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Short communication

Isolating the effects of climate change in the variation of secondary inorganic aerosols (SIA) in Europe for the 21st century (1991–2100)

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ABSTRACT

The analysis of the influence of future climatic variations on air quality needs of methods that give a space-time display of large atmospheric data related to air pollution. Here a new approach in order to assess the impacts of climate change on the patterns of variation of secondary inorganic aerosols (SIA) over Europe is presented. The most widely used method of analysis (selected time-slices, future-minus-present method) is very sensitive to the chosen control and future periods because of the internal variability of the climate system. In order to overcome this limitation, full transient simulations for the period 1991–2100 under the SRES A2 scenario are analysed by the Empirical Orthogonal Functions (EOFs) methodology in order minimise the uncertainty associated to the internal variability due to the longer time series obtained. The results indicate that the EOF1 accounts for around 30–45% of the total variance for the SIA levels and points out a general increase of its trend over the entire domain ($p < 0.005$), except in the case of nitrate, whose change signal is not significant ($p > 0.1$). The correlation between SIA and meteorological parameters indicates that the trends and patterns of variation of aerosols are related to the higher temperature projected for the future climate. It favours the formation of sulphates and ammonium (increasing the concentrations of atmospheric oxidants) and the decomposition of ammonium nitrate, remaining in the gas phase. Further, the decreases in precipitation have a strong effect on the frequency of the washout and therefore in the levels of aerosols. The concentrations of aerosols decrease with increasing precipitation as wet deposition provides the main aerosol sink. The trend from a decreasing mixing height found in several areas of Europe is frequently related to a decrease in precipitation, representing an adding effect for the enhanced future SIA concentrations.

1. Introduction

Climate change impacts on air quality may affect long-term air quality planning. However, the policies aimed at improving air quality have not accounted for the variations in the climate (Racherla and Adams, 2006; Giorgi and Meleux, 2007). Furthermore, the coarse horizontal resolution of current global climate-chemistry simulations does not permit an estimate of the effects of climate change on tropospheric pollutants distributions on the regional scale (Forkel and Knoche, 2006). Climate change alone will influence future aerosol concentrations through modifications of gas-phase chemistry, transport, removal, and natural emissions (Avise et al., 2009; Jacob and Winner, 2009; Dawson et al., 2009). Of particular importance are the secondary inorganic aerosols (SIA, including sulphate–nitrate–ammonium), which are significant contributors to particulate matter levels.

Up to date, the most frequent approach adopted for the evaluation of climate change impacts on projected air quality has been the future-minus-present method. This is based on the assumption that biases in simulated present-day and future climate simulations should tend to cancel each other, and thus their difference captures the signal of the concentration anomalies. Although widely supported in most future climate–air quality interactions studies (e.g. Forkel and Knoche, 2006; Liao et al., 2009; Chen et al., 2009; Pye et al., 2009), this approach is very sensitive to the chosen control and future periods due to the inherent internal variability of the climate models, especially at regional scales (Raisanen et al., 2004).

The analysis of the climate change impacts on air quality needs of methods that give a space-time display of large atmospheric data coming from the full transient simulations. Empirical orthogonal

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functions (EOFs) were first used in meteorology in the late 1940s and are common tools in climate change analysis (Zorita et al., 2005; Monahan et al., 2009), but to the authors' knowledge they have not been used for the long-term analysis of air quality fields influenced by climate impacts. The EOFs methodology applied to full transient simulations minimizes the uncertainty associated to the internal variability due to the longer time series obtained, although they are computationally very expensive.

Hence, the goal of this study is to determine the spatio-temporal patterns of variability for SIA under a changing climate (SRES A2 Scenario) for Europe in the period 1991–2100 under the aforementioned full transient simulation approach, focusing mainly in southern Europe.

2. Methods

2.1. Models and domains

The regional modelling system consists on a climatic version of the Fifth-Generation PennsylvaniaState University – National Center for Atmospheric Research Mesoscale Model (MM5) (Dudhia, 1993), which has been extensively used in a number of regional climate simulations (e.g. Tagaris et al., 2007; Lynn et al., 2010; Gomez-Navarro et al., 2010), coupled to CHIMERE chemistry transport model (Bessagnet et al., 2004). The regional domain of study covers most of Europe with a resolution of 80 km (Fig. 1). 24 sigma levels are considered in the vertical, with the top at 100 hPa.

