Reviewer 2

General: Well-written summary of NO2 monitoring data and trends across Europe. Ready for final publication as is. A few comments:

Response: We thank the reviewer for the time they spent reviewing the manuscript, and are pleased that they consider the paper ready for publication. Below we respond to each of the reviewer's specific comments in turn.

1) In terms of the drivers, it would have been interesting to assess correlations between actual met measurements (assuming some are nearby, if not onsite) to see if more robust assessments about the contribution drivers could be determined.

Response: We agree, it would be an interesting analysis to assess variability in NO_2 concentrations in relation to variation in measurements of wind speed, wind direction, temperature and other meteorological parameters. However, a number of fundamental steps are required before the meteorological measurements collected across Europe, and the network of atmospheric composition measurements are effectively able to be used to undertake the analysis the reviewer outlines.

Specifically, at the majority of atmospheric composition monitoring sites there is not co-located measurement of meteorological parameters. This has recently been highlighted in the UK Air Quality Expert Group's recent analysis of the utility of the UK air quality compliance monitoring network, and the need for collocated met and atmospheric composition data was one of the key recommendations from this report: 'For some scientific and research applications the evidential value of compliance data would be greatly enhanced through the co-measurement and reporting of meteorological parameters' (AQEG, 2015).

In the absence of sufficient co-located meteorological measurements, an assessment such as that outlined by the reviewer would then rely on meteorological measurements made at nearby stations. While at rural locations nearby met and atmospheric composition sites may have a relatively high degree of representativeness, a more careful assessment of the representativeness of the urban meteorological station location, in relation to urban background and urban traffic monitoring sites would be required before the met data could be considered applicable to comparison with NO_2 measurements. Previous studies have highlighted i) the large gradients in local scale meteorological conditions within urban areas (Kanda, 2007), and ii) the importance of local-scale meteorology in determining NO_2 concentrations (Carslaw and Carslaw, 2007). We therefore consider the incorporation of meteorological measurement analysis a very substantial additional piece of analysis beyond the scope of this work. Within the

existing text, we have referred to previous studies that assess the relationship between NO_2 concentrations and meteorological conditions at different types of sites (e.g. P14 L24).

However, the reviewer raises an important point about the importance of meteorological conditions in determining NO_2 concentrations, and we have therefore referenced the above AQEG report in the discussion, and reiterated the conclusion of that report that widespread co-location of met data alongside atmospheric composition monitors could increase the ability to assess the drivers of NO_2 (and other air pollutant) variability, especially at urban locations.

Additional Text P18 L23: 'Co-located meteorological measurements with atmospheric composition measurements, as recommended by AQEG (2015), would also allow meteorological drivers of annual NO_2 concentrations (and other impacts) to be assessed in more detail.'

2) Wonder if a companion meteorological clustering analyses would have proven valuable, in both the trends analyses and the composition/contribution analyses (i.e., during stagnation events does one see different trends, or greater contribution from rush hours)?

Response: As outlined above, the availability of suitable meteorological data across Europe is not sufficient to undertake a meteorological cluster analysis that would be comparable with the cluster analysis of NO_2 measurement data. Within the manuscript we refer to previous work that has assessed the importance of meteorological conditions in determining NO_2 concentrations, which gives some insight into the reviewers question. Schafer et al. (2006) assessed the relationship between boundary layer height and NO_2 concentrations in Hannover, Germany, and found substantially less correlation with boundary layer height and NO_2 concentrations at roadside sites than at urban background sites. Hence whether stagnation events produces different trends, such as greater contribution from rush hour periods may depend on the location of the sites (next to a road or removed from a road).

3) In the US, we often don't have as much confidence in the metadata associated w/ AQ monitoring sites (e.g., site environs change over time and metadata is not updated). Appears that isn't the case in Europe, but might be interesting to "doublecheck" urban/rural & traffic/background against emissions data, or satellite landuse, or population / other surrogates for emissions.

Response: In line with the reviewer's request, we have 'double checked' the relationship between site classification and population and gridded NO_x emission estimates. The population density from the GEOSTAT 2011 dataset

(http://ec.europa.eu/eurostat/web/gisco/geodata/reference-data/population-distribution-demography/geostat) in the 1 km grid in which the site was located was determined. Similarly, the total NO_x emission in the 0.1º grid containing each site was determined from gridded NO_x emissions developed by the European Monitoring and Evaluation Programme (EMEP) Centre for Emission Inventories and Projections (CEIP, http://www.ceip.at/new_emep-grid/01_grid_data_2014).

The resulting range of population density and NO_x emissions in the vicinity of different classifications of monitoring sites clearly show distinctions between rural and urban/suburban sites (Figures 1 and 2 below). There is less difference between population density and NO_x emissions between suburban and urban sites, and little distinction for background and traffic sites, although there is a distinction between rural industrial and other classifications of rural sites.

