

Interactive comment on “Dynamic Complex Network Analysis of PM_{2.5} Concentrations in the UK using Hierarchical Directed Graphs (V1.0.0)” by Parya Broomandi et al.

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29/05/2020 Dear GMD editorial board, Subject: Submission of revised paper gmd-2019-342 Thank you for your email dated 1st May 2020 enclosing the reviewers' comments. We have carefully reviewed the comments and have revised the manuscript accordingly. Our responses are given in a point-by-point manner below. Changes to the manuscript are highlighted. We hope the revised version is now suitable for publication and look forward to hearing from you in due course. Sincerely, Jong R. Kim, PhD, PE, CMP Professor Department of Civil and Environmental Engineering School of Engineering and Digital Sciences Nazarbayev University Nursultan (Astana), Kaza-

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They mention the need to “overcome the high dimensionality challenge” on line 70, but there are plenty of existing tools for studying the high dimensionality in atmospheric composition, chiefly among them chemical transport models. What does this work provide that more traditional simulation experiments cannot? Is it instead a demonstration of a new technique in atmospheric pollution research? If so, the authors should state this and demonstrate its utility in relationship to existing knowledge.

Response: (Updated in Paper to the discussion section): We are happy to provide more context here. For sure we are demonstrating a new technique that may be used alongside more established/traditional methods. The general framing of our approach is at the national level, trying to demonstrate (via a data-driven correlation and causal network), the statistical relationship between pollution data between multiple cities. This data-driven low-dimensional network enables us to examine seasonal trends and infer root causal mechanisms. This, we believe, is of a much lower complexity than deploying a chemical transport model of UK, where inference of causality remains challenging. Of course, what our model lacks is the relationship back to the physical flow models, and our future work would like to connect the models together.

We also respond in more detail (not in paper): As the reviewers are well aware, chemical transport models require emission inventory data (local or regionally originated) and a meteorological core to predict the dispersion and deposition of pollutants such as PM_{2.5}. Beside the notable amount of required data, high performance computing [HPC] platforms are required to deploy and evaluate model outputs, not least including experience with the pre and post processing software environments. In the current study we attempt to investigate the behaviour of PM_{2.5}, using a 2-dimensional (2D) network constructed from observational data alone. This leads to, first, the correlation

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network, and then a causation network. We can identify two things of note. (1) is the presence of root causations of pollution in certain seasons across a large region of UK, and (2) the stability of the transport network to potential disturbances. Both provide a level of simplistic insight at a very low complexity.

â€” The references are quite sparse for this manuscript. Additional background and motivational clarity should include more details of previous applications of causality in the geosciences (e.g. Ebert-Uphoff, Imme, and Yi Deng. “Causal Discovery for Climate Research Using Graphical Models.” *Journal of Climate*).

Response: The proposed study and also some other related references will be addressed in revised version of our manuscript. (Updated in the Paper to the discussion section): To infer causality, correlation-based methods such as lagged linear regression are already used in climate variability studies. This method can provide valuable information about causal relationships, but is susceptible to overreporting significant relationships when one or more of the variables has substantial autocorrelation (memory)(Ebert-Uphoff and Deng, 2012; McGraw and Barnes, 2018; Runge et al., 2017). On the other hand, Granger causality considers the autocorrelation of data and as a result is not susceptible to overreporting significant relationships. Since Granger causality is straightforward to calculate, it can be a preferred option to traditional lagged regression analyses when one or more datasets has substantial autocorrelation (memory). In addition, the establishment of a relationship between two variables is not sufficient in determining the true causality, but also determining the direction of causality is also needed; A more difficult task and challenge to overcome. The correlation-based methods cannot provide any information regarding directionality (but are still popular and useful for identifying lagged relationships among climate variables). However, the Granger approach has its own limitations as it does not account for mediating variables or indirect effects. Also, it requires assumptions of stationary and linear processes(Davidson et al., 2016; McGraw and Barnes, 2018; Wang et al., 2015, 2004). In previous studies, Nowack et al. (2020) showed that causal model evaluation provides

