Sedimentary Archives Reveal the Concealed History of Micropollutant Contamination in the Seine River Basin

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Abstract Sedimentary archives provide long-term records of particulate-bound pollutants (e.g. trace metal elements, PAHs). We present the results obtained on a set of selected cores from alluvial deposits within the Seine River basin, integrating the entire area's land uses upstream of the core location, collected upstream and downstream of Paris megacity and in the estuary. Some of these cores go back to the 1910s. These records are complemented by in-depth studies of the related pollution emissions, their regulation and other environmental regulations, thereby establishing contaminant trajectories. They are representative of a wide range of contamination intensities resulting from industrial, urban and agricultural activities and their temporal evolution over a 75,000 km^2 territory. A wide set of contaminants, including metals, radionuclides, pharmaceuticals and up to 50 persistent organic pollutants, have been analysed based on the Seine River sediment archives. Altogether, more than 70 particulate contaminants, most of them regulated or banned (OSPAR convention, European Water Framework Directive (WFD 2000/60/EC)), were measured in dated cores collected at 7 sites, resulting in a large data set.

After drawing a picture of the literature devoted to sedimentary archives, the findings resulting from several decades of research devoted to the Seine River basin will be used, together with other studies on other French and foreign rivers, to illustrate the outstanding potential of sedimentary archives. The limitations of using sedimentary archives for inter-site comparison and the approaches developed in the PIREN-Seine to overcome such limitations such as selecting pertinent indicators (specific fluxes, per capita release, leakage rate, etc.) will be described. The very complex interactions between humans and their environment will be addressed through questions such as the impact on the spatial and temporal trajectories of contaminants of factors such as wastewater management, deindustrialisation within the Seine River basin, implementation of national and EU environmental regulations, etc. This chapter will show how such studies can reveal the persistence of the contamination and the emergence of new pollutants, e.g. antibiotics. It will propose indicators for the evaluation of the environment resilience and the efficiency of environmental policies.

1   Introduction

Sedimentary archives have been used since the early 1970s to reconstruct the past contaminant contents of river particles settled in the river continuum, particularly in the United States. They were used to reconstruct the past contamination of the Chesapeake Bay [1], the progressive eutrophication of the Great Lakes and their gradual contamination with mercury [2] and polychlorinated biphenyls (PCBs) [3]. Cores taken in the Mississippi River Delta integrate the contamination over 3.2 M km^2 [4, 5] for trace metals, polycyclic aromatic hydrocarbons (PAHs) and chlorinated organic compounds. Their analysis demonstrated that the lead (Pb) profile showed a doubling of concentrations from 1900 to 1970, followed by a significant decline of Pb contamination over this wide territory. These trends could not be demonstrated by the regulatory monitoring performed on filtered and unfiltered (whole-water) water sample pairs [6]: the analysis of a single core provided more valuable results on river contamination than 15 years of water quality surveys over the entire United States. To overcome the difficulties raised by this protocol, between 1991 and 2015, the United States Geological Survey (USGS) added the analysis of fine deposited river sediments to regulatory monitoring (Horowitz, pers. comm.) The sedimentary archive approach was then recommended by the National Oceanic and Atmospheric Administration (NOAA) [7].

In Europe, sedimentary archives were also considered in the 1970s in Alpine lakes [8] and the Rhine River delta [9]. In this basin, contamination for As, Cd, Cr, Cu, Hg, Ni, Pb and Zn started before 1920 and peaked in the 1970s, a trend confirmed later [10]. Most of these studies focused on the evolution of contaminant levels, generally at the river mouth or in deltas, therefore integrating the whole river basin.

In France, sediment archive analysis started with the metal contamination in the Garonne estuary (Gironde) [11] and then in the Lot reservoirs, a tributary of the Garonne, downstream of a former zinc mining site, which was the major source of Zn and Cd for this system [12, 13]. More recently, sediment cores have been analysed in the Seine basin [14, 15], the Loire basin [16] and the Rhône basin [17, 18]. In addition to trace elements and persistent organic pollutants (POPs), pharmaceutical products have been analysed more recently [19, 20]. This brief and non-comprehensive review does not cover the many lake cores analysed in France [21]. At all French sites, the sediment archives reveal the evolution of micropollutant contamination for the period when their regulatory analyses in water were limited or insufficient (<1990s) and long before, over periods that may exceed 100 years.