The transient climate and air quality simulations were performed from 1991 to 2100, driven by ECHO-G General Circulation Model under the SRES A2 scenario. A more detailed description of the experiment can be found in Zorita et al. (2005), Gomez-Navarro et al. (2010) and Jerez et al. (2010). The physico-chemical options for the regional modelling system (Table 1) have been chosen in order to minimise the computational cost, since none of the configurations included within the MM5-RCM provides the best performance (Fernandez et al., 2007).

In order to isolate the possible effects of climate change on the ground concentrations of air pollution, unchanged anthropogenic emissions are assumed, derived from EMEP database-officially reported emissions for the year 2005. Changes in natural biogenic emissions were generated dynamically using MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006) but fire-derived emissions are kept constant in the simulations. Fixed chemical boundary conditions for the European domain were implemented in CHIMERE and therefore a blending area of five grid points is excluded from the analysis hereafter.

2.2. Empirical orthogonal functions (EOFs)

In order to investigate the concentration signal along the 21st century, we apply a principal component analysis that decomposes the SIA and meteorological fields into Empirical Orthogonal Functions (EOFs) characterizing space-time patterns of variability. This methodology allows increasing the signal to noise ratio, reducing the high dimensionality of complex phenomena and summarizing its main properties in a much smaller number of prominent modes of variability (Hannachi et al., 2007). The EOFs analysis is conducted on the anomalies of geophysical fields. Here, mean annual series of modelled SIA and meteorological values (corresponding to the first layer of the model, aprox. 15 m, for the period 1991–2100) are used for conducting the analysis; the SIA concentrations have been standarised in order to avoid the representation of a larger variability in most emitting areas. Following Fiore et al. (2003) we construct a covariance matrix of elements r_{ij} representing the correlation of anomalies between grid square i and grid square j over the 21st century. The EOFs are the eigenvectors of the