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stronger relationships for constraining precipitation projections under climate change as compared to traditional evaluation metrics for precipitation or storm tracks (Nowack et al., 2020). As a result, casual network analyses could be a promising tool to constrain long-term uncertainties in climate change projections. When a method relies on the assumption that previous model skill can be related to projected future changes will definitely suffer from certain limitations, including; the existence of some processes which are not (or not well) represented in current climate models and might become important in the future, and there is possibility that not all of relevant processes be well captured through the studied model (Nowack et al., 2020).

Materials and Methods Section 2.1 The description of the data in this section was inadequate for assessing the quality of the research in this work. How often were measurements taken? How was averaging done? What instruments were used? Was data quality assessed in any way? Is there a DOI or citation appropriate for any of the data used?

Response: How often were measurements taken? The measurement are taken from UK Automatic Urban and Rural Network (AURN) (<https://uk-air.defra.gov.uk/data/openair>). More information about UK Automatic Urban and Rural Network is available online from the DEFRA website (DEFRA, 2015).

How was averaging done? Data coverage were checked to have minimum missing data and having at least 75% of hourly based measured data for all stations, before averaging the hourly PM_{2.5} concentration. Only available data for 20 hours a day were averaged. While zero, NAN, and negative values were removed from the data set, and if the remained values were at least 20 hours a day, we averaged it representing the daily PM_{2.5} concentration, if not we report that day as NAN.

What instruments were used?

For PM_{2.5} measurement in UK monitoring system, for daily and hourly averaged concentrations, the instrument of FAI SWAM 5a was used by Defra (Defra approved

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instrument) which was certified to MCERTS (The Environment Agency's Monitoring Certification Scheme) for UK particulate Matter, and also certified to MCERTS for CAMs (Continuous Ambient Measurement Systems) of particulate Matter (<https://uk-air.defra.gov.uk/networks/monitoring-methods?view=mcerts-scheme>) (DEFRA, 2015). Reference equivalent method FDMS (Filter Dynamic Measurement System) is used for PM_{2.5} measuring at studied stations, which is allowed by EU for regulatory purposes (AQEG, 2012).

Was data quality assessed in any way?

Characterisation of PM_{2.5} temporal variability is important when it can help us to observe the high levels of pollutant causing health problems. Due to the data unavailability in the UK, it is not possible to conduct the historically long-term temporal trend analysis of PM_{2.5} (AQEG, 2005). Based on the AQEG (2012) report, there are no monitoring stations with long term (> 5years) using reference equivalent instruments for PM_{2.5} monitoring. From 2008–2009 onward, with the increase in the number of monitoring stations using reference equivalent method (such as FDMS allowed by EU for regulatory purposes) it is possible to study the temporal changes in PM (PM₁₀ & PM_{2.5}) in the UK (Munir, 2016). Minimum performance requirement for PM₁₀ & PM_{2.5} analysers were outlined in standard method of EN12341:2014 PM₁₀ and PM_{2.5} (EN16450:2017 Automatic PM analysers). These methods are proposed to ensure that measurement methods are complying with the DQO (Data Quality Objectives) set down in the Ambient Air Quality Directive (2008/50/EC) and in the amending Directive (EU) 2015/1480. The monitoring techniques used the UK's AURN for PM₁₀ & PM_{2.5} (with the exception of the automatic PM₁₀ analysers) are; Tapered Element Oscillating Microbalance, Beta Attenuation monitor, Gravimetric monitor, Filter Dynamics Measurement System (FDMS), Optical light scattering, and Fine dust Analysis System (FIDAS) (DEFRA, 2015). Is there a DOI or citation appropriate for any of the data used?