Apart from some notable exceptions, the archive-based studies, conducted by geochemists, have rarely documented the river basin history, which is needed to

correlate the sediment archives with the evolution of the river basin upstream of the coring site, and its complex interactions with societies.

The interdisciplinary approach, developed by the PIREN-Seine to go further than a geochemical description of the contamination trends, is presented in Sect. 2. Section 3 briefly presents the Seine River basin, our coring strategy and core analysis. Greater detail on the basin and its pollutant sources can be found in other chapters of this volume and in original papers for coring, dating and analysis details. Sections 4–7 present the different steps of the historical reconstruction of the basin contamination to allow for intra- and interbasin comparisons, as conceptualised in Fig. 1, illustrated with different particulate contaminants. The final section links the sediment archives with the basin history (past pressures, awareness, conflicts, societal responses and regulations, creation of institutions), i.e. defining the contamination trajectory, taking PCB as an example.

2 Long-Term Reconstruction of Past Contamination Trajectories Based on River Sedimentary Archives

2.1 *Conceptual Steps for the Study of Contaminants in Cores*

These studies can be schematised by a succession of steps, from the most common ones to those developed on the Seine River by the PIREN-Seine programme, as follows.

Step A. *Depth contaminant profile*: cored fine sediments – clay and silt deposits – are sliced, and then targeted contaminants are analysed on each slice. To minimise the effect of grain size on the contents of pollutants, samples can be sieved at the clay–silt fraction, typically <63 μm. Al, Th or Sc should also be measured as quantitative tracers of this fraction. Particulate organic pollutant contents are often normalised to the particulate organic carbon (POC). When the sedimentation rate is regular, the slices are averaged temporal windows, e.g. yearly averages. The best cores (e.g. lakes and reservoirs) provide an annual resolution. Cores taken in alluvial plains record the deposits corresponding to high flows and floods: the temporal record is more fragmented. Sometimes a single flood event corresponds to a multi-centimetre layer.