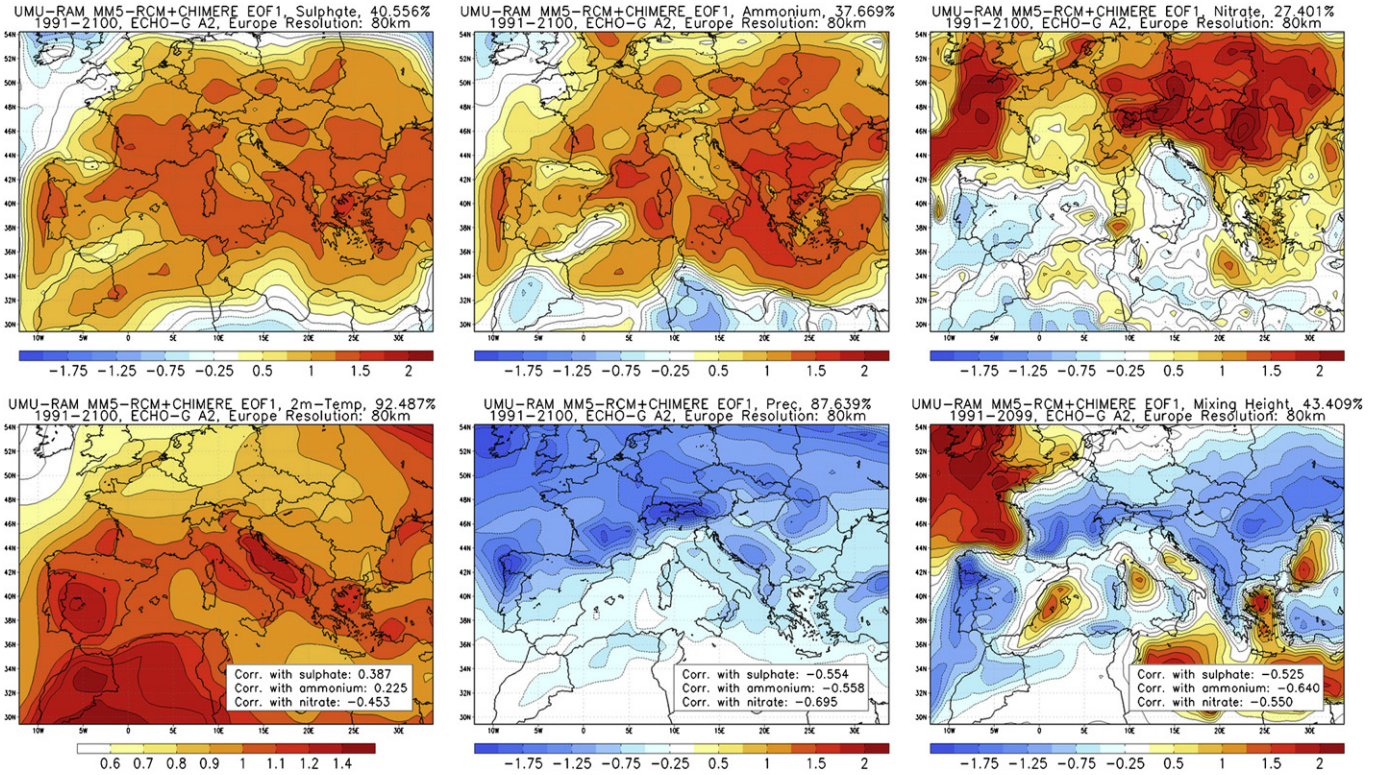

Fig. 1. EOF1 obtained from SIA sulphate, ammonium, nitrate series (up) and from meteorological parameters 2-m temperature, precipitation, mixing height (bottom). The percent of variance associated is depicted in the header of each figure.

Table 1
Configuration and parameterizations of the regional modelling system used.

Model	MM5	CHIMERE
Parameterizations	Microphysics: Simple Ice Cumulus: Grell PBL: MRF Radiation: RRTM Soil: Noah LSM	Chemical Mechanism: MELCHIOR2 Aerosol chemistry: Inorganic (thermodynamic equilibrium with ISORROPIA) and organic (MEGAN SOA) scheme) aerosol chemistry Natural aerosols: dust, re-suspension and inert sea-salt BC: LMDz-INCA + GOCART

correlation matrix and represent linear independent modes of variability. The sum of all eigenvalues equals the total variance in the original data.

The first EOF (EOF1) is defined as that accounting for the maximum amount of variance in the data set, and much of the variance can be represented only with the EOF1, making this methodology a useful data reduction technique applied to air quality data and modelling results. Projection of the SIA concentration or meteorological fields onto the EOFs defined the principal components (PCs); these time series describe the temporal variation in the contribution of the associated EOF to the total variance in the original data.

We analyse the correlations of EOF1s between the aerosol component and the meteorological parameters, which is an index of the similarity between the spatial patterns of variation (Fig. 1), and also the correlation of PC1s, depicting the temporal evolution associated to the spatial pattern of change (Fig. 2). We also applied a Mann–Kendall test to the PC1 of these variables in order to obtain the statistical significance of the obtained trends. This non-parametric test is widely used in climate studies and is capable to detect the trends or inhomogeneities in the series of data.

3. Results

The EOFs and PCs are extracted from our modelling results for the 1991–2100 period both for SIA concentrations and for those meteorological variables whose change would have a significant impact on aerosols levels such as temperature, precipitation and mixing height (Wu et al., 2008). The EOF1 accounts for around 30–45% of the total variance for the sulphate–nitrate–ammonium levels and mixing height and this variance is even larger for temperature and precipitation (92.5% and 87.6%, respectively) (Fig. 1). Therefore we will focus on EOF1 and the associated PC1, since the EOF1 captures the global trend and EOFs beyond the first individually account for less than 5% of the total variance. Moreover,

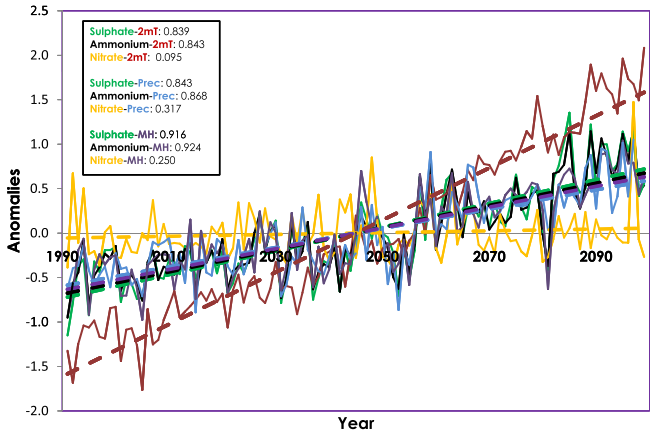

Fig. 2. PC1 trends and correlation obtained between sulphate, ammonium and nitrate and the meteorological variables included in this analysis (2-m temperature, precipitation and mixing height).

the PCs beyond the first (not shown) present no trend and a much smaller variance.