Yes, there is: Department for Environment, Food and Rural Affairs, United Kingdom:

<http://uk-air.defra.gov.uk/>, Last Access: 27 July 2015.

(Updated in the paper to the data availability section): Data availability: The measurements are taken from UK Automatic Urban and Rural Network (AURN) (<https://uk-air.defra.gov.uk/data/openair>). More information about UK Automatic Urban and Rural Network is available online from the DEFRA website (DEFRA, 2015). Data coverage were checked to have minimum missing data and having at least 75% of hourly based measured data for all stations, before averaging the hourly PM2.5 concentration. Only available data for 20 hours a day were averaged. While zero, NAN, and negative values were removed from the data set, and if the remained values were at least 20 hours a day, we averaged it representing the daily PM2.5 concentration, if not we report that day as NAN. For PM2.5 measurement in UK monitoring system, for daily and hourly averaged concentrations, the instrument of FAI SWAM 5a was used by Defra (Defra approved instrument) which was certified to MCERTS (The Environment Agency's Monitoring Certification Scheme) for UK particulate Matter, and also certified to MCERTS for CAMs (Continuous Ambient Measurement Systems) of particulate Matter (<https://uk-air.defra.gov.uk/networks/monitoring-methods?view=mcerts-scheme>) (DEFRA, 2015). Reference equivalent method FDMS (Filter Dynamic Measurement System) is used for PM2.5 measuring at studied stations, which is allowed by EU for regulatory purposes (AQEG, 2012). Characterization of PM2.5 temporal variability is important when it can help us to observe the high levels of pollutant causing health problems. Due to the data unavailability in the UK, it is not possible to conduct the historically long-term temporal trend analysis of PM2.5 (AQEG, 2005). Based on the AQEG (2012) report, there are no monitoring stations with long term (> 5years) using reference equivalent instruments for PM2.5 monitoring. From 2008–2009 onward, with the increase in the number of monitoring stations using reference equivalent method (such as FDMS allowed by EU for regulatory purposes) it is possible to study the temporal changes in PM (PM10 & PM2.5) in the UK (Munir, 2016). Minimum performance requirement for PM10 & PM2.5 analysers were outlined in standard method of EN12341:2014 PM10 and PM2.5 (EN16450:2017 Automatic PM analyzers). These methods are proposed to

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Section 2.2 – The software used (e.g. PAST, EVIEW) should be appropriately cited.

Response: – PAST software: (Hammer et al., 2001)

– EvIEWS (version 11) software: (Software, 2019)

– Line 139: What is this threshold for, how is it selected, and how is it calculated?

(Updated in the Paper to the Materials & Methods section): Based on previous similar study conducted in Switzerland to characterize the spatial distribution and seasonal changes of PM2.5 and PM10 concentrations using long-term monitoring data (Gehrig and Buchmann, 2003), we decided to choose 70% as our threshold cross-correlation.

Section 2.3 This section outlines a set of methods unfamiliar to the majority of the geoscientific modelling community. I suggest the authors include more details and relevant citations for the broader community should they be inclined to dig deeper into this sort of analysis. At the very least, this section should be carefully edited for clarity. The large number of parenthetical elements throughout the section make it challenging to parse what is being stated. The three sentences within a parenthetical statement on lines 150-155 are emblematic of this.

Response: The section is revised based on valuable comment provided by reviewer. (Updated in the paper to the Materials & Methods section): The Granger causality test statistically ascertains if one time series can cause the other. Thus to see that prior values of a time series contain the information about the future values of another time se-

