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Persistent toxic substance inputs to the river Seine basin (France) via atmospheric deposition and urban sludge application

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Abstract

Paris constitutes a major direct and indirect source of persistent toxic substances (PTS) to the river Seine, its tributaries and its basin, by atmospheric depositions and sewage sludge land-filling. The contaminant cycle and transfer pathways were investigated from 1999 to 2003 at local and inter regional scales in order to determine the respective importance of the main input and diffusion processes (wastewater, rainwater and runoff) from urban to rural areas. Paris constitutes an atmospheric emission hot spot for PAHs and PCBs. For example, for 2002, atmospheric concentrations ranged from 0.5 to 3 ng m^{-3} for PAHs (∑ 6 WHO) and from 0.06 to 0.69 ng m^{-3} for PCBs (∑ 7, EEC) and concentrations in bulk deposition ranged from 6.6 to 647 ng L^{-1} for PAHs (∑ 14) and from 0.6 to 8.1 ng L^{-1} for PCBs. At Paris, annual atmospheric deposition inputs of PAHs (∑ 6) and PCBs (∑ 7) reached 104 g km^{-2} and 35 g km^{-2}, respectively. PAHs followed a marked seasonal cycle in relation with winter domestic heating and bulk deposition concentrations were 5 to 15 times lower in remote areas. No seasonal cycle was observed for PCBs which varied little according to the area considered. PCB deposition fluxes were ruled by the rainfall amount, while for PAHs, the fluxes depended on local anthropogenic characteristics. At the scale of the *Seine-Aval* treatment plant comparison of annual inputs of PTS in wet period indicated that PCBs essentially come from atmospheric sources whereas PAHs are derived from both atmospheric and urban runoff sources. At the scale of the sub-basin, atmospheric inputs to the soil (∑ 3 PAHs: 14–25 g km^{-2}, ∑ 7 PCBs: 5.6–25 g km^{-2}) represent the prevailing source for PAHs and PCBs, as compared to that from the disposal of urban sludge on agricultural plots (∑ 3 PAHs: 3–8 g km^{-2}, ∑ 7 PCBs: 0.5–2 g km^{-2}).

Keywords: Polycyclic aromatic hydrocarbon; Polychlorinated biphenyl; Atmosphere; River; Basin; Urban; Sludge; Seine

1. Introduction

Investigation of sources, dispersion pathways and fate of pollutants is a concern to both scientists and environmental quality managers (UNEP, 2005). A clear understanding of processes that intervene at the different transfer steps through the catchment system is essential for the protection of environment quality and water resorts, particularly in the Paris urban area where surface water constitutes 50 to 95% of the drinking water resource. Considering the number of toxins dispersed, especially synthetic organic components unintentionally arising from anthropogenic activities, the European Commission has defined a list of 33 priority substances in complement to that of Persistent Toxic Substances (PTS) listed by the United Nation Environmental

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Programme (UNEP, 2003). In this context, the sanitary and environmental effects, subsequent to dumping of a wide variety of contaminants belonging to different chemical classes, are of major concern (Nielsen et al., 1996). The Geological Survey identified 95 molecules (such as medicines, hormones, plasticizers) in treatment effluents and rivers in the USA (Kolpin et al., 2004). Although most of these molecules can be easily degraded, the occurrence of constant input fluxes leads to dynamic equilibrium of alarming concentrations that might induce long term effects upon the environment and human health.

This paper examines the pollution for the Seine basin where urbanization extent and intensive agriculture constitute major pollutant sources to the river. Half of the 16 million inhabitants of the basin are concentrated within 2% of its area (75 000 km^2). Because of the high population density, half of the trace metal found in the river Seine arise from domestic and industrial wastewaters of Paris (Observatoire de l'hydrologie urbaine, 2005). Two types of persistent and ubiquitous pollutants are examined in this paper: polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs), used as contamination indicators of water, soil and atmosphere of the river Seine basin (Fernandes et al., 1997; Guidotti et al., 2000; Ollivon et al., 2002; Teil et al., 2004; Motelay-Massei et al., 2004; Azimi et al., 2005).

PAHs of anthropogenic origin are produced by incomplete combustion of fossil fuel or oil leakage during domestic heating or motor vehicle traffic (Baek et al., 1991). In France, 250 tons of PAHs were released to the atmosphere in 2000 by combustion of petroleum products mainly from domestic heating and automobile traffic (CITEPA, 2006). PCBs are a group of 209 organochlorinated aromatic compounds which are chemically stable and found in various fluids and materials. They were mainly used in electric equipment until their ban in 1986. These chemicals appear to be involved in hazardous effects upon the biota, particularly human health. They might induce carcinogenesis, mutagenesis and hormonal disruption (Moore et al., 2002; Akingbemi et al., 2004; Sharpe and Irvine, 2004).

Our objective is to evaluate input sources of persistent toxic substances to the river Seine basin in relation with atmospheric deposition, wastewater discharge and sludge disposal.

2. Methods

The main thrust of our work was to show a link between urban sources and atmospheric transfer, wastewater collection and treatment and sludge landfilling, so as to give a balance of the relative importance of the different transfer pathways in the basin. For this, initially, atmospheric pollution behaviour and atmospheric deposit evolution were investigated at Paris, in central urban area, for an annual cycle from November 1999 to October 2000 (Ollivon et al., 2002). For atmospheric deposition sampling, a stainless steel funnel (0.36 m^2 surface) collected bulk deposition (rainwater plus dry deposition) and for ambient air sampling, a high volume sampler collected both the gaseous and the particulate phases (Blanchard et al., 2006). Then, the role played by the atmosphere in the dispersion of pollutants at different spatial and temporal scales was considered from 1999 to 2003. The experimental schedule included different scales of time and space and stretched beyond the Seine basin limits. Bulk deposition that included both rainwater and particulate dry deposition was used to assess the qualitative and quantitative fluctuations of the atmospheric compartment contamination, over a large transect through Northern France. Investigations were performed in different environments with various population densities, to determine the impact magnitude of PTS emissions from Paris agglomeration upon deposit contamination and subsequent fluxes. The sampling sites were: Ouessant (Island), Pleumeur (coastal), Evreux (semi-rural), Coulommiers and in 2001 and 2002, Eclaron (rural), Abreschviller (forest), Paris (urban). Their location is indicated on Fig. 1. Two sampling campaigns were performed first, from November 1999 to October 2000 weekly, then, from February 2001 to January 2002 every fortnight and last, from May 2002 to April 2003.

Observations were also carried out about the origin and variability of PTS inputs to the sewerage system and about the fate of contaminants during wastewater treatment procedures either with liquid effluents or with sewage sludge. Sludge represents a pollutant sink if incinerated or a pollutant vector if land-filled. The set of data concerning urban sludge for agricultural recycling in the river Seine basin was provided by the SIAAP and six other firms. However, our study did not take into account inputs to agricultural soils by waste from industrial activities such as food, textile, chemistry or paper industries.

PCBs were quantified by HRCG with a GC 8000 (Blanchard et al., 2006) and PAHs, by HPLC (Ollivon et al., 1995). For PCBs, 27 congeners were studied, including 7 designated in European legislation (Coleman et al., 1997): #28, 52, 101, 118, 138, 153 and 180. In addition, some results were also expressed as Aroclor equivalents to allow comparisons with previous data.