3.1. Temperature

The EOF1 for temperature can be associated to the spatial structure of the future warming pattern (Zorita et al., 2005; Gomez-Navarro et al., 2010), more intensified towards southern Europe. This spatial pattern does not seem to be importantly correlated to sulphate (0.387) and ammonium (0.225) indicating a larger impact in the spatial signal of change of other meteorological variables, as will be shown below. The warming patterns shows a slight anti-correlation to nitrate EOF1 (−0.453). This anti-correlation may be influenced by the decomposition of ammonium nitrate with higher temperatures, remaining in the gas phase (nitrate aerosol may slightly increase with cool temperature, <290 K, but decrease with hot temperature, >290 K, as temperature increases).

However, this analysis differs for the PC1, which indicates the temporal variation of SIA. The sulphate and ammonium PC1s are positively correlated to that of 2-m temperature (correlation of 0.839 and 0.843, respectively) showing a statistically significant increasing trend ($p < 0.005$). The high correlation found could be at first attributed to the trend observed for the temporal series; however, when detrended, this correlation is still above 0.6. The temporal variation of sulphate and ammonium seems to be somehow impacted by the higher temperatures modelled for the future climate; however, as shown above, the EOF1s are not spatially related. This could be attributed to the fact that temperature does not directly impact the concentrations of sulphate and nitrate, but modifying the kinetics and the concentrations of atmospheric oxidants. As reported by other authors (e.g. Dawson et al., 2007; Kleeman, 2007), temperature enhances the formation of sulphates due to a to faster SO_2 oxidation (higher rate constants and higher oxidant concentrations, increasing the formation of condensable compounds). Unger et al. (2006) point out that higher water vapor in the future climate leads to higher concentrations of H_2O_2, the principal SO_2 oxidant, thus increasing sulfate concentrations.

The slight positive trend found for nitrate PC1 is not significant ($p > 0.1$) and does not show any temporal correlation with temperature PC1 (0.095), since the effects of temperature change on nitrate are more complicated than on sulphate and ammonium due to the high vapour pressure for particle-phase ammonium nitrate. For the European domain the change signal of nitrate is very noisy, as also found by Aw and Kleeman (2003).

3.2. Precipitation

The changes in precipitation have a strong effect on the frequency of the washout and therefore in the levels of aerosols. The EOF1 for precipitation is mainly negative for all the domain and rather homogeneous, related to a general pattern of future decrease for all Europe. This decrease is strongly spatially anti-correlated to SIA (−0.554 for sulphate, −0.558 for ammonium and −0.695 for nitrate), indicating a resemblance between the future precipitation

patterns and the patterns of change of SIA concentrations. Jacob and Winner (2009) indicate that the critical variable is precipitation frequency rather than precipitation rate, since scavenging within a precipitating column is highly efficient. Temporally, the important correlation of the PC1 for sulphate and ammonium with precipitation (0.843 and 0.868) indicates that future concentrations of aerosols will increase with decreasing precipitation ($p < 0.005$) (EOF1 times PC1 is negative for all the domain indicating a decreasing signal) as wet deposition provides the main aerosol sink (Racherla and Adams, 2006; Dawson et al., 2009). Nitrate represents a singular case among the SIA components, since the PC1s correlation for precipitation-nitrate is just 0.317 and therefore no significant correlation can be attributed.

3.3. Mixing height

The changes in ventilation, and specifically, in the mixing height have strong effects on aerosols because of their low background concentrations. Strong correlation of particulate matter with stagnant conditions is expected and has been previously reported by Leung and Gustafson (2005).

The trend from a decreasing mixing height found for mostly all continental Europe is highly spatially anti-correlated to the concentrations of all SIA components (correlation of −0.525, −0.640 and −0.550 between the mixing height and sulphate, ammonium and nitrate EOF1s, respectively) leading to higher future concentrations of these pollutants, as also observed from PC1 analysis (correlation of 0.916 and 0.924 between the temporal signal of mixing height and sulphate and nitrate; no important correlation is again observed for nitrate, 0.250).