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ries. This method was applied (using Eviews, version 11)(Software, 2019) to each pair of cities in the network during different seasons. Following this, statistically significant results ($p < 0.05$) were used to determine which time series contain information about the future values of another. The Granger Causality test assumes that both x and y time series (x and y represent PM2.5 concentration series for different stations in our network) are stationary, which was not the case in current study. As a result, de-trending was firstly employed before using the Granger Causality test(Papagiannopoulou et al., 2017, 2016). To retain the same degrees of freedom (DF) (mathematically, DF represents the number of dimensions of the domain of a random vector, or how many components should be known before the vector is fully determined.), with annual data, the lag number is typically small (1 or 2 lags). For quarterly data (in our case), the appropriate lag number is 1 to 8. If monthly data is available, 6, 12, or 24 lags will be used given enough data points. The number of lags is critical since a different number of lags lead to different test results. Consequently, the optimal lag number of 7 ensures the stability of model in this case study (based on Akaike Information Criterion (AIC). There is possibility of causation in one or both directions (x Granger-causes y and y Granger causes x). The chosen direction was based on the lowest p-value. For example according to our analysis, in spring we infer that ‘activities’ in Manchester is statistically influencing concentrations measured in Preston with a p-value= 5×10^{-29} , while Preston is statistically affecting Manchester with a p-value= 3×10^{-8} . Therefore, the first statement (pollution from Manchester is influencing Preston’s concentrations) is the correct one to be selected due to its lower p-value. Please note the language chosen reflects the statistical inference for the network analysis; However, the mapping of inference to atmospheric behavior and known challenges around PM2.5 source apportionment is important and discussed.

âĀĀ Line 149: Detrending a dataset does not always correct for non-stationarity. Please cite a reference for its appropriateness here.

Response: The following references are added to the manuscript:

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(Papagiannopoulou et al., 2017, 2016)

â€” Line 157: What is a lag used for in this case?

Response: The optimal chosen lag is based on the Akaike Information Criterion (AIC). This ensures the model will be stable, and we found a value of 7 (unitless) was appropriate in our study.

â€” Lines 345-348: It is not clear in the text why this basic description of a directed graph is in the middle of the section on Granger Causality?

Response:

The previous undirected graph indicates the existence of correlation. The directed graph shows the direction of potential causal mechanism (e.g. pollution from A leads the pollution from B, possibly indicating a transport process). We use the Granger causality to build the directed graph and go further to analyse its stability using hierarchical trophic coherence.

Section 2.4 The language regarding trophic coherence and sources/sinks is unreferenced and thus assumed to be an innovation of this work. All cities in this region are known to be atmospheric sources of PM2.5 due to anthropogenic and natural processes. The discussion here and elsewhere in the manuscript of only some cities classified as sources of PM2.5 is at odds with reality and should be clarified further.

Response: Yes, you are right!! But we looked at only one year to demonstrate the usefulness of our approach in the first instance. About site selection, again to demonstrate the usefulness of our approach we decided to conduct a small study first. Besides, we were trying to show the impact of regional sources on PM2.5 level in the UK, therefore decided to focus more on urban background sites. In some cities such as London, Birmingham, and Chesterfield we have various urban background sites, since we were not interested in inferring causality between sites across a city, the number of stations reduced in different cities. Focusing on larger networks and smaller regions is

something that can follow in future studies.

While trophic coherence has been shown to provide interesting results for the analysis of food webs, it's not clear from the text that this is the most appropriate method for assessing important vertices in a causal graph for PM2.5. Why was it used here?

Response: Trophic coherence is used to analyse the general directionality coherence of the causal network. If we had perfect coherence ($q=0$), then there is a source of pollution that is affecting others. If we had perfect incoherence ($q=1$), then all the cities are polluting each other equally. This gives us an idea of both the nature and geography of the transport ecosystem for different seasons, as well as its stability.

On line 39 in the abstract, the authors claim that winter is the most coherent of the seasons. They attribute this to meteorological features like wind speeds and inversions. Table 3 shows that summer has nearly the same incoherence factor as winter, and yet this is not discussed at all in the manuscript. Given the vastly different meteorological features in summertime, does this influence the interpretation that meteorology is a driving factor?