Step B. *Core dating*: generally using the measurement of Cs-137, an artificial radioisotope which provides three temporal markers: (1) the Cs-137 emergence in 1945, (2) a first maximum in 1962 originating from atmospheric nuclear test bombs and (3) a second and sharper maximum generated by the Chernobyl nuclear accident in 1986; Pb-210 decay can be used as well [22]. The evolution of the sedimentation rate is then used to convert the depth profile into a temporal evolution of contamination. Reliable geochemical tracers for dating before 1945 are lacking.
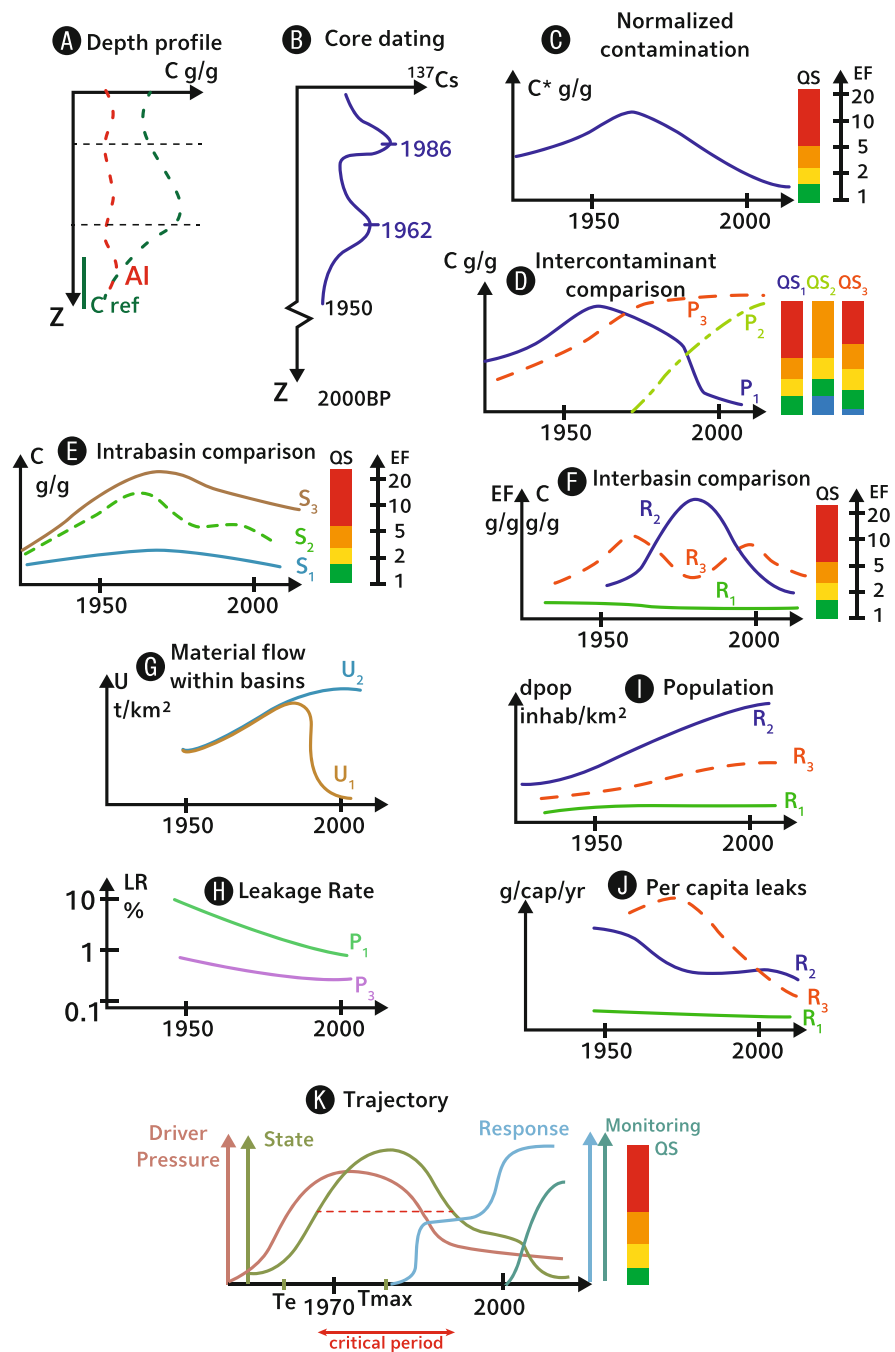
Fig. 1 Environmental contamination history based on multistep analysis of sedimentary archives, from state to environmental indicators and trajectories

Step C. *Temporal evolution of contamination and normalisation*. The contaminant contents expressed on a dry weight basis ($\mu g\ g^{-1}$ dw) are presented on a temporal scale. If the core slices are relatively thick, this evolution is presented as a succession of average contents in time slices. At this stage, there are two different options to interpret these profiles: (1) in the geochemical approach for metals, enrichment factors (EF) represent the excess contamination, which are the contents at time t doubly normalised to the background contents determined in natural conditions and to a tracer of the finer fraction. A variation of EFs is the Igeo index [23], which has a geometric progression to account for the increase of EFs, up to two orders of magnitude; (2) the regulatory approach is based on environmental quality standards for sediment quality or predicted no-effect concentrations [24]. Before the 1990s, these criteria were not available in France.

Once dated and normalised, the results can be used to compare how various pollutants have evolved at a given station (Step D), to compare stations within a given basin (Step E) and finally to compare basins (Step F). The understanding of these pollution trajectories needs historical socio-economic data (Steps G–K), as will be discussed in the following sections.

2.2 Contamination Fluxes Estimated from Floodplain Cores

In this approach, the average composition of the core at a given period is multiplied by the sediment flux for the same period to generate the contaminant flux. Then excess river fluxes are calculated [25].

This approach has several limitations, however. The extension of core analysis to river contaminant fluxes is based on the following hypothesis: (H_1) these deposits represent the flux-averaged quality of the particles carried by the river; (H_2) once deposited (without hiatus, which makes the age model uncertain), these particles are not (bio)turbated; and (H_3) the targeted compounds do not decompose (H_4) or migrate in the profiles. This set of assumptions is applied only over a long period, i.e. decades and more, considering that the contaminant fluxes are first controlled by the evolution of the chemical composition of river suspended particulate matter (SPM) much more than sediment fluxes. Note that H_1 is discussed for metals and radionuclide trends in [26]. The interpretation should also take into account the possible retention of contaminated particulates in the river basin [27], as in reservoirs, which could result in a decreased leakage not connected to environmental measures (e.g. reduction of point pollution sources). In some basins, the sediment loads can be reduced up to 95% as a consequence of large dam or reservoir cascades [28].

