ASSESMENT OF TEMPORAL TRENDS OF TRACE METALS OVER FRANCE BETWEEN 1996 AND 2011: HOW TO INCLUDE THE EFFECTS OF THE PROTOCOL AND THE GEOGRAPHICAL VARIATIONS?

Emeline Lequy, A. Pascaud, S. Sauvage, S. Leblond

Museum National d'Histoire Naturelle, Service du Patrimoine Naturel, Paris, France
Mines Douai, SAGE, Douai, France
emeline.lequy@mnhn.fr

Keywords: Cadmium; Lead; Generalized mixed models; atmospheric deposition; moss species

Introduction

Atmospheric pollution has been a worldwide challenge, especially in Europe where the Convention on Long Range Transboundary Atmospheric Pollution was agreed in November 1979. The International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops was created in 1987 and includes a moss bio-monitoring of trace metals. This network made it possible to determine spatial and temporal trends in Europe (Harmens et al., 2010, 2015). But some variability remains in the results due to the sampling protocol features (*eg*. the moss species or the sampling period), which the former studies overlooked. This study aims to assess temporal trends in France after taking into account some co-variables of the concentrations, for the trace metals (TM) Cd, Hg, Ni, and Pb, by use of mixed models.

Methods

Mosses were sampled in France during 4 surveys from 1996 to 2011, every 5 years, with the financial support of ADEME (French Agency for Environment and Energy Management). Five moss species were collected according to the IPC-Vegetation guidelines. Sampling sites were considered a repetition between surveys if they were within 500m of the sampling site of the former survey and under the same biotope, giving 280 repeated sites in our database. They are roughly homogeneously distributed over France, except the North and the French Brittany (NW). Samples were analyzed by IAAS, ICP-AES, ICP-MS in 1996, 2001, 2006-2011, respectively. Non-conformed concentrations according to standard samples were corrected. Data for Hg were unavailable for the 1996 survey. Due to the structure of the data, *ie* the repetition of the sites over time, we had to analyze the effect of the time associated with other co-variables (moss species, sampling period, region in France among a regular 6-cell raster) with mixed modeling. The sampling period was computed as the day of years of each sample, categorized by quartiles of this distribution. As fixed effects we entered the time, the regions, the moss species, the sampling period, and the interaction between time and regions into the model. As random effects we had the site, and the interaction between time and moss species. The TM concentrations in mosses were log-normally distributed, so we opted for a generalized linear mixed model (GLMM) with a log distribution. We used the R software with the lme4 package (Bates, Maechler, Bolker, & Walker, 2014) and the glmer function.

Results

Significant temporal trends over France were found for Cd and Pb (Table 1), despite significant effects of the considered features of the protocol. Regional differences appeared for Pb, Ni, while it was less clear for Cd and Hg. Significant negative interactions between time and region were found for Hg and Ni in the ME region (Rhodanian valley impacted by heavy industries) and on the Northern part of France, respectively. Interestingly, Pb had a positive interaction time:region in the SE of France. The moss species was significant for Pb but this effect was less clear for the other TM.

Table 1. Coefficients of the GLMM for the TM Cd, Hg, Ni, and Pb. Significant values (5%) are in bold.

Variable		Cd	Hg	Ni	Pb
Time		**-0.03**	-0.02	0.01	**-0.09**
Region (ref: NW)	NE	**0.52**	-0.03	**0.44**	**0.2**
	MW	0.06	-0.04	0.06	**-0.32**
	ME	**0.36**	**0.26**	**0.66**	-0.04
	SW	0.09	-0.04	0.19	**-0.29**
	SE	0.07	0.01	**0.47**	-0.16
Moss species (ref: M1)	M2	0.14	0.24	-0.29	**-0.25**
	M3	0.16	0.06	**-0.39**	**-0.29**
	M4	0.11	-0.07	-0.22	**-0.3**
	M5	0.01	0.13	-0.03	**-0.33**
Interaction time:region	NE:time	**-0.03**	0	**-0.02**	**-0.02**
	MW:time	**-0.03**	-0.01	**-0.03**	**-0.02**
	ME:time	**-0.03**	**-0.02**	**-0.04**	0
	SW:time	**-0.03**	-0.01	-0.01	0
	SE:time	**-0.02**	-0.01	-0.02	**0.01**
Sampling period* (ref: SP1)	SP2	-0.09	-0.1	-0.27	**-0.32**
	SP3	**-0.37**	**-0.17**	**-0.32**	**-0.31**
	SP4	**-0.58**	-0.13	**-0.35**	**-0.3**

* SP corresponds to the days of years of each sample, then categorized according to the quartiles of the distribution.

Conclusion

Significant decreasing trends were established for Cd and Pb, while Hg and Ni had no significant trends. The moss species had a strong effect for Pb, and the sampling period for the 4 TM. Regional trends are suspected for Cd, Ni, and Pb. The effect of the features of the protocol is thus needed to be taken into account to assess robust temporal trends.

References

Bates, D., Maechler, M., Bolker, B., & Walker, S. (2014). "lme4: Linear mixed-effects models using Eigen and S4." ArXiv e-print; submitted to _Journal of Statistical Software_. Retrieved from http://arxiv.org/abs/1406.5823

Harmens, H., Norris, D. A., Sharps, K., Mills, G., Alber, R., Aleksiayenak, Y., … Zechmeister, H. G. (2015). Heavy metal and nitrogen concentrations in mosses are declining across Europe whilst some "hotspots" remain in 2010. *Environmental Pollution*, *200*, 93–104. http://doi.org/10.1016/j.envpol.2015.01.036

Harmens, H., Norris, D. A., Steinnes, E., Kubin, E., Piispanen, J., Alber, R., … Zechmeister, H. G. (2010). Mosses as biomonitors of atmospheric heavy metal deposition: Spatial patterns and temporal trends in Europe. *Environmental Pollution*, *158*(10), 3144–3156. http://doi.org/10.1016/j.envpol.2010.06.039