INFLUENCE OF CLIMATE CHANGE SCENARIOS ON HEALTH SAFETY LIMITS FOR THE PRESENCE OF ATMOSPHERIC BENZO[a]PYRENE IN EUROPE

Nuno RATOLA^{1,2}, Juan P. MONTÁVEZ¹, Pedro JIMÉNEZ-GUERRERO¹

¹Department of Physics, Regional Campus of International Excellence "Campus Mare Nostrum", University of Murcia, Murcia, Spain ^{.2}LEPABE, Universidade do Porto, Portugal nrneto@um.es, montavez@um.es, pedro.jimenezguerrero@um.es

RESUMEN

El impacto del cambio climático en la calidad del aire a largo plazo es un factor que no se está considerando actualmente en los planes de mejora de la calidad del aire establecidos por las diferentes administraciones regionales, nacionales y europeas. Por tanto, es necesario actualizar la información disponible sobre contaminantes orgánicos persistentes (COP) con el fin de incluir los impactos climáticos regionales sobre la calidad del aire. El uso de nuevos modelos climáticos/transporte químico con alta resolución nos permite obtener resultados en periodos climáticamente significativos. Con este objetivo, se ha caracterizado, para el escenario SRES A2, cómo el cambio climático afecta a las concentraciones de fondo de benzo(a)pireno (BaP), un producto químico cancerígeno para los que existen ciertas directrices sobre sus niveles. Pero, sobre todo, se desea estudiar cómo (y dónde) las zonas que superan umbrales peligrosos para la salud sufren de la influencia del cambio climático sobre el continente europeo. Se han utilizado simulaciones del modelo MM5-RCM forzadas por ECHAM5 SRES A2, por un período de tiempo que abarca desde el año 1991 hasta 2050. Los resultados indican un aumento en el riesgo general de cáncer de pulmón en Europa (1 x 10⁻⁶ para el escenario actual), que se convirtió en un orden de magnitud mayor, como consecuencia del aumento de las concentraciones medias de BaP.

Palabras clave: modelización regional, cambio climático, contaminación atmosférica, contaminantes orgánicos persistentes, modelización climática

ABSTRACT

The impact of climate change on air quality in the long term is a factor that is not currently being considered in plans to improve air quality in the different regional, national and European administrations. It is therefore necessary to update the information currently available on persistent organic pollutants (POPs) to include regional climatic impacts on air quality, using new regional climate/chemistry transport models with high resolution that allow the establishment of the climate impacts on persistent pollutants, considering the results for significant periods. To this aim, we have characterised, for SRES A2 scenario, how climate change affects background concentrations of benzo(a)pyrene (BaP), a carcinogenic chemical for which some guidelines and legislation already exist. But above all, how (and where) would these current health safety concentration thresholds suffer from the influence of different scenarios outlined for the European continent. MM5-simulations driven by ECHAM5 SRES A2 forcing were used for a time period covering the years 1991-2050. The results indicate a general increased risk of lung cancer in Europe (1 x 10^{-6} for present

scenario), which is turned into an order of magnitude higher as a consequence of the increase in average BaP concentrations over all Europe.

Key words: regional modelling, climate change, air pollution, persistent organic pollutants, climate modelling

1. INTRODUCTION

The impact of climate change on air pollution, especially on aerosols, has been previously described in a number of studies (e.g. Katragkou *et al.*, 2011; Jiménez-Guerrero *et al.*, 2012; Jerez *et al.*, 2013, among many others). Variations in climate affect pollutants by changing the dispersion (wind speed, mixing layer height, convective fronts), deposition by precipitation, dry deposition, photochemistry, natural emissions and background concentrations (Jacob and Winner, 2009). Hence, the effects of climate change on air quality should be studied in the broader context of chemistry/climate interactions (Giorgi and Meleux, 2007).

The magnitude and extent of potential impacts of climate change on air quality in Europe urged major government agencies to undertake several important actions in this regard (Amann *et al.*, 2004). The impact studies to be performed under this umbrella inevitably require downscaled scenarios of air quality under climate change frameworks, with as high spatial resolution as possible. To achieve this, the most common methodology is the online or offline coupling of chemistry transport models (CTMs) to regional climate models (RCMs). However, recent studies applying both approaches (Juda-Rezler *et al.*, 2012; Manders *et al.*, 2012, among many others) highlight a wide spread in the results related to the influence of climate change on regional air pollution at regional scales over Europe.

Incidentally, polycyclic aromatic hydrocarbons (PAHs) can be found in the atmosphere associated with the gaseous or the particulate phases (Lammel *et al.*, 2009), but also in other environmental matrices such as soil, water, sediment or vegetation (Ratola *et al.*, 2010a; 2010b). They have natural and anthropogenic sources derived from processes involving the use of fossil fuels and mainly their combustion (Srogi, 2007). These chemicals raise concern among the scientific community and have been widely studied due to their carcinogenic and mutagenic character. Having high lipid solubility, they are absorbed in the lung tissue, skin, breast or intestines, posing risks to the human health (Kim *et al.*, 2013). Benzo(a)pyrene (BaP), with molecular structure of 5 aromatic rings (and thus present in the atmosphere mainly as particulate matter) is possibly the most studied of this family of compounds, and the European Commission (Directive 2004/107/EC, amended by Regulation 219/2009) defined an average limit of 1 ng m⁻³ over one year as a reference for PAH air quality standards. Although legal limits for atmospheric PAHs are scarce, some guidelines have been presented by studies in literature that can help to assess their harmful potential (Butterfield and Brown, 2012).

As such, the objective of this work is to evaluate the impact of climate change on future levels of benzo[a]pyrene over Europe for the first half of the XXI century (simulations cover 1991-2050) driven by A2 SRES scenario.

2. METHODOLOGY

The regional modelling system consists of a climatic version of the Fifth-Generation Pennsylvania State University - National Center for Atmospheric Research Mesoscale Model (MM5) (Dudhia, 1993; Grell et al., 1994), coupled to CHIMERE chemistry transport model (EC4MACS, 2012). The spatial model configuration consists of one domain covering most of Europe of MM5-RCM simulations with spatial resolutions of 25 km, respectively. Twenty four sigma levels are considered in the vertical, with the top at 100 hPa. The fields are interpolated to CHIMERE working grids (resolutions of 0.2 degrees for the European domain). The physicochemical options for the regional modelling system have been chosen in order to minimize the computational cost, since none of the configurations included within the MM5-RCM provides the best performance for all seasons and locations (Jiménez-Guerrero et al., 2013). A blending area of five grid points is excluded from the analysis hereafter. The regional climate simulations driving the CTM were performed using the regional climate model MM5 driven by the European Centre/Hamburg 5-Run1 simulation forced by the SRES-A2 scenario. This scenario has been selected since it lies towards the upper end of the IPCC emission range, and hence maximized the effect of climate changes on air quality. No nudging of the regional model toward the global climate model was used. For further description of the regional climate simulations and the validation of the present-day simulated climatologies, the reader is referred to Jerez et al. (2010, 2013).

Simulations cover the period 1991-2050. Up to date, the most frequent approach adopted for the evaluation of climate change impacts on projected regional air quality has been the futureminus-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. This method is widely supported in most future climate-air quality interactions studies (e.g. Liao et al., 2009; Pye et al., 2009), and therefore this approach is followed here. Simulations for present-day climatologies (1991-2010) have been compared to a time slice covering 2031-2050 (denoted as 2050s in several parts of this contribution). In order to isolate the possible effects of climate change on the ground concentrations of air pollution, unchanged anthropogenic emissions (derived from the European Monitoring and Evaluation Programme -EMEP- database) are assumed. Natural emissions depend on climate conditions, and consequently the only ones to vary between reference and future climate simulations. Therefore, the effects of climate change on air pollutants are estimated without considering possible changes on vegetation, land use, anthropogenic pollutant emission changes or any feedbacks from the chemical compounds to the meteorological fields, but allowing changes in natural emissions (Jiménez-Guerrero *et al.*, 2012). Biogenic emissions were generated dynamically using MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006) with the parameterized form of the canopy environment model. The model estimates these emissions as a function of hourly temperature and ground level shortwave radiation from MM5-RCM.

3. RESULTS

3.1. Present *versus* future climatologies

Modelled BaP climatologies (Figure 1) for 1991-2010 do not show defined spatial patterns. Instead, several areas in Europe exceed 100 pg m⁻³ as annual averages (Portugal, Spain, Netherlands, Germany, Italy or Poland, among other countries), reaching more than 500 pg m⁻³

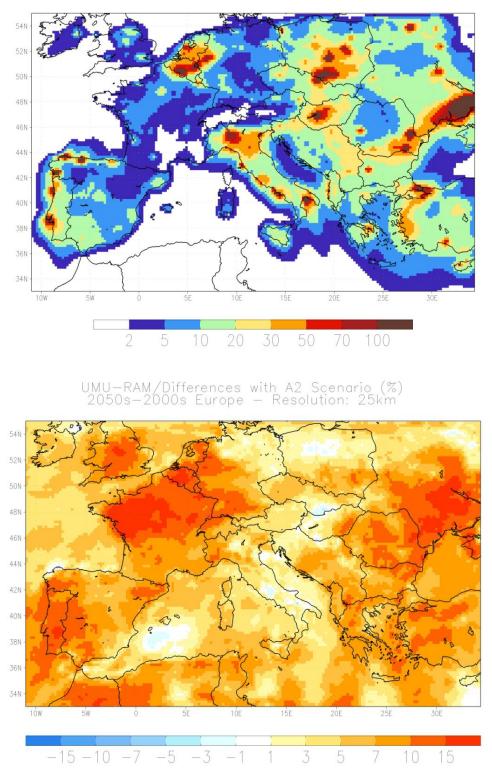
³ over some areas in Poland. The highest concentrations modelled are found in urban (e.g. Lisbon, Portugal) and industrial settings (Ruhr area, Germany), followed by rural and remote areas. This reflects the accumulation of anthropogenic sources like traffic, building heating or industrial processes involving combustions. When estimating the difference of this 1991-2010 average values with the concentrations obtained for the 2031-2050 period driven by SRES A2 scenario, the highest increases are up to 25% over western France. However, we should bear in mind that this region presents very low concentrations of BaP currently, so considerable percentage increases due to climate change alone are not reflected in high levels of this pollutant. Significant rises (around 15%) are modelled over eastern Europe (mainly Ukraine) and western Iberian Peninsula (especially over Portugal), where the enhancement of BaP levels coincide with those areas with the highest BaP concentrations over southern Europe. Since BaP is predominantly on the particulate phase, the strong decreases in precipitation modelled for the Iberian Peninsula for future climates (e.g. Jiménez-Guerrero *et al.*, 2012) reduce the wet deposition of this pollutant and imply a significant increase in its atmospheric concentration.

3.2. Assessment of increased health risks

It is also the intention of this study to provide an example of how these tools can help in the assessment (and correction) of potentially hazardous effects to human health. When the European Union set the target value in Directive 2004/107/EC, a Quantitative Risk Assessment (QRA) method was based on different studies, such as Andersen *et al.* (1982) or Lindstedt and Sollenberg (1982). These studies focused on the increased risk of lung cancer due to industrial exposure to PAHs. This QRA method is laid out in the European Union's "Ambient air pollution by Polycyclic Aromatic Hydrocarbons (PAH)" Position Paper (European Union, 2001). Using this method and the World Health Organisation estimate for PAHs unit risk of lung cancer (87×10^{-6} BaP m⁻³ for lifetime exposure), the European Union calculated the increased risk for three possible target values: (1) target value of 0.01 ng m⁻³ with an associated increased risk of 1 x 10⁻⁶; (2) target value of 0.1 ng m⁻³ (increased life-time risk of 1 x 10⁻⁵); and (3) target value of 1 ng m⁻³ with an associated increased risk of 1 x 10⁻⁴. Based on the health evidence and acceptance that the upper limit of the additional life-time risk should be less than 1 x 10⁻⁴, the target value reached for the annual mean concentration of BaP was 1 ng m⁻³ (Butterfield and Brown, 2012).

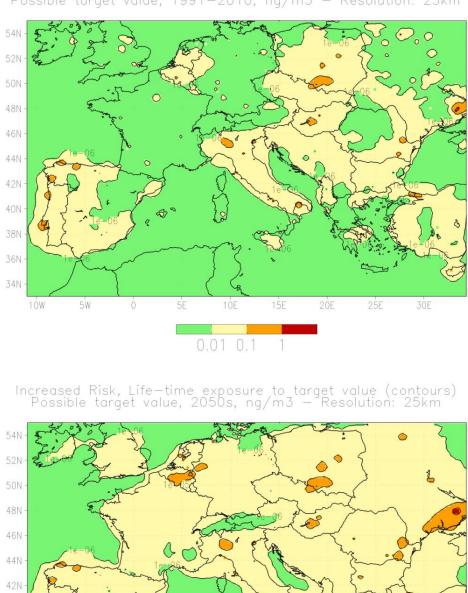
As seen in Figure 2, this target value for present-day climatologies is only exceeded in Ukraine. While some areas surpass the target value of 0.1 ng m⁻³ with associated increased risk of 1 x 10^{-5} for lung cancer, life-time exposure (mainly in the largest Portuguese cities, Lisbon and Porto, some areas in northern Spain and Poland and the Po Valley in Italy), the rest of Europe falls under it (below 0.1 ng m⁻³ in southern and eastern Europe and 0.01 ng m⁻³ in most of Europe). This involves increased risks of lung cancer under 1 x 10^{-5} and 1 x 10^{-6} , respectively. However, for SRES A2-driven simulations, the risks of lung cancer increase substantially all over Europe. While the 1 ng m⁻³ target value is only exceeded in Ukraine, a much larger number of spots exceeding the 0.1 ng m⁻³ threshold with respect to present-day climatologies (e.g. over the Ruhr Valley in Germany or the Netherlands). The general increased risk of lung cancer over land in Europe (1 x 10^{-6} for the present scenario) is turned into an order of magnitude higher (1 x 10^{-5}).

INFLUENCE OF CLIMATE CHANGE SCENARIOS ON HEALTH SAFETY LIMITS FOR THE PRESENCE OF ATMOSPHERIC BENZO[a]PYRENE IN EUROPE



UMU-RAM/Mean Concentration BaP (pg/m3) 1991-2010 - Resolution: 25km

Fig. 1: (Top) Present day mean concentrations of BaP over Europe (1991-2010) (pg m⁻³) and (bottom) percentual increase in the levels of this compound for the 2031-2050 period under the SRES A2 scenario compared to present-day levels.



Increased Risk, Life-time exposure to target value (contours) Possible target value, 1991-2010, ng/m3 - Resolution: 25km

Fig. 2: Exceedances of the possible target values (shaded, ng m⁻³) and associated increased risk of lung cancer, life-time exposure to the target value (contours), as defined by the Quantitative Risk Assessment included in Butterfield and Brown (2012). (Top) Present-day conditions; (bottom) SRES A2 scenario.

1

10E

0.01 0.1

40N 38N

34N

10W

5W

4. CONCLUSIONS

A regional climate modelling system (coupling MM5-RCM and CHIMERE chemistry transport model) has been applied to a simulation covering a present-climate condition (1991-2010) and the future SRES A2 scenario (2031-2050) in order to obtain regional distributions PAHs concentrations. Results show the plausible influence of climate change alone on the levels of BaP.

Regarding BaP, its response to changes in future climate conditions shows a wide range of variations. In general, precipitation drives the modification in the concentration of most particulate components, since the general decrease of precipitation modelled for the A2 scenario over the target domain, together with the enhanced oxidative capacity of the atmosphere, leads to a regional increase in the levels of BaP (over 15% in areas of France, southern and eastern Europe). In addition, the increased stagnation under future climates (decrease in the mixing height by 15% percent) favours the increase of BaP in polluted regions.

An important remark has to be made regarding the inherent uncertainties in the climate projections which may affect the results depicted here. Not all climatic variables are affected by the same degree of uncertainty. Future climate simulations tend to agree in a warm trend due to the increase of greenhouse gases concentrations, and only differ in the intensity and spatial distribution details of the warming. However, there is not such a good agreement in the projections of precipitation changes, which strongly affect the results for particles, such as BaP (more than 90% of this compound is in the particle phase). This larger uncertainty is partly due to the complex mechanism that governs precipitation, which involves a wide variety of spatial scales and it is approximated by different approaches among the state-of-the-art models. It is therefore important to characterise and reduce the uncertainties of model projections by having a better idea of the regional air pollution meteorology (especially related to temperature and precipitation, which influences considerably the difference between aerosol simulations).

5. REFERENCES

Amann, M.; Cofala, J.; Klimont, Z.; Hoglund, L.; Wagner, F. and Winiwarter, W. (2004). *Synergies between GHG mitigation and air pollution control.* 2004 IIASA Contribution to Conference of Parties (COP10), Buenos Aires, Argentina; 8 Dec.

Andersen, A.; Dahlberg, B.E.; Magnus, K. and Wannag, A. (1982). "Risk of cancer in the Norwegian aluminium industry". *International Journal of Cancer*, 29, pp. 295-298.

Butterfield, D.M. and Brown, R.J.C. (2012). *Polycyclic aromatic hydrocarbons in Northern Ireland*. National Physical Laboratory Report AS66, Teddington, Middlesex, UK.

Dudhia, J. (1993). "A nonhydrostatic version of the Penn State NCAR mesoscale model: Validation tests and simulation of an Atlantic cyclone and cold front". *Monthly Weather Reviews*, 121, pp. 1493-1513.

EC4MACS - European Consortium for Modelling of Air Pollution and Climate Strategies, 2012. *The CHIMERE Atmospheric Model*. Eds. B. Bessagnet, E. Terrenoire, F. Tognet, L. Rouil, A. Colette, L. Letinois, L. Malherbe. Issued by INERIS, March 2012.

Giorgi, F. and Meleux, F. (2007). "Modeling the regional effects of climate change on air quality". *Comptus Rendus Geosciences*, 339, 721-733.

Grell, A.G.; Dudhia, J. and Stauffer, D.R. (1994). A description of the fifth-generation PennState/NCAR mesoscale model (MM5). NCAR Technical Note NCAR/TN-398+STR, Natl Cent Atmos Res, Boulder, Colorado: 138. On-line: http://www.mmm.ucar.edu/mm5

Guenther, A.; Karl, T.; Harley, P.; Wiedinmyer, C.; Palmer, P.I. and Geron, C. (2006). "Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature)". *Atmospheric Chemistry and Physics*, 6, pp. 3181-3210.

Jacob, D.J. and Winner, D.A. (2009). "Effect of climate change on air quality". *Atmospheric Environment*, 43, pp. 51–63.

Jerez, S.; Montávez, J.P.; Gómez-Navarro, J.J.; Jiménez-Guerrero, P.; Jiménez, P. and González-Rouco, J.F. (2010). "Temperature sensitivity to the land-surface model in MM5 climate simulations over the Iberian Peninsula". *Meteorologische Zeitschrift*, 19, pp. 363–374.

Jerez, S.; Jiménez-Guerrero, P.; Montávez, J.P. and Trigo, R.M. (2013). "Impact of the North Atlantic Oscillation on European aerosol ground levels through local processes: a seasonal model-based assessment using fixed anthropogenic emissions". *Atmospheric Chemistry and Physics*, 13, pp. 11195-11207.

Jiménez-Guerrero, P.; Montávez, J.P.; Gómez-Navarro, J.J.; Jerez, S. and Lorente-Plazas, R. (2012). "Impacts of climate change on ground level gas-phase pollutants and aerosols in the Iberian Peninsula for the late XXI century". *Atmospheric Environment*, 55, pp. 483–495.

Jiménez-Guerrero, P.; Jerez, S.; Montávez, J.P. and Trigo, R.M. (2013). "Uncertainties in future ozone and PM10 projections over Europe from a regional climate multiphysics ensemble". *Geophysical Research Letters*, 40, pp. 5764–5769.

Juda-Rezler, K.; Reizer, M.; Huszar, P.; Kruger, B.; Zanis, P.; Syrakov, D.; Katragkou, E.; Trapp, W.; Melas, D.; Chervenkov, H.; Tegoulias, I. and Halenka, T. (2012). "Modelling the effects of climate change on air quality over Central and Eastern Europe: concept, evaluation and projections". *Climate Research*, 53, pp. 179-203.

Katragkou, E.; Zanis, P.; Kioutsioukis, I.; Tegoulias, I.; Melas, D.; Krüger, B.C. and Coppola, E. (2011). "Future climate change impacts on summer surface ozone from regional climate-air quality simulations over Europe". *Journal of Geophysical Research*, 116, pp. D22307.

Kim, K.-H.; Jahan, S.A.; Kabir, E. and Brown R.J.C. (2013). "A review of airborne polycyclic aromatic hydrocarbons (PAHs) and their human health effects". *Environment International*, 60, pp. 71–80.

Lammel G.; Sehili A.; Tami B.; Feichter J. and Grassl H. (2009). "Gas/particle partitioning and global distribution of polycyclic aromatic hydrocarbons – A modelling approach". *Chemosphere*, 76, pp. 98-106.

Liao, K.-J.; Tagaris, E.; Manomaiphiboon, K.; Wang, C.; Woo, J.-H.; Amar, P.; He, S. and Russell, A.G. (2009). "Quantification of the impact of climate uncertainty on regional air quality". *Atmospheric Chemistry and Physics*, 9, pp. 865-878.

Lindstedt G, and Sollenberg J. (1982). "Polycyclic aromatic hydrocarbons in the occupational, environment, with special reference to benzo[a]pyrene measurements in Swedish industry". *Scandinavian Journal of Work Environment and Health*, 8, pp. 1-19.

Manders, A.M.M.; van Meijgaard, E.; Mues, A.C.; Kranenburg, R.; van Ulft, L.H. and Schaap, M. (2012). "The impact of differences in large-scale circulation output from climate models on the regional modeling of ozone and PM". *Atmospheric Chemistry and Physics*, 12, pp. 9441–9458.

Pye, H.O.T.; Liao, H.; Wu, S.; Mickley, L.J.; Jacob, D.J.; Henze, D.K. and Seinfeld, J.H. (2009). "Effect of changes" in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. *Journal of Geophysical Research*, 114, pp. D01205

Ratola, N.; Amigo, J.M. and Alves, A. (2010a). "Comprehensive assessment of pine needles as bioindicators of PAHs using multivariate analysis. The importance of temporal trends". *Chemosphere*, 81, pp. 1517-1525.

Ratola, N.; Amigo, J.M. and Alves, A. (2010b). "Levels and sources of PAHs in selected sites from Portugal: biomonitoring with *Pinus pinea* and *Pinus pinaster* needles". *Archives of Environmental Contamination and Toxicology*, 58, pp. 631-647.

Srogi, K. (2007). "Monitoring of environmental exposure to polycyclic aromatic hydrocarbons: A review". *Environmental Chemistry Letters*, 5, pp. 169–195.