# MODELING THE EFFECTS OF A CLIMATE CHANGE SCENARIO ON THE DISTRIBUTION OF ORGANIC POLLUTANTS IN THE GLOBAL ATMOSPHERE

Lamon Lara<sup>1,2</sup>, von Waldow Harald<sup>3</sup>, MacLeod Matthew<sup>3</sup>, Scheringer Martin<sup>3</sup>, Marcomini Antonio<sup>1,2</sup>, Hungerbühler Konrad<sup>3</sup>

<sup>1</sup> Department of Environmental Sciences, University Ca' Foscari of Venice, Calle Larga S. Marta 2137, 30123 Venice, Italy; <sup>2</sup>Euro Mediterranean Centre for Climate Change, CMCC, Via Augusto Imperatore 16, 73100 Lecce, Italy; <sup>3</sup>Safety and Environmental Technology Group, Swiss Federal Institute of Technology, Zürich, Hönggerberg 8093, CH

#### Abstract

We have applied a global multimedia fate model to evaluate changes in the atmospheric distribution of two polychlorinated biphenyls, PCB 28 and PCB 153, under the influence of climate change. We defined a climate scenario representing the last twenty years of the 20<sup>th</sup> century ("20CE scenario") and a scenario representing the global climate under the assumption of strong future greenhouse gas emissions ("A2 scenario"). These two climate scenarios are defined by four groups of environmental factors: 1) temperature in the atmosphere, 2) wind speeds and directions, 3) oceanic current velocities and directions, and 4) precipitation rate and geographical pattern. As a fifth factor in our scenarios, we consider the effect of temperature on primary volatilization emissions of PCBs. Comparison of model results for the 20CE scenario against long-term monitoring data of concentrations. Temperature and wind differences between the two scenarios are the dominant factors determining the difference in atmospheric distribution of PCB under the A2 scenario. This study indicates that, in a future impacted by climate change, we can expect increased volatilization emissions and increased mobility of persistent organic pollutants with properties similar to the PCBs.

# Introduction

Climatologic studies show that never in the past have changes been as rapid as those we are experiencing in modern times, and there is now strong evidence that this is due to human influence<sup>1</sup>. Dated ice cores show that the atmospheric concentration of carbon dioxide ( $CO_2$ ) had been fluctuating between 180 and 300 ppm over the last 650 000 years, but a sharp increase from 280 ppm in the pre-industrial period to 379 ppm in 2005 has been documented. Numerous changes in climate have been observed at both the global and local scales; these include changes in surface temperatures and ice cover in the Arctic, widespread changes in precipitation amounts, ocean salinity, wind patterns and aspects of extreme weather including droughts, heavy precipitation, heat waves and intensity of tropical cyclones<sup>2</sup>.

Persistent Organic Pollutants (POPs) are defined in the UNECE (United Nations Economic Commission for Europe) Convention on Long-Range Transboundary Air Pollution (CLRTAP) as persistent, toxic and bioaccumulative compounds<sup>3</sup>. Because of an increasing concern about these contaminants, treaties were signed at international level: the United Nations Environmental Program (UNEP) Stockholm Convention signed by 127 countries in 2001 is an important example of the worldwide attention focused on POPs<sup>4</sup>.

The environmental behavior of chemicals like PCBs, which are archetypal POPs, depends on a complex interaction between many variables; therefore any alteration of climate conditions may affect their distribution and fate in a multitude of different ways. Providing experimental evidence of changes in POPs fate due to climate change is extremely challenging, as long-term monitoring data are needed for each environmental compartment<sup>5</sup>. In this project we approach this problem considering a manageable sub-set of possible changes in climate scenarios we used here are defined by differences in 1) temperature, 2) atmospheric circulation patterns, 3) ocean circulation patterns and 4) precipitation rate. In addition, we also considered the effect of temperature differences between the climate scenarios on the rate of primary volatilization emissions of PCB 28 and PCB 153, since this has been recognized as the dominant mechanism of release of PCBs to the atmosphere<sup>6</sup>.

# **Materials and Methods**

BETR-Global is a multimedia fate and transport model with a spatial resolution of  $15^{\circ} \times 15^{\circ}$ . The model represents advective transport between the regions in air and water, intermedia transport and partitioning processes. The model is capable of both dynamic (time varying) and steady state (time invariant) calculations. Atmospheric and oceanic data (i.e. precipitation rates, 3-dimensional temperature fields, global atmospheric and oceanic circulation fluxes) resolved on a monthly resolution for a representative year<sup>7</sup> describe environmental conditions in dynamic model runs, and yearly average conditions describe the environment in steady state runs. Each region consists of 7 environmental compartments (oceanic water, fresh water, lower and upper atmosphere, soil, sediment, vegetation). The model represents advective transport between the regions in air and water as well as intermedia transport processes like deposition and revolatilization within the regions<sup>7</sup>.