Response: (Updated in the Paper to the Results section): Table 3 shows a similar incoherence factor for winter and summer. With a q value of 0.3-0.4, this suggests having a single source of pollution and similar stability. The summer and winter periods have similar values but different sources. Figure 4B (summer) suggests that the source of network are Liverpool, London Road, Southampton, Norwich, and Birmingham. While, in winter (Figure 4 D), the sources of network are Chesterfield, Manchester, Preston, London Bexely. Winter is also inferred to be represented as a national network, while summer is more local.

We would also add that, according to the figures 2-4, the less incoherent network was witnessed during wintertime comparing to the rest of seasons. During springtime, due to well mixing of the lower atmospheric layer, the network was well formed. On the other hand, during wintertime, the meteorology is characterized by frequent inversions, form-

ing an efficient obstacle for the distribution and homogenization of PM. As a result, only tight spatially embedded parts of network (below 100Km with the highest percentage of restored network) could 'withstand' meteorological influences and larger distances across the network (above 100Km) started to collapse from a network perspective. In winter time, the plausible reason of connecting the cities out of the initial network (81% of connected cities were out of the initial network with distance above 200 Km) might be higher average seasonal wind speeds (in all studied stations), probably due to the balance among greater dilution and shorter transport times at higher wind speeds, which allows less time for PM dispersion and deposition over further distances(Harrison et al., 2012).

Discussion In general, this discussion seems incomplete and largely conjecture without appropriate referencing or analysis presented herein.

â€” Lines 378-381: What in the previous analysis indicates that meteorological conditions and diurnal emissions from regional sources dominate? I acknowledge that this is known the be case throughout the field a priori, but it's not clear how this analysis in this manuscript leads to that conclusion.

Response: Harrison et al., (2012) showed how meteorological parameters with the focus on wind speed and wind direction can influence PM_{2.5} level in the UK and provide insight into the origin the measured PM_{2.5} concentrations. Based on their study, a notable consistency in the patterns across the UK exists. When the winds are coming the south-southeast clockwise through to north, the PM_{2.5} concentrations are generally lower than the annual average value, while when the winds are coming from the northeast through to southeast, the PM_{2.5} concentrations are higher than the annual average value (Harrison et al., 2012). In addition, secondary aerosols secondary sources are dependent on meteorological conditions, and the abundance of precursors, that can have a significant contribution in PM_{2.5} concentrations in the UK, where a large proportion of transboundary secondary PM_{2.5} are transferred from different parts of Europe (AQEG, 2012; Vieno, et al., 2016). As a result, one plausible reason

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of connection within a network can be common transboundary sources.

âĀĀ Lines 399-409: How are these relationships known without detailed trajectory modelling? The fact that the causal networks are consistent with known transport mechanisms is interesting but does not provide evidence for these sweeping assessments of pollutant transport.

Response: The reviewer is correct. We did not conduct detailed trajectory modelling but tried to interpret our casual network based on previous studies and try to explain the reason behind generated clusters in south and north of the UK. Inferring causal mechanisms from data is not new, and the fact that our findings corroborate with previous detailed modeling and known qualitative causal mechanisms we hope demonstrates the usefulness of this approach.

âĀĀ Lines 410-415: There is not evidence presented in this manuscript that this transport mechanism is attributable to PM2.5 variability in the Northern UK.

Response: Our results (Figures 2-4) showed coherence throughout the patterns across Group A and Group B in the UK. Based on previous studies, high PM2.5 concentrations in Group B (southern sites) are more attributed to winds from the east through to southeast, which are often attributed to a blocking high pressure over the Nordic countries, giving rise to a south-easterly or easterly air flow that cause transportation of emissions from eastern Europe, northern Germany, and the Belgium and Netherlands to the southern cities in the UK. High PM2.5 concentrations in Group A (northern cities or close to northern part) are attributed to the winds blowing from the northeast through to east, drawing air flow (likely to start blowing when a low pressure runs up the English Channel) northward across European emission sources (to mainly be emission sources of precursors of secondary PM), out into the North Sea, then reaching northern parts of the UK from a north-easterly direction (Barry, R.G., Chorley, R.J., 2010; Harrison et al., 2012).