In the Seine River, some of these hypotheses have been checked. Floodplain particles are generally deposited from the peak discharge throughout the receding

stage. Cd analysis on river SPM during Seine River floods has shown that actually the maximum content is observed at the receding stage, i.e. when particulates settle in the floodplain [29], so that the sediment contamination is not underestimated and hypothesis H_1 is satisfied. For the assessment of excess metal fluxes, the effect of interannual variations of suspended loads (H_2) is minimised by taking a 2 to 5 cm core slice, homogenised and then analysed (corresponding to an average period of 4–6 years, depending on the sedimentation rate), to which an average Seine River interannual sediment load (700,000 t year^{-1}; [30]) has been attributed throughout the core. In some cores of the Seine River (e.g. [14]), the Cs-137 peak maximum is very high, an indication of the limited mobility of this element and the absence of post-depositional bioturbation (H_3, H_4).

Finally, it must be remembered that sediment transport in fluvial systems results in a mixing of particulates of various ages of mobilisation/remobilisation, ranging from a few hours (remobilised bottom sediments) to a few hundred years and more (erosion of floodplain banks). Thus, the comparison of contemporary river output of polluted particles to the present-day circulation of material is an approach that is mostly used to compare the life cycle of different chemicals (they are affected by the same bias) or to assess the general trend of exported pollutants. These reservations explain why modelling particulate pollutant transfer in the anthroposphere is so difficult.

2.3 Circulation of Contaminants Within River Basins

The river's excess fluxes of particulate contaminants are then compared to the circulation of products and materials within the anthroposphere, i.e. the material flow [31] such as imports and exports, transformation and use of products and recycling (e.g. for metals). In the case of metals, one should also take into account mining and smelting, which are not always present within the river basin intercepted by the core. The anthroposphere also gradually stores contaminants in contaminated soils, infrastructures, etc. In large basins, only a very small proportion of this enormous cycling of material actually leaks into the aquatic environment. In small urban streams, it may be greater. Such material flow analysis has been done for Paris for the metal circulation [32, 33] and for the whole basin for specific elements such as nitrogen and phosphorus [34]. Fluvial sediment archives are key elements in the validation of contaminant circulation in river basins. They concern environmental transfers between the major Earth system components – atmosphere, biosphere, hydrosphere, soils and sediments – which intercept different diffuse and point sources of contamination, which are considered as leaks to the river system: contaminated atmospheric fallout, agricultural runoff (sometimes impacted by reuse of urban sewage sludge on cropland), sewage outfalls and leaks from waste dumps.

The ratio of the exported river pollutant fluxes to the circulation of the related material within the intercepted basin generates the *leakage ratio* (Fig. 1, Step H), a dimensionless factor, which is a performance indicator for regulation measures

(pressures/driver). Another indicator of environmental pressure, rated to the basin population, is the per capita *excess load* (Fig. 1, Steps I and J), i.e. the excess flux divided by the basin's population upstream of the coring station (ELcap), expressed in g cap^{-1} year^{-1}. Both indicators can be used to compare the efficiency of environmental regulations in river basins and their trajectories, provided that past metal contents are reconstructed on the basis of dated sediment archives and past metal demands are recovered by an analysis of the area's economic history [25, 35, 36].

3 The Seine River Basin and Its Contamination by Micropollutants

The Seine River basin has one of the highest ratios of pressures to river sediment fluxes, which results in a very high contamination level in the lower Seine River, downstream of Paris. This contamination by metals and by numerous organic pollutants was not assessed before 2006 when new monitoring strategies were applied, following the EU Water Framework Directive (WFD). At that time, the first cores taken in the Seine River basin revealed the unknown long-term history of contamination, which reached very severe levels, when compared to current sediment quality guidelines.

