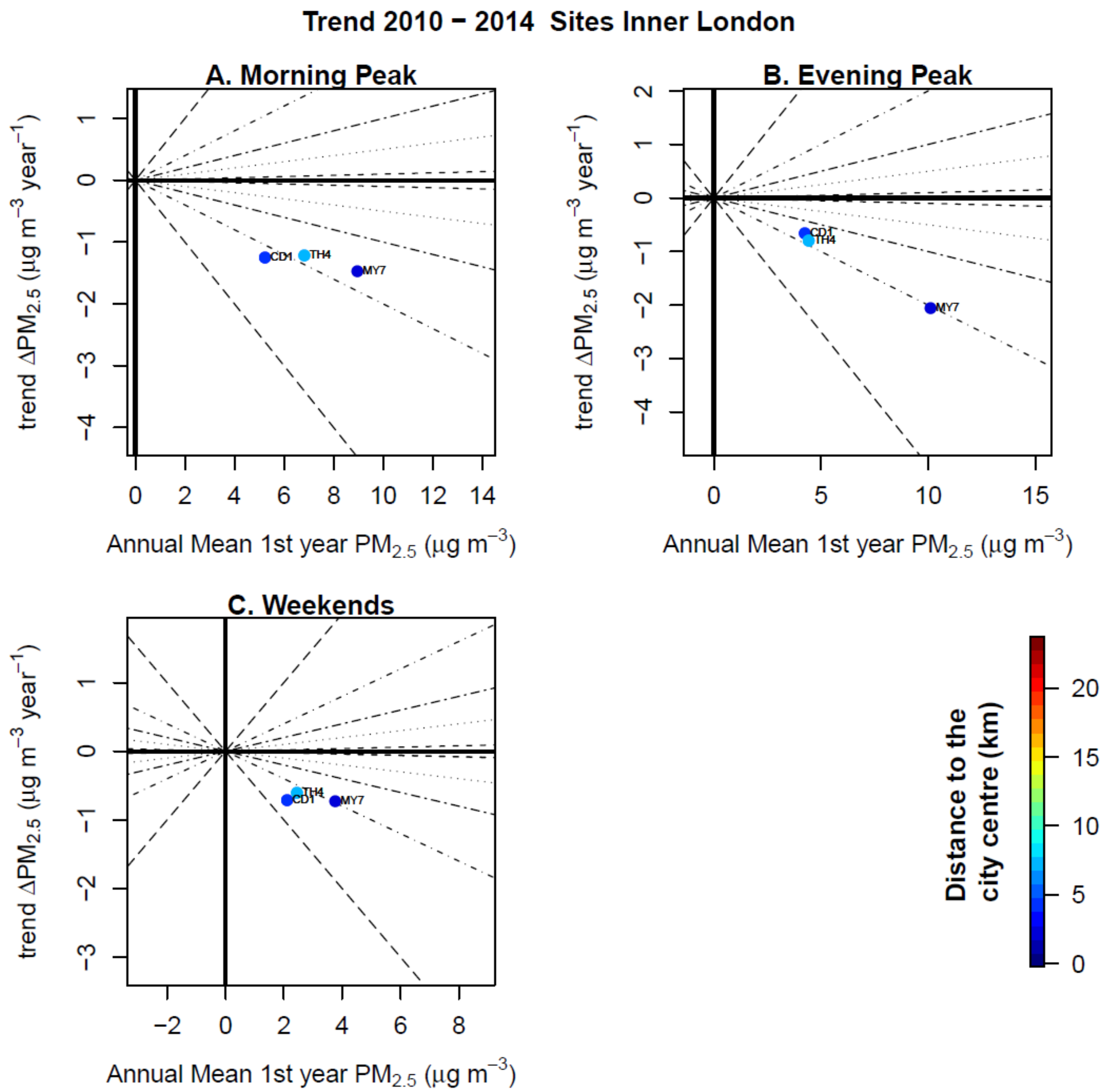
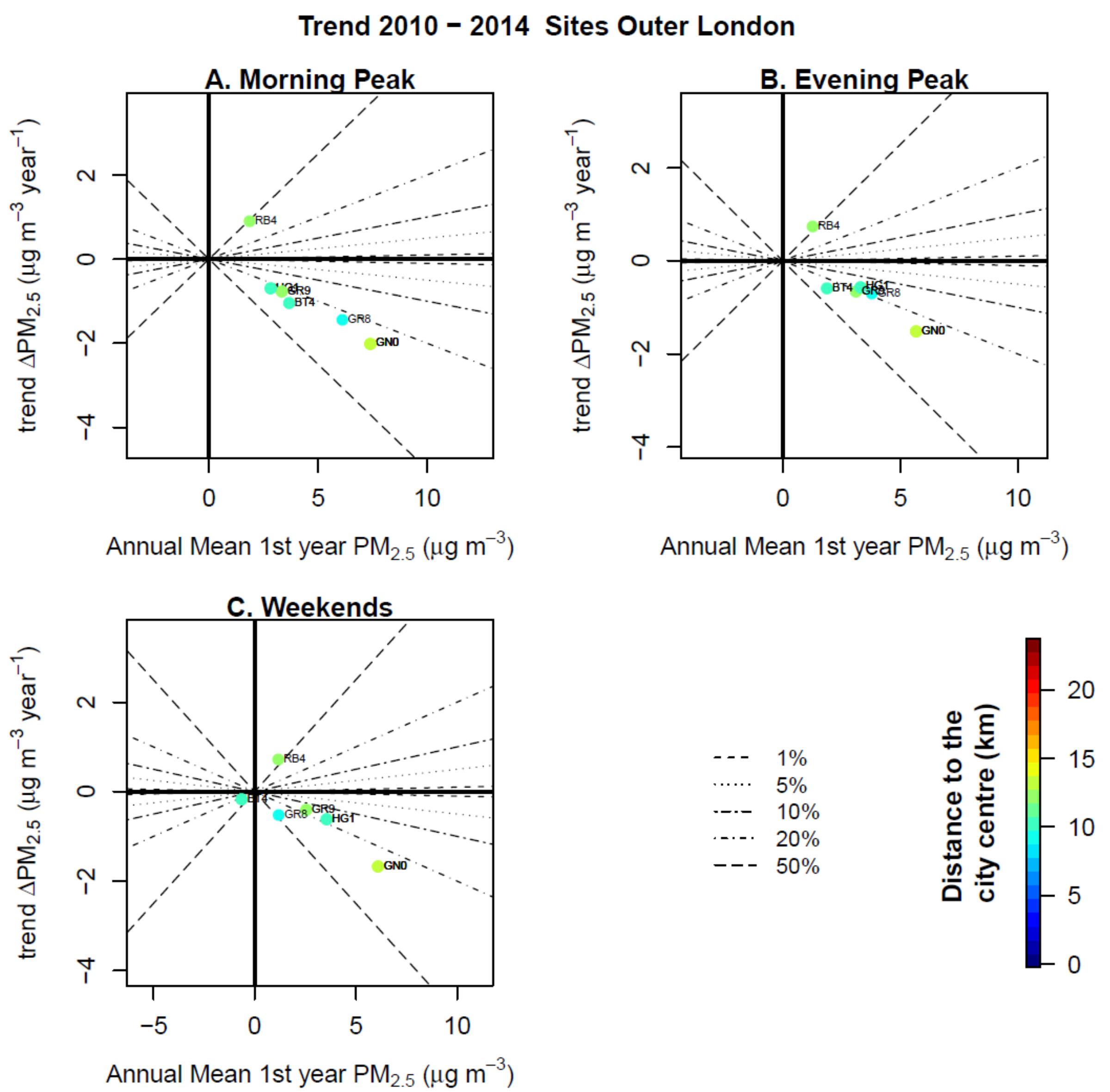


Figure 12. Median trend for $PM_{2.5}$ roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for inner London sites (<10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.



Supplementary Figure 9. Median trend for $PM_{2.5}$ roadside increments between 2010 and 2014 for sites with statistically significant trend against the mean roadside concentration in 2010 for outer London sites (>10 km from the city centre) for morning peaks (A), evening peaks (B) and weekends (C). Distance to the city centre is indicated by coloured points.