In contrast, there is a particularly strong trend to an increase in the mixing height that can be found over some parts of the Mediterranean coast and may be associated with drying and warming. In these coastal regions, the EOF1 for SIA are the lowest in all the domain (for instance, aprox. zero for ammonium) and therefore any increase in SIA concentrations due to increased temperatures or decrease in precipitation is somewhat offset by increases in the mixing heights (Avise et al., 2009), and therefore the correlation between aerosols and meteorological parameters is not obvious in these areas. However, in those other regions where reductions in mixing height occur simultaneously with increases in temperature, changes in both meteorological parameters generally lead to a positive relation in the change signal.

4. Conclusions

The EOFs analysis allows filtering the change signal of the concentrations of SIA and to identify the patterns of variability of their future concentration. The results indicate that climatic change will impact air quality by increasing the mean concentrations of these aerosol components matter in most regions over Europe.

It is established that for sulphate and ammonium the statistically significant increasing signal is driven by an enhanced secondary production as a consequence of the temperature increase, the changes in precipitation patterns, the decrease of the mixing heights hampering the dilution of pollutants and the stagnant conditions found for Europe, which are substantially modified in the future SRES A2 scenario. However, nitrate does not show a significant trend for change and no meteorological causes can be attributed to the slight increase modelled for the future concentrations of nitrate.

It should be noted that the results presented here correspond to just one model and scenario. Therefore, quantitative analysis of the implications of climatic variation for future air quality will require of further studies, which should be devoted to analyse the patterns of variability under different SRES scenarios, global-driving models o regional modelling systems (in order to increase the significance of simulation results and to characterise the uncertainty) and to quantify the impact of the physico-chemical parameterizations of the regional model on the patterns and magnitude of the change signal.

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References

Avise, J., Chen, J., Lamb, B., Wiedinmyer, C., Guenther, A., Salathe, E., Mass, C., 2009. Attribution of projected changes in summertime US ozone and PM2.5 concentrations to global changes. Atmos. Chem. Phys. 9, 1111–1124.

Aw, J., Kleeman, M.J., 2003. Evaluating the first-order effect of intraannual temperature variability on urban air pollution. J. Geophys. Res. 108 (D12), 4365. doi:10.1029/2002JD002688.

Bessagnet, B., Hodzic, A., Vautard, R., Beekmann, M., Cheinet, S., Honore, C., Liousse, C., Rouil, L., 2004. Aerosol modeling with CHIMERE – preliminary evaluation at the continental scale. Atmos. Environ. 38, 2803–2817.

Chen, J., Avise, J., Lamb, B., Salathe, E., Mass, C., Guenther, A., Wiedinmyer, C., Lamarque, J.-F., O'Neill, S., McKenzie, D., Larkin, N., 2009. The effects of global changes upon regional ozone pollution in the United States. Atmos. Chem. Phys. 9, 1125–1141.

Dawson, J.P., Adams, P.J., Pandis, S.N., 2007. Sensitivity of PM2.5 to climate in the eastern U.S: a modeling case study. Atmos. Chem. Phys. 7, 4294–4309.

Dawson, J.P., Racherla, P.N., Lynn, B.H., Adams, P.J., Pandis, S.N., 2009. Impacts of climate change on regional and urban air quality in the eastern United States: role of meteorology. J. Geophys. Res. 114, D05308. doi:10.1029/2008JD009849.

Dudhia, J., 1993. A nonhydrostatic version of the Penn State NCAR mesoscale model: validation tests and simulation of an Atlantic cyclone and cold front. Month. Weather Rev. 121, 1493–1513.

Fernandez, J., Montavez, J.P., Saenz, J., Gonzalez-Rouco, J.F., Zorita, E., 2007. Sensitivity of the MM5 mesoscale model to physical parameterizations for regional climate studies: annual cycle. J. Geophys. Res. 112 (D4), D04101.

Fiore, A.M., Jacob, D.J., Mathur, R., Martin, R.V., 2003. Application of empirical orthogonal functions to evaluate ozone simulations with regional and global models. J. Geophys. Res. 108 (D14), 4431. doi:10.1029/2002JD003151.

Forkel, R., Knoche, R., 2006. Regional climate change and its impact on photooxidant concentrations in southern Germany: simulations with a coupled regional chemistry-climate model. J. Geophys. Res. 111, D12302.

Giorgi, F., Meleux, F., 2007. Modeling the regional effects of climate change on air quality. Compt. Rendus Geosci. 339, 721–733.

Gomez-Navarro, J.J., Montavez, J.P., Jimenez-Guerrero, P., Jerez, S., Garcia-Valero, J.A., Gonzalez-Rouco, J.F., 2010. Warming patterns in regional climate projections over the Iberian Peninsula. Met. Zeit. 19 (3), 275–285.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P.I., Geron, C., 2006. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). Atmos. Chem. Phys. 6 31813210.

Hannachi, A., Joliffe, I., Stephenson, D., 2007. Empirical orthogonal functions and related techniques in atmospheric science: a review. Int. J. Climatol. 27, 1119–1152.

Jacob, D.J., Winner, D.A., 2009. Effect of climate change on air quality. Atmos. Environ. 43, 51–63.

Jerez, S., Montavez, J.P., Gomez-Navarro, J.J., Jimenez-Guerrero, P., Jimenez, J., González-Rouco, J.F., 2010. Temperature sensitivity to the land-surface model in MM5 climate simulations over the Iberian Peninsula. Met. Zeit. 19 (4), 363–374. doi:10.1127/0941-2948/2010/0473.

Kleeman, M.J., 2007. A preliminary assessment of the sensitivity of air quality in California to global change. Clim. Change 87, S273–S292.

Leung, L.R., Gustafson, W.I., 2005. Potential regional climate change and implications to U.S. air quality. Geophys. Res. Lett. 32, L16711. doi:10.1029/2005GL022911.

Liao, K.-J., Tagaris, E., Manomaiphiboom, K., Wang, C., Woo, J.-H., Amar, P., He, S., Russell, A.G., 2009. Quantification of the impact of climate uncertainty on regional air quality. Atmos. Chem. Phys. 9, 865–878.

Lynn, B.H., Rosenzweig, C., Goldberg, R., Rind, D., Hogrefe, C., Druyan, L., Healy, R., Dudhia, J., Rosenthal, J., Kinney, P., 2010. Testing GISS-MM5 physics configurations for use in regional impacts studies. Clim. Change 99, 567–587. doi:10.1007/s10584-009-9729-5.

Monahan, A.H., Fyfe, J.C., Ambaum, M.H.P., Stephenson, D.B., North, G.R., 2009. Empirical orthogonal functions: the medium is the message. J. Clim. 22, 6501–6514.

Pye, H.O.T., Liao, H., Wu, S., Mickley, L.J., Jacob, D.J., Henze, D.K., Seinfeld, J.H., 2009. Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. J. Geophys. Res. 114, D01205.

Racherla, P.N., Adams, P.J., 2006. Sensitivity of global tropospheric ozone and fine particulate matter concentrations to climate change. J. Geophys. Res. 111, D24103.

Raisanen, J., Hansson, U., Ullerstig, A., Doscher, R., Graham, L.P., Jones, C., Meier, H.E.M., Samuelsson, P., Willen, U., 2004. European climate in the late twenty-first century: regional simulations with two driving global models and two forcing scenarios. Clim. Dynam. 22, 13–31.

Tagaris, E., Manomaiphiboon, K., Liao, K.-J., Leung, L.R., Woo, J.-H., He, S., Amar, P., Russell, A.G., 2007. Impacts of global climate change and emissions on regional ozone and fine particulate matter concentrations over the United States. J. Geophys. Res. 112, D14312. doi:10.1029/2006JD008262.

Unger, N., Shindell, D.T., Koch, D.M., Amann, M., Cofala, J., Streets, D.G., 2006. Influences of man-made emissions and climate changes on tropospheric ozone, methane, and sulfate at 2030 from a broad range of possible futures. J. Geophys. Res. 111, D12313.

Wu, S., Mickley, L.J., Jacob, D.J., Rind, D., Streets, D.G., 2008. Effects of 2000–2050 global change on ozone and air quality in the United States. J. Geophys. Res. 113, D06302.

Zorita, E., Gonzalez-Rouco, J.F., von Storch, H., Montavez, J.P., Valero, F., 2005. Natural and anthropogenic modes of surface temperature variations in the last thousand years. Geophys. Res. Lett. 32, 755–762.