Conclusions âĀĀ Line 423-425: The authors haven't shown any results related to pre-

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dicting future PM_{2.5} between cities. Response: We apologise for this confusion and have removed the word “predict”. Past values have information which is statistically significant to future values. We use this in our causal analysis, but we do not make active predictions, only statistical inferences.

âĀĀ Line 439-431: This work is not the first to demonstrate that meteorology drives much of the variability in PM_{2.5}. It is not clear from this manuscript how connections beyond conjecture can be made to meteorological variability and PM_{2.5}. Response: This is a fantastic point and as we touched on in our earlier response, our hope is to link our network findings back to meteorology findings and models. What we are keen to show and add to is that there is a topological aspect, which highlights the complex web of cascade pollution transport between cities.

Minor Comments âĀĀ Line 78: “Atmospheric particulate matters” should be “Atmospheric particulate matter” Response: Corrected and highlighted in text.

âĀĀ Figure captions should contain more detail regarding the content of the figured. For example in Figure 2, what do edge thicknesses correspond to? Response: In the revised version of figures, we will try to add more details. âĀĀ Line 232: “%50” should be “50%”. Response: Corrected and highlighted in text.

âĀĀ Line 393: “Primary” should be “Primary”. Response: Corrected and highlighted in text.

References: AQEG, 2012. Fine Particulate Matter (PM_{2.5}) in the UK. AQEG, 2005. Particulate Matter in the United Kingdom. London. Barry, R.G., Chorley, R.J., 2010. Atmosphere, Weather and Climate, ninth ed. ed. Routledge, Abingdon. DEFRA, 2015. Department for Environment, Food and Rural Affairs, United Kingdom. Gehrig, R., Buchmann, B., 2003. Characterising seasonal variations and spatial distribution of ambient PM₁₀ and PM_{2.5} concentrations based on long-term Swiss monitoring data. Atmos. Environ. 37, 2571–2580. [https://doi.org/10.1016/S1352-2310\(03\)00221-8](https://doi.org/10.1016/S1352-2310(03)00221-8) Hammer, O., Harper, D., Ryan, P., 2001. PAST: Paleontological Statistics Software

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Package for Education and Data Analysis. *Palaeontol. Electron.* 4, 1–9. Harrison, R.M., Laxen, D., Moorcroft, S., Laxen, K., 2012. Processes affecting concentrations of fine particulate matter (PM_{2.5}) in the UK atmosphere. *Atmos. Environ.* 46, 115–124. <https://doi.org/10.1016/J.ATMOSENV.2011.10.028> Munir, S., 2016. Analysing temporal trends in the ratios of PM_{2.5}/PM₁₀ in the UK. *Aerosol Air Qual. Res.* 17, 34–48. <https://doi.org/10.4209/aaqr.2016.02.0081> Papagiannopoulou, C., Decubber, S., Miralles, D.G., Demuzere, M., Verhoest, N.E.C., Waegeman, W., 2017. Analyzing Granger Causality in Climate Data with Time Series Classification Methods BT - Machine Learning and Knowledge Discovery in Databases, in: Altun, Y., Das, K., Mielikäinen, T., Malerba, D., Stefanowski, J., Read, J., Žitnik, M., Ceci, M., Džeroski, S. (Eds.), . Springer International Publishing, Cham, pp. 15–26. Papagiannopoulou, C., Miralles, D., Verhoest, N., Dorigo, W., Waegeman, W., 2016. A non-linear Granger causality framework to investigate climate–vegetation dynamics. *Geosci. Model Dev. Discuss.* 1–24. <https://doi.org/10.5194/gmd-2016-266> Software, Q.M., 2019. Eviews, Version 11.

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