In this project, parameters describing the environment in BETR-Global were derived from the output of ECHAM5/MPI-OM, an atmosphere-ocean general circulation model (AOGCM). The datasets were obtained from the Coupled Model Intercomparison Project (CMIP3) and are representative of the forecasts of the ensemble of models that were used for the Fourth Assessment Report (AR4) of IPCC<sup>8</sup>.

In order to evaluate the influence of a climate change scenario on the environmental distribution of POPs we used two different climate scenarios. One scenario represents current climate conditions (i.e. 20CE) and the other one represents a possible future climate in the year 2100 (i.e. A2). Our 20CE scenario is based on the AOGCM output for the years 1981 to 2000 of the CMIP3-"climate of the 20<sup>th</sup> century experiment" (20C3M), and our A2 scenario is based on the output for the years 2080 to 2099 of the CMIP3 SRES A2 experiment<sup>9</sup>.

The highly resolved datasets from the AOGCM were averaged temporally and spatially to match the resolution of BETR-Global, and used as an input to our chemical fate model in dynamic and steady state model simulations.

For our model evaluation we used the dynamic version of the model, whereas we examined the effect of the climate change scenario using steady-state calculations. We considered dynamic calculations to evaluate the model against recent monitoring data since PCBs are not currently near steady-state in the global environment. We selected instead steady-state concentrations to evaluate the effect of the A2 climate scenario because they reflect the long time-scale associated with POPs, and the steady-state concentration is a surrogate for cumulative environmental exposure, regardless of the temporal pattern of emissions<sup>12</sup>.

First, we carried out steady state calculations using generic climate scenarios in order to assess the relative importance of each climate variable in influencing PCBs environmental distribution. Then, we used generic emission scenarios in order to identify the effect of climate change on the distribution of PCBs released from different source regions. Attention was focused on the concentration of PCBs in the atmosphere of the model because PCBs 28 and 153 have been subject to long term monitoring programs that characterize concentration in the atmosphere.

#### **Results and discussion**

Using emission estimates from Breivik et al.<sup>6</sup>, we compared the hindcast of present conditions with available field data for different years and locations, provided by  $EMEP^9$  for European countries and by IADN for North American areas<sup>10</sup>. The model results are in satisfactory agreement with observations: Figure 1 compares modeled and observed seasonal (three-month) average concentrations of the two PCB congeners in air at the 16 monitoring stations for the default and the maximum emission scenarios as defined by Breivik et al.<sup>6</sup>, diagonal lines represent perfect agreement and agreement within factors of  $10^{0.5}$  (= 3.16) and 10 for comparison. Figure 1 shows that atmospheric concentrations are generally under-predicted with the default emission scenario, and are slightly over-predicted when assuming the maximum emission scenario. In the maximum emission scenario, 64% of the modeled concentrations are within a factor of 3.16 of the measured concentrations, and 96% of the data points for PCB 28 and PCB 153 lie within one order of magnitude of the 1:1 line. We view the agreement between measured and modeled concentrations as satisfactory in that the model provides a description of PCB 28 and 153 concentrations in air that is in reasonable agreement with observations. Since the maximum emission scenario provides better agreement with field data, we used the maximum emission scenario in our calculations that compare the two climate scenarios.

#### First model experiment

Different hypothetical steady state climate scenarios assuming a change in only one factor, i.e., temperature, total precipitation, atmospheric and oceanic circulation, demonstrate that the most influential factors in determining the changes in concentration in lower air in the A2 scenario are changes in wind speed, wind direction and temperature. A realistic spatial distribution of emissions<sup>6</sup> was assumed, and the results were compared with the original A2 climate scenario, where all parameters change compared to the 20CE scenario.

Figure 2 shows the ratio of concentrations of PCBs under the A2 climate scenario to the concentrations under the 20CE scenario (first column). For both congeners, concentrations in air are higher everywhere in the global atmosphere under the A2 scenario. The spatial pattern of increase in concentrations is similar for PCB 28 and PCB 153 but the increase is higher for PCB 153, up to a factor of 2.5, compared to a factor of 1.8 for PCB 28. This is possibly due to its higher sensitivity of vapor pressure to changes in temperature, and higher half life in air (29-90 d compared to 14-29 d for PCB 28<sup>13</sup>).

Areas of notable increase in modeled concentrations are the Arctic and the equatorial Pacific Ocean.

