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Understanding Polycyclic Aromatic Hydrocarbon transfers at the catchment scale combining chemical and fallout radionuclide analyses

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Reducing environmental contamination constitutes a major challenge for industrialized countries. Furthermore, in the European Union, Water Framework Directive (WFD; Directive 2000/60/EC) requires that the member state water bodies reach good ecological and chemical status by 2015. Polycyclic Aromatic Hydrocarbons (PAHs) are a group of persistent organic pollutants considered as priority pollutants because of their mutagenic and carcinogenic properties. They are mostly emitted by human activities such as household heating or road traffic. Although emissions have decreased during the last decades, a large amount of PAHs have been released into the atmosphere for the last two centuries. In recent years, studies dealing with PAHs have grown in number but most of them were restricted to the measurement of PAHs concentrations in the different compartments of the environment (air, soil, sediment, water, etc.). In this context, there remains a lack of knowledge about the transfers and, consequently, about the persistence of these compounds in the environment. This question is particularly acute in the Seine River basin where very high concentrations in PAHs are reported in sediment, thereby compromising the achievement of the good chemical status required by WFD.

Our study aims to quantify PAHs transfers at the catchment scale by combining chemical analysis with gamma spectrometry. Atmospheric fallout, soil, river water and sediment samples were collected in two upstream sub-catchments of the Seine River basin during one year. Chemical analyses, restricted to 15 of the 16 PAHs selected by the US Environmental Protection Agency (USEPA), were carried out to determine PAHs concentrations in all samples. Contamination spectra were used to outline the potential origin of pollution. Measurement of fallout radionuclides (Beryllium-7, Lead-210, Caesium-137) in both rainfall and river sediment provided a way to discriminate between freshly eroded sediment vs. material that deposited on the riverbed. These data are crucial to estimate PAH residence time in the Seine River basin and to know if current river pollution is resulting from recent emissions or from the persistence of former sources.

Results show that spectra of contaminants in sediment are similar in most of the sampling sites. This underlines the possibility of a common primary source. Moreover, local variations of atmospheric PAHs fallout are poorly significant within the subcatchments, even though concentrations greatly vary in sediment. This underlines the presence of local secondary sources where accumulation of atmospheric fallout is more important but their deposition ways still remain to determine. At other sites, impact of urbanized area is more likely to be the source of light HAPs in sediment. Measurements in soils show larger concentrations in forest than in grassland or in cropland. This can be explained by a bigger affinity of PAHs for the soils richer in Organic matter (OM) and lower erodibility of woodland.