The lack of distinction across urban sites is likely due to the spatial resolution of the gridded population and NO_x emission datasets that are available with full European coverage. One km grids, and 0.1º grids in particular will contain both roadside and background locations within a city.

We do not think that it is necessary, or advisable, to reclassify sites to differ from those used in the AirBase data repository. This is especially the case as these official sites classifications have been used in previous analyses (e.g. Joly and Peuch, 2012), and will likely be used in future work given that these classifications are underpinned by EU legislation (referenced on P5 L13 of the main paper). Creating a discrepancy with how sites are classified in this work and other studies would be unhelpful to those attempting to consider this work in the context of previous studies. More useful are studies which specifically aim to assess the classification of sites based on different variables and methodologies, of which there are multiple examples for Europe, which are referred in the discussion of this work, and to which we have now directed readers at the point in which site classification is discussed in the Methods.

Based on the reviewer's suggestion, we have now referenced previous studies that have looked at monitoring site classification across Europe in the Methods. We have not reproduced Figures 1 and 2 contained in this response document in the main text or supplement of the manuscript because i) the key conclusions of the analysis would not be affected by a small number of sites having characteristics of a different site classification, ii) these are only two proxy variables for site classification, and there may have been changes that have occurred in the site environment that affect the classification that are not reflected either in a change in population of NO_x emissions at 0.1º grid size that would require site-by-site analyses to identify robustly, and iii) to keep focus on the key aspects of our paper, which relate to the contribution of hourly NO_2 variability to annual NO_2

concentrations across Europe. Specifically, for the NO_x emission figure (Figure 2 below), we have not included it because of the relatively coarser scale of the gridded NO_x emissions (0.1º grids). We want to avoid any confusion or suggestion that these gridded emissions represent the variability in NO_x emissions within the immediate environment of the site. This is unlikely to be the case given the spatial heterogeneity of NO_x emissions within a city at scales below 0.1º. We also note that this response document will be publicly available for those readers interested in understanding variation in these proxy variables in relation to the site classifications used in this work.

Additional text P5 L19: 'European monitoring site classification has been evaluated previously (Flemming et al., 2005; Joly and Peuch, 2012; Spangl et al., 2007).'

However, we have not included the NO_x emission figure (Figure 2 below) in the supplement because of the coarser scale of the gridded NO_x emissions (0.1º grids). We want to avoid any confusion or suggestion that these gridded emissions represent the variability in NO_x emissions within the immediate environment of the site. This is unlikely to be the case given the spatial heterogeneity of NO_x emissions within a city at scales below 0.1º.

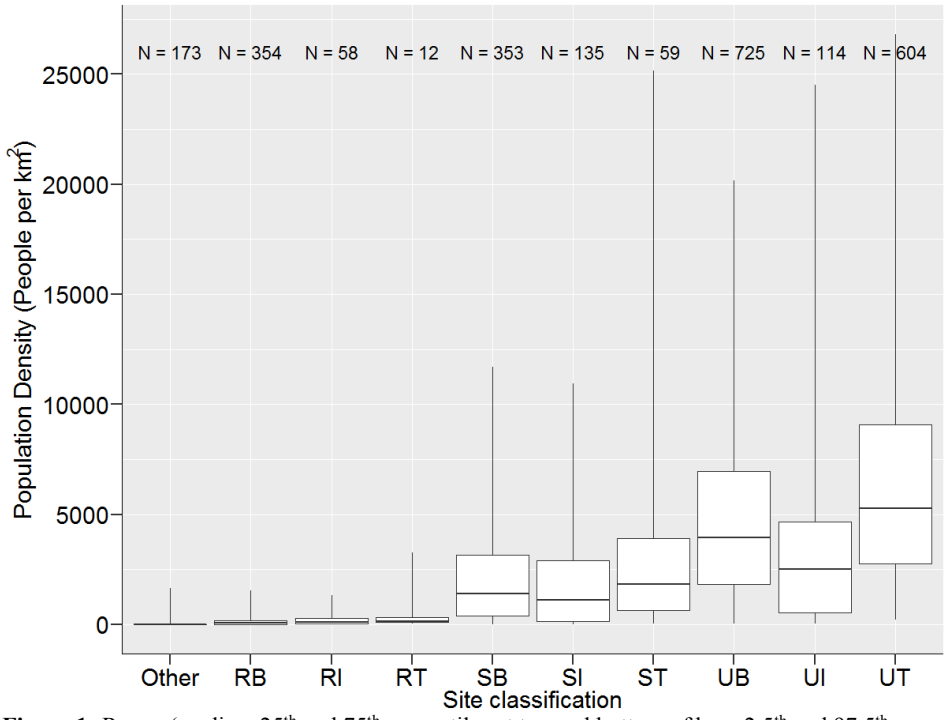

Figure 1: Range (median, 25th and 75th percentiles at top and bottom of box, 2.5th and 97.5th percentile at top and bottom of whiskers) of estimated population density in 2011 in the 1km grids in which NO_2 monitoring sites of different classifications are located. Classification abbreviations denote the site area (R = Rural, S = Suburban, U = Urban), and site type (B = Background, I = Industrial, T = Traffic).

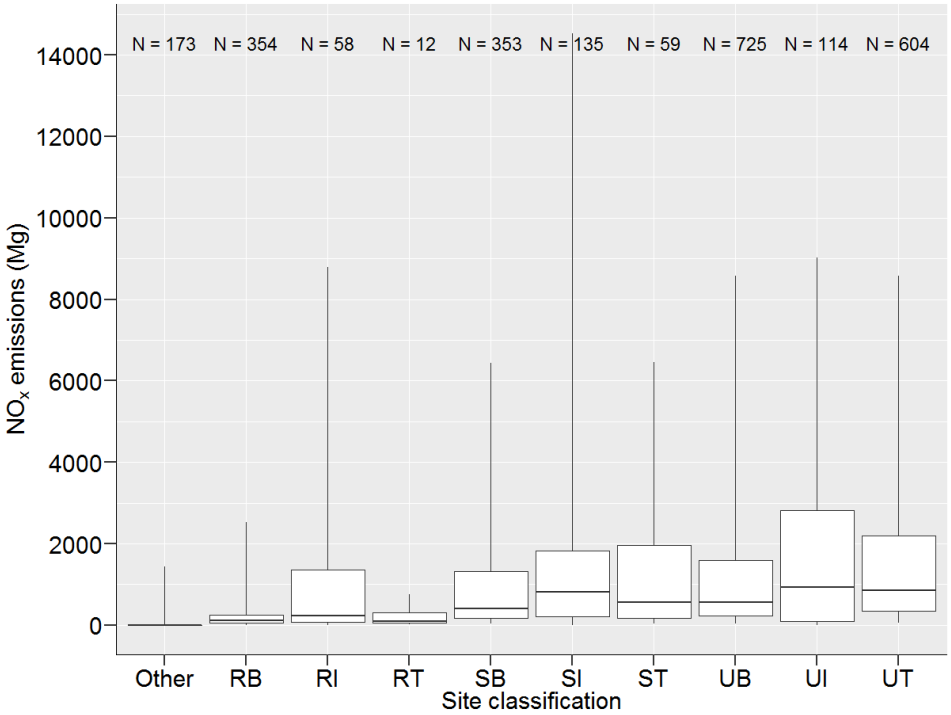

Figure 2: Range (median, 25th and 75th percentiles at top and bottom of box, 2.5th and 97.5th percentile at top and bottom of whiskers) of estimated NO_x emissions in 2014 in the 0.1º grids in which NO_2 monitoring sites of different classifications are located. Classification abbreviations denote the site area (R = Rural, S = Suburban, U = Urban), and site type (B = Background, I = Industrial, T = Traffic).

Specific:

Page 7: May want to combine the first two full paragraphs on page 7 w/ the last paragraph from page 6.

Response: Paragraphs have been combined in line with the suggestion.

Pag 12: Am struggling to understand sentence that starts on line 31. If those sites in N. Italy have annual averages dominated by winter months, why would photochemical drivers "be a more important" factor. Is this supposed to read "less important"?

Response: The sentence does not state that photochemistry is an important driver in producing *high* NO_2 concentrations. Rather it is stating that photochemical conversion of NO_2 to NO and O_3 may be a more important driver of what the concentration of NO_2 is at sites in northern Italy compared to other regions. In the context of a low summertime contributions to annual average NO_2 at northern Italian sites, this means producing low summer NO_2 concentrations (and increasing O_3 concentrations). We have rephrased to make this clearer.

Original text P13 L21: 'This indicates that in this region the photochemical conversion of NO_2 to NO and O_3 in summer may be a more important factor in determining NO_2 concentrations at these sites than at other sites across Europe with similar NO_{2AA} concentrations.'

Amended text P13 L21: 'This indicates that in this region the photochemical conversion of NO_2 to NO and O_3 in summer may be a more important factor in determining NO_2 concentrations (i.e. lowering NO_2 concentration during summer) at these sites than at other sites across Europe with similar NO_{2AA} concentrations.'

Page 13: add "a" before "heavily industrialised".

Response: We have amended the sentence in line with the suggestion.

References

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