The other panels of Figure 3 show results from the first model experiment, where we constructed hypothetical climate scenarios from one climate parameter taken from the A2 climate scenario and all other parameters from the 20CE scenario. Shown are the three parameters with the strongest influence on the concentration patterns in air: temperature effects on emissions (second column), temperature effects on dynamic re-partitioning and degradation (third column) and wind speeds (fourth column). This Figure shows that different factors contribute to the difference in atmospheric concentration between A2 and the 20CE scenarios for different locations. The effect of temperature on primary emissions (second column in Figure 2) results is a general increase in concentrations throughout the globe that is stronger for PCB 153 than for PCB 28. The increase is strongest over central Asia, which is caused by increased primary volatilization emissions in eastern Europe and Russia. The effect of higher temperatures under the A2 scenario on environmental partitioning and degradation of PCBs results in higher concentrations globally, especially in the Arctic and, to a lesser extent, the Antarctic (third column in Figure 2). The changes in atmospheric circulation patterns account for higher concentrations of both congeners in the equatorial Pacific, and lower concentrations in the equatorial Atlantic and over Antarctica (Figure 2, last column).

The other parameters considered in this study, i.e. oceanic currents and precipitation, lead to smaller changes in environmental behavior of PCBs in lower air.

# Second model experiment

We considered four hypothetical emission scenarios assuming respectively North America, Europe, Asia, and South America as emission regions, and we compared the steady state model results for lower air concentrations under the A2 and the 20CE scenarios. The results are shown in Figure 3, where each emission region is highlighted with a squared box. Figure 3 shows that for PCB 28 the modeled concentrations in air in the hemisphere where emissions occur increases, and there is a slight decrease in the other hemisphere, reflecting that the average half-life of PCB 28 in air is not long enough for efficient inter-hemispheric mixing. Modeled concentrations of PCB 28 are higher in the direction "downwind" (i.e., eastward) of the source regions under the A2 scenario, and are in some cases lower immediately "upwind" (i.e., westward). This trend is most evident when the European region is considered as the source region, and is attributable to the accelerated wind speed in the eastward direction in both the lower air and the upper air compartments under the A2 scenario.

For PCB 153, modeled concentrations in air increase in both hemispheres under the A2 scenario compared to the 20CE scenario. In all scenarios and for both PCBs, the modeled PCB concentrations in equatorial regions over the Pacific Ocean are higher under the A2 scenario than under the 20CE scenario.

Downwind of the source regions the model forecasts higher concentrations under the A2 scenario for both congeners. This trend implies an increased potential for inter-continental transport and transport into the Arctic, and is most obvious for emissions in Europe, but is also apparent for emissions in North America and Asia.

Our results identified temperature and atmospheric circulation pattern as climate variables affecting the PCB distribution in the atmosphere most strongly. The increase in primary volatilization emissions as a result of higher temperature in the A2 scenario is the most influential factor affecting the atmospheric concentration of PCB 28 and PCB 153. A first consideration is then that the more efficient mobilization of POPs-like substances from primary sources in a future impacted by climate change may dominate all other effects of such change on global scale POPs dynamics. And, our model experiments show also that PCBs are more efficiently transported

in the global atmosphere under the A2 scenario compared to the 20CE scenario. However the overall persistence in the multimedia environment is lower under the A2 scenario (2.7 years in the 20CE scenario versus 3.8 years in the A2 scenario for PCB 28), therefore our experiments show that PCBs are less persistent in a future impacted by global-scale warming.

# Acknowledgements

We acknowledge the modeling groups, the Program for Climate Model Diagnosis and Intercomparison (PCMDI) and the WCRP's Working Group on Coupled Modeling (WGCM) for their roles in making available the WCRP CMIP3 multi-model dataset. Support of this dataset is provided by the Office of Science, U.S. Department of Energy.

We gratefully acknowledge the Euro-Mediterranean Centre for Climate Change (CMCC; Lecce, Italy) for financial support.

# References

- Solomon, S., Qin, D., Manning, M., Alley, R.B., Berntsen, T., Bindoff, N.L., Chen, Z., Chidthaisong, A., Gregory, J.M., Hegerl, G.C., Heimann, M., Hewitson, B., Hoskins, B.J., Joos, F., Jouzel, J., Kattsov, V., Lohmann, U., Matsuno, T., Molina, M., Nicholls, N., Overpeck, J., Raga, G., Ramaswamy, V., Ren, J., Rusticucci, M., Somerville, R., Stocker, T.F., Whetton, P., Wood, R.A., Wratt, D., 2007. Technical summary. In: Climate Change 2007: the Physical Science Basis. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. <u>http://www.ipcc.ch/pdf/assessmentreport/ar4/wg1/ar4-wg1-ts.pdf</u>
- Trenberth K.E., Jones P.D., Ambenje P., Bojariu R., Easterling D., Klein Tank A., Parker D., Rahimzadeh F., Renwick J.A., Rusticucci M., Soden B. and Zhai P., 2007, Climate Change, The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, 2007, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. http://www.ipcc.ch/pdf/assessment-report/ar4/wg1/ar4-wg1-chapter3.pdf
- 3. UNECE, Protocol to the 1979 convention on long-range Transboundary air Pollution on Persistent Organic Pollutants, 1998. <u>http://www.unece.org/env/lrtap/full%20text/1998.POPs.e.pdf</u>
- 4. OECD, 2004. OECD Series on Testing and Assessment No. 45, Guidance Document on the Use of Multimedia Models for Estimating Overall Environmental Persistence and Long-range Transport.
- 5. Lamon L., Dalla Valle M., Critto A., Marcomini A., *Environmental Pollution* 2009; 157: 1971.
- 6. Breivik K., Sweetman A.M., Pacyna J. and Jones K.C. Sci. Total Environ 2007; 377: 296.
- 7. MacLeod M., Riley W.J. and McKone T.S., Environ. Sci. Technol. 2005; 39: 6749-56.
- 8. Roekner E., Brasseur G.P., Giorgetta M., Jacob D., Jungklaus J., Reick C., Sillmann J., Max Plank Intitut für Meteoreologie, 2001; pp 32.
- Nakicenovic, N.; Alcamo, J.; Davis, G.; de Vries, B.; Fenhann, j.; Gaffin, S.; Gregory, K.; Grubler, A.; Jung, T.; Kram, T.; La, E.; Michaelis, L.; Mori, S.; Morita, L.; Pepper, W.; Pitcher, H.M.; Price, L.; Riahi, K.; Roehrl, A.; Rogner, H.; Sankovski, A.; Schlesinger, M.; Shukla, P.; Smith, S.J.; Swart, R.; van Rooijen, S.; Victor, N.; Dadi, Z. Special Report on Emissions Scenarios: A Special Report of Working Group III of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, 2000. http://www.grida.no/publications/other/ipcc\_sr/?src=/climate/ipcc/emission/
- 10. EMEP, EMEP data online, http://www.emep.int/.
- 11. IADN, Integrated Atmospheric Deposition Network, <u>http://www.msc-smc.ec.gc.ca/iadn/index\_e.html</u>
- 12. von Waldow H., Scheringer M., Hungerbuehler K. Ecological Modelling 2008; 219: 256.
- 13. Mackay D., Shiu W.Y, Ma K.C., Lee S.C., Handbook of Physical-Chemical Properties and Environmental Fate for Organic Chemicals, Second Edition, 2006, CRC.

Figure 1: Comparison of seasonally averaged measured and modelled concentrations of PCB 28 (left column, ○) and PCB 153 (right column, △) assuming the default (first panel) and the maximum (second panel) emission scenarios as defined by Breivik et al.<sup>6</sup>

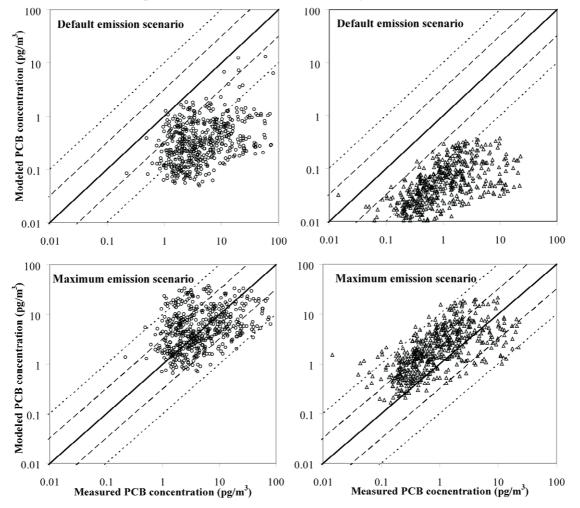


Figure 2: Ratio of modeled PCBs concentrations at steady state in the atmosphere under hypothetical climate conditions for PCB 28 (first line) and PCB 153 (second line). The hypothetical climate conditions refer to: (first column) the A2 scenario; (second column) the 20CE scenario with a change in the volatilization from primary sources to reflect the temperature in the A2 scenario; (third column) temperature fields from the A2 scenario (here volatilization is considered only from water and soil), other parameters from the 20CE scenario; (fourth column) wind fields from the A2, other parameters from the 20CE scenario.

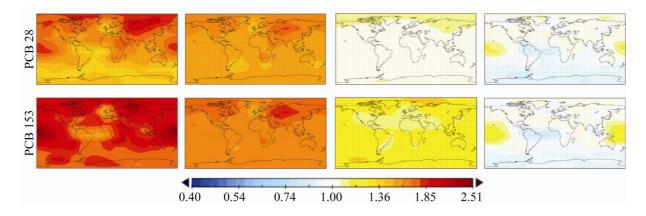


Figure 3: Ratio of modeled PCB concentrations at the steady state in the atmosphere under the A2 climate scenario to concentrations under the 20CE scenario for PCB 28 (first line) and PCB 153 (second line) for four emission regions, marked with a black box.