3.1 The Seine River Basin: A Sensitive Territory Under High Pressures

In addition to the pressures (e.g. climate, morphology, land use, etc.) described earlier in this volume [37], the basin is characterised by a highly biased population distribution: about 70% of the basin's population is concentrated in the Paris megacity (2,750 km^2 with ten million inhabitants), which covers only 4% of the basin area. The upper Seine River and Marne tributary meet at Paris (Fig. 1 [37]), and the lower Seine River receives the other main tributary, the Oise River, downstream of the Paris megacity. The Paris megacity's main wastewater treatment plant (WWTP) at Achères, today called "Seine-Aval", discharges the treated effluent just upstream of the Seine-Oise confluence, some 70 river km downstream of the centre of Paris. Its capacity gradually grew in three decades to 8 M people at the end of the 1980s and then decreased to 6.5 M people because of the construction of additional WWTPs, which still makes it the largest WWTP in Europe.

The lower Seine River is structurally very sensitive to wastewater inputs, due to its minimal sediment transport and, above all, to its limited summer dilution during low flows (110 m^3 s^{-1}).

Most Seine River cores presented here intercept the 1945–2005 time window, which covers the reconstruction period after the World War II, from 1945 to 1974, also termed the Glorious Thirties. During that period, important changes and innovations occurred in the industrial sector. From 1970 to 2000, sewage collection and treatment plants were generalised in the Paris megacity (ten million inhabitants), i.e. more than two-thirds of the contamination point sources. Industries, which used to discharge their wastes directly into river courses, installed recycling processes and were gradually connected to sewage networks after 1970 [25]. Many heavy industries were delocalised outside the Paris area and then outside the Seine basin. During that period, new types of products were consumed, new drugs were used, and some products were legally withdrawn.

Since its beginning, the PIREN-Seine programme launched a series of studies on particulate micropollutants in the basin, particularly on metals [29, 38–41] and PCBs [42]. Other compounds such as PAHs, alkylphenols, PBDEs [15] and pharmaceuticals [19] were later studied. Various strategies have been set up: river profiles upstream and downstream of Paris, temporal variations with river flow and TSS at selected sampling stations, spatial distribution of the contamination using of fresh floodplain deposits [43], in-depth source inventories and circulation of compounds within the anthroposphere including atmospheric inputs. At the same time, the long-term history of human activities on the basin and of the social responses to water quality issues was addressed [33, 44–46]. The selection of core sites, core analysis and interpretation greatly benefited from this accumulated knowledge.

3.2 Regulatory Surveys Failed to Assess Micropollutants (1971–2006)

In France, trace metals and chlorinated insecticides were first surveyed within three national river quality inventories (1971, 1976 and 1981) on unfiltered waters, to comply with World Health Organization (WHO) drinking water criteria. Analyses were carried out 4 to 6 times per year at 25 key stations in the Seine River basin. Due to substantial bias (contamination during field and laboratory operations, excessive quantification limits in untrained laboratories), the first water quality inventory carried out by the newly created Agence de l'Eau Seine-Normandie (AESN) did not provide usable data [47]. The first environmental assessment of the European Environmental Agency in 1994 [48] also failed to produce quantitative figures on micropollutant concentrations.

The pioneering micropollutant studies in the Seine River basin were conducted on river SPM and concerned trace elements such as Cd and Hg [49, 50]. These academic studies failed to assess the state of extreme contamination they observed given the lack of sediment quality criteria and a reference natural background. Therefore, their use for environmental assessment remained very limited. The use of sediment archives had been recommended very early by marine scientists [51] and by the

Ministry of Transport's technical laboratory [52], but this approach was not used by the river basin authorities.

From 1982 to 1992, the six French river basin agencies developed their own monitoring strategies. The AESN turned to deposited sediments and rotating surveys on 204 sediment stations. The bed sediment analyses were carried out on the fraction <2 mm, i.e. including the sand fraction; unfortunately, ancillary data (e.g. % fine fraction, aluminium), which would have allowed for grain-size effect correction, were lacking, limiting data interpretation [53]. The official assessment of the metal contamination (Ministère de l'environnement, 1985) stated: "one finds high concentrations only for copper: metals in sediments are low". This statement ignored the severe Cd and Hg contamination occurring at that time, gradually unveiled by the PIREN-Seine programme in the 1990s using SPM analysis (e.g. [41]). In 2006, the monitoring within the WFD considered the dissolved fraction only. It therefore appears that the shifting strategies of micropollutant monitoring from 1971 to 2006 did not generate a general spatial and temporal assessment for contamination.

