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Geoscientific Model Development Discussions

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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gmd-2019-342-RC2#

âĂć the small subset of data used (e.g. 15 sites in 14 cities) Response: Yes, you are right!! But 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 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.

âĂć the small temporal period considered (just 1 year) Response: Yes, you are right!! But we looked at only one year to demonstrate the usefulness of our approach in the first instance. âĂć small about of parameters considered (solely PM observations and a national inventory of emissions) Response: As we mentioned in Conclusion, our study has some limitations. As a result, to have a better understanding of the network, evaluating a predictive network based PM2.5 model using meteorological parameters, and contributions from identified clusters in the UK, would be helpful and will be investigated in our future research.

âĂć the lack of consideration of known variables that influence PM (e.g. meteorology) Response: As we mentioned in Conclusion, our study has some limitations. As a result, to have a better understanding of network, evaluating a predictive network based PM2.5 model using meteorological parameters, and contributions from identified clusters in the UK, would be helpful and will be investigated in our future research.

âĂć The insufficient of placing this work in the context of existing related publications Response: We thank the reviewer for identifying further references to add to this work.

Based on the above issues raise, we change our paper as demonstrated in the following text.

(Updated in the paper to the introduction section): A number of studies have deployed a range of techniques to overcome challenges in computational performance of chemical transport models. Solving atmospheric chemical kinetics is a stiff numerical problem, with choice of solvers used reflecting the need to ensure numerical stability(Sandu and Sander, 2006). Consequently, the integration of the chemical kinetics takes 50%–90% of the computational cost of an atmospheric chemistry model such as GEOS-Chem (Eastham et al., 2018; Hu et al., 2018; Nielsen et al., 2017). Dynamical reduction (adaptive solvers) in solving the chemical mechanism was previously demonstrated to increase the efficiency of the integration at the expense of a reduction in accuracy (Cariolle et al., 2017). Other attempts to reduce the computationally of chemical kinetics include repro-modelling (approximation of the chemical kinetics using polynomial functions)(Turányi, 1994), quasi-steady state approximation(Whitehouse et al., 2004), and separation of fast and slow species(Young and Boris, 1977). Other studies use reduced chemical mechanisms with fewer species (Kelp et al., 2018; Whitehouse et al., 2004). Recent attempts have also used machine learning to replace the use of traditional integrators (Porumbel et al., 2014). For example, using a neural network emulator for an atmospheric chemistry box model, an order-of-magnitude speed up was found, but the new implementation suffered from rapid error propagation when applied over multiple time steps (Kelp et al., 2018). Numerical emulators have also been used to directly forecast air pollution concentrations across future time steps (Mallet et al., 2009). This approach was also applied in chemistry– climate simulations with the focus on model forecasting of time averaged concentrations of selected species such as OH (hydroxyl radical), and O3 (ozone) over timescales of days to months(Nicely et al., 2017; Nowack et al., 2018). Keller and Evans (2019) studied the replacement of suitably trained machine-learning based approach (random forest regression) for the gas-phase chemistry in atmospheric chemistry transport models (GEOS-Chem). As noted within this particularly study, this approach suffers also from some limitations in-

cluding; (a) being only applied within the range of data used for the training, (b) studying scenarios with significant changes in the emissions (being outside of used data for the training) can lead to inaccurate predictions by the model, and (c) machine learning algorithm may not capture model resolution caused by the non-linear nature of chemistry (the numerical solution of chemical kinetics is resolution-dependent)(Keller and Evans, 2019).

âĂć Many years of data are freely available for hundreds of sites across the UK. Why have only 15 sites from 14 locations been considered here? Why were these 15 sites (listed in the SI) chosen above the hundreds of others? Are the authors saying they are representative of the sites? Why only 15 sites? Is it due to limitations in a computer resource or does this approach not scale well? If this is the case could a representative average be taken of the sites in a city? Why has a mixture of types been used (e.g. Traffic urban, Background suburban, Background Urban, and Industrial Urban)? What impact could the choice of data set have on the conclusions? 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 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.

âĂć All of these issues need to be explored through sensitivity testing. Response: Our data enables us to construct an undirected correlation and a directed Granger causality network, using PM2.5 concentrations in 14 UK cities over a year-long period. We show for both reduced-order cases, the UK is divided into two northern and southern connected city communities, with greater spatial embedding in spring and summer. We go on to infer stability to disturbances via the network trophic coherence parameter, whereby we found that winter had the greatest vulnerability. As a result of our novel graph-based reduced modelling, we are able to represent high-dimensional knowledge into a causal inference and stability framework. Our statistical p values demonstrate confidence in results which embeds robustness. We would like to expand this further, but we cannot do it at the end of an 8-month manuscript review due to realistic researcher employment and practical reasons. Had this been raised much earlier, we may have had the resources.

âĂć This analysis must be expanded in breadth (# sites) and depth (e.g. years of data) if it is to be considered a case study or proof of concept is. 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 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.

âĂć How many air quality events happened in this year? What does this approach show about these events? Often pollution events in the UK are synoptic in scale, so would affect multiple cities. It would be interesting to see what information could be drawn out from this analysis. Response: The paper is showing correlations and potential causal pathways inferred from data across UK cities. We show for both reduced-order cases, the UK is divided into two northern and southern connected city communities, with greater spatial embedding in spring and summer. We have not studied certain potential air quality events and are they synoptic in scale and how they can influence different cities. In further investigations it would be an interesting lead to follow.

âĂć The discussion needs to be expanded to explain what the pros and cons of this approach are over the generally taken approaches (e.g. chemical transport models). Also consider this approach against other non-explicit approaches taken in this field (e.g. Nowack et al 2020, Keller et al 2019). ML methods that consider much more input data (e.g. meteorology, emissions, chemistry) have already been demonstrated in a much more thorough way within this journal (e.g. Keller et al 2019). Also make it clear what new information that this approach will provide and why this would be advantageous above these other approaches or complementary to them.

Response: (Updated in Paper to the discussion section): 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 data from multiple cities. This data-driven, low-dimensional, network enables us to examine seasonal trends and infer root causal mechanisms. We believe this approach requires evaluations across multiple scales. Nonetheless, we believe this approach will offer an additional approach to traditional models 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 will incorporate this. Machine Learning models are used to predict, but we are here to infer causality and demonstrate topological patterns via the network. 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 PM2.5. 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 PM2.5, using a 2-dimensional (2D) network constructed from observational data alone. This leads to, first, the correlation 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. (Updated in the paper to the introduction section): Previous studies tried different techniques to overcome difficulties in simulation of atmospheric chemistry transport processes. One of the faced challenges is about computationally expensive nature of these models. The numerical solution of chemical kinetics is computationally expensive due to the numerically stiff equations needs implicit integration schemes (like Rosenbrock solvers) to ensure numerical stability(Sandu and Sander, 2006). Consequently, the integration of the chemical kinetics takes 50%–90% of the computational cost of an atmospheric chemistry model such as GEOS-Chem (Eastham et al., 2018; Hu et al., 2018; Nielsen et al., 2017). Involving dynamical reduction (adaptive solvers) in the chemical mechanism was previously tried methods to increase the efficiency of the integration, associated with accuracy reduction(Cariolle et al., 2017). Some other previous attempts to reduce computationally cost chemical kinetics are repro-modelling (approximation of the chemical kinetics using polynomial functions)(Turányi, 1994), quasi-steady state approximation(Whitehouse et al., 2004), and separation of fast and slow species(Young and Boris, 1977). Some other studies tried to simplify the chemistry causing a decrease in the number of species and reactants (Kelp et al., 2018; Whitehouse et al., 2004). Using machine learning, the chemical integrator was replaced for other chemical systems and were faster than solving the ODEs (chemical systems like those found in combustion) (Porumbel et al., 2014). Recently, using a neural network emulator for an atmospheric chemistry box model, an order-of-magnitude speed ups was found, but it suffered from rapid error propagation when applied over multiple time steps (Kelp et al., 2018). Machine learning emulators have also been tried to directly forecast the air pollution levels in future time steps(Mallet et al., 2009). This approach was also applied in chemistry– climate simulations with the focus on model forecasting of time averaged concentrations of selected species such as OH (hydroxyl radical), and O3 (ozone) over timescales of days to months(Nicely et al., 2017; Nowack et al., 2018). Keller and Evans (2019) studied the replacement of suitably trained machine-learning based approach (random forest regression) for the gas-phase chemistry in atmospheric

chemistry transport models (GEOS-Chem) compares well to the standard modelling. The new approach was to forecast the concentration of each transported specie including NOx and O3 (Keller and Evans, 2019), made it comparable to previous attempts in speeding up the solution of the chemical kinetics through more efficient integration. Current approach suffers also from some limitations including; (a) being only applied within the range of used data for the training, and applying the method outside of this range can cause inaccurate outputs, (b) studying scenarios with significant changes in the emissions (being outside of used data for the training) can lead to inaccurate predictions by model, and (c) machine learning algorithm may not capture model resolution caused by the non-linear nature of chemistry (the numerical solution of chemical kinetics is resolution-dependent)(Keller and Evans, 2019).

Line 102 - This is sentence is far too strong. The authors have not demonstrated that this technique could be this useful. “This is a timely study as strategic investments in national and local air quality monitoring networks require an evaluation on the usefulness, or not, of network design” Response: We are not sure why the reviewer feels this sentence is too long or is invalid. We are not claiming the method can be used immediately but have submitted this study for scientific peer review. We use a case study of selected sites to confirm whether inferred causality makes sense based on known relationships in PM2.5 and variable conditions and transport mechanisms. Much like studies demonstrating the potential use of ML variants of existing models are far from wide scale adaption, for reasons discussed in the literature, we feel our study demonstrates potential for the method presented here.

âĂć Line 112 - See earlier point about the limitations of data used. 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 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.

âĂć Line 117 - Was this emissions inventory used for the same year? Given more details here. Response: At the current stage we decided not to use NAEI data and will be deleted from revised version in the materials & methods section.

âĂć Line 163 - What is the link to the broader picture here? If we accept that a background urban site in Manchester can be used to predict future concentrations in Preston, is the suggestion that something like this could be done for other sites and a national level of prediction gained at a computational cheap cost? Response: Yes, there is a possibility that we can understand how UK cities cross-pollute across regional and national distances, and we are now investigating it in our ongoing research.

âĂć Line 424 - There is no supporting evidence given for this predictive capacity. 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.

Technical points

âĂć Title / other text - Some lines are highlighted blue. Why? Response: We had a minor revision based on editor's comment asked us to revise and highlight revised parts, which is reason for blue highlighted parts through the manuscript. âĂć Table 2 - typo in units? (Kelvin metres instead of kilometres?) Response: Corrected in the text. âĂć Figure 1 - resolution needs to be improved. Most of UK coastline should be shown for context. Which map layer was used? Response: The resolution will be improved in revised manuscript. âĂć Figure 2 - resolution needs to be improved. Response: The

resolution will be improved in revised manuscript.

âĂć Figure 3 - resolution needs to be substantially improved. Axis labels are not large enough or readable. Response: The resolution will be improved in revised manuscript.

âĂć Line 9 - Specify the region over which the deaths occur. Europe? Global? Response: Global âĂć Line 32 - Have people not already been successful in doing this? (e.g. the climate science community - Nowack et al 2020) Response: The causal networks in Nowack et al are causal networks between confounding factors (an analogy here would be: causal network between industrialisation, production, and air pollution). Our causal networks are geographic, in that we are looking at whether cities causally influence each other. âĂć Line 34 - "12 month", "52 week" are used interchangeably in the manuscript: would a single phrasing (e.g. year-long) be easier for the reader? Response: Sure!! Corrected and highlighted through the text. âĂć Line 35 - Sentence does not scan. superfluous "two" and "a"? Response: Checked and corrected in the text.

âĂć Line 80 - Globally? The focus of the article is the UK, so you need to be more specific. Response: The following part will be added to the text;

(Updated in the paper to the introduction): In the UK, long-term exposure to PM from anthropogenic sources has an impact on the equivalent of around 29,000 deaths a year(COMEAP, 2010; Gowers etal., 2014). Also, short-term exposure to air pollution events can increase the daily emergency hospital admissions (for cardiovascular and respiratory conditions) and mortality(Macintyre et al., 2016). Focusing on two air pollution events (12– 14 March and 28 March–3 April 2014) with the highest PM2.5 concentrations, about 600 deaths were brought forward from short-term PM2.5 exposure, representing 3.9% of total all-cause death during these 10 days.

âĂć Line 112 - This time-split would be better placed in a table and in the SI. It is not a very readable way of presenting this information. Response: Table S2 including time-split data created and added to the supplementary material.

âĂć Line 116 - Was data downloaded for the same period. This sentence does not need to start with" also". Response: At the current stage we decided not to use NAEI data, it was left here by accident and will be deleted from revised version. âĂć Line 393 - formatting error of "y" in word primary. Response: Corrected and highlighted in text. âĂć Data availability - Add a data availability section. Include specific details on which version of data was used and how to access this. Response: Data availability section will be added to the revised version, including all above-mentioned details. (Updated in the Paper to the data availability section): Data availability section: The measurement were hourly based 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 be 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).

Characterisation 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 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 & PM2.5 (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). âĂć References - remove full stops at starts of lines. Response: Checked and corrected in the text.

âĂć S1 list - please update the information to be in a table format. The current format is unwieldy, not read-friendly. Response: Table S including the studied Monitoring stations' data created and added to the supplementary material.

âĂć Citations Keller, C. A. and Evans, M. J.: Application of random forest regression to the calculation of gas-phase chemistry within the GEOS-Chem chemistry model v10, Geosci. Model Dev., 12, 1209–1225, https://doi.org/10.5194/gmd-12-1209-2019, 2019. Nowack, P., Runge, J., Eyring, V. and Haigh, J.D., 2020. Causal networks for climate model evaluation and constrained projections. Nature communications, 11(1), pp.1-11. Interactive comment on Geosci. Model Dev. Discuss., https://doi.org/10.5194/gmd-2019-342, 2020. Response: Proposed references will be added to the reference section and will be addressed in our manuscript in revised version.

References: AQEG, 2012. Fine Particulate Matter (PM2.5) 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. COMEAP, 2010. The Mortality Effects of Long-term Exposure to Particulate Air Pollution in the United Kingdom. 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 PM2.5 concentrations based on long-term Swiss monitoring data. Atmos. Environ. 37, 2571–2580. https://doi.org/10.1016/S1352-2310(03)00221-8 Gowers, A.M., Miller, B.G., Stedman, J.R., 2014. Estimating Local Mortality Burdens Associated With Particulate Air Pollution © Crown copyright 2014, licenced under the Open Government Licence (OGL). Hammer, O., Harper, D., Ryan, P., 2001. PAST: Paleontological Statistics Software 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 (PM2.5) in the UK atmosphere. Atmos. Environ. 46, 115–124. https://doi.org/10.1016/J.ATMOSENV.2011.10.028 Macintyre, H.L., Heaviside, C., Neal, L.S., Agnew, P., Thornes, J., Vardoulakis, S., 2016. Mortality and emergency hospitalizations associated with atmospheric particulate matter episodes across the UK in spring 2014. Environ. Int. 97, 108–116. https://doi.org/https://doi.org/10.1016/j.envint.2016.07.018 Munir, S., 2016. Analysing temporal trends in the ratios of PM2.5/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,

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