The sediment quality guidelines (SQG) that were or could have been used to assess the contamination of micropollutants is another limitation to contamination assessments. From 1971 to 2003, the guidelines concerning metal contamination evolved considerably, depending on the media (e.g. unfiltered waters, agricultural soils, dredged river materials, urban sludges). For a long time, the SQG did not exist in France, until they were finalised in 2003 by the French water quality assessment system, the SEQ-Eau [54]; sediment guidelines were therefore replaced by the WFD criteria on waters in 2006. Metal contamination in rivers has not been assessed on a national scale [55, 56], ignoring this critical issue.

3.3 Spatial Position of Sedimentary Archives Within the Seine River Basin

The core position within river systems should be chosen depending on the expected environmental information. In the Seine River basin, our coring strategy has considered very different positions of the intercepted basin area (Fig. 2). In addition, the coring sites are geographically located in Fig. 1 [37]. Coring sites are listed in Table 1 and greater detail can be found in the related articles. Cores taken directly from the river bed by a diver in 1994 [57], which cannot be dated, presented sharp contamination boundaries, suggesting erosion of previous deposits, are not used here.

Our pristine or sub-pristine site (PRI) is a headwater reservoir, which intercepts forested area. The medium impacts are studied on two floodplain cores, downstream of the city of Troyes (URB), on the Upper Seine River, and in the middle part of the Oise River (IND), a major Seine River tributary impacted by heavy industries. The maximum impact is assessed on a core taken 20 km downstream of the centre of Paris, in a navigated reach blocked by a sluice, which actually acts as a sediment trap
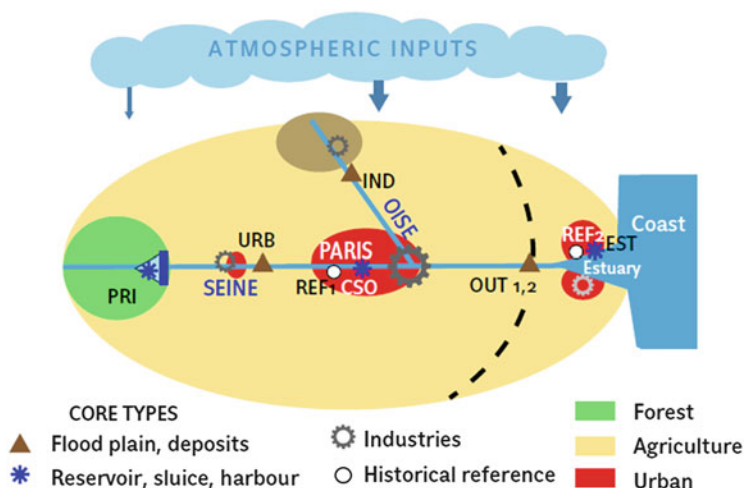
Fig. 2 Schematic positions of sediment archives used in the Seine River basin (see text)

Table 1 Principal coring sites considered in the Seine River basin

	Site (period covered)	Basin area (km^2)	Dpop[a] (cap. km^{-2})	Contaminants[b]	Reference(s)
EST	Rouen harbour (1965–2008)	75,000	208	Metals, major elements, lead isotopes, radionuclides, PAH, PCB, PBDE, OCP	[26, 60, 61]
OUT$_1$	Bouafles (1945–2004)	64,277	277	Metals, major elements, lead isotopes, radionuclides, PAH, PCB, PBDE, alkylphenols, antibiotics	[14, 15, 19, 60–63]
OUT$_2$	Muids (1933–2003)	64,484	276	Metals, major elements, lead isotopes, radionuclides, PAH	[14, 63, 64]
IND	Chauny (1954–2004)	3,280	45	Metals, major elements, lead isotopes, radionuclides	[14, 63]
CSO	Chatou (no datable archive)	44,670	na	Metals (Cd, Cu, Pb, Zn, Fe)	[65, 66]
URB	Troyes (1972–2001)	3,410	47	Metals, major elements, lead isotopes, radionuclides	[14, 63]
PRI	Lac de Pannecière (1931–2008)	917	9	Metals, major elements, lead isotopes, radionuclides, PAH	[64]

na not available
[a]Population density (Dpop)
[b]See related articles for further detail

for the main Paris combined sewer overflows (CSO) at low flows. The Seine River basin outlet is sampled on floodplain cores (OUT$_1$ and OUT$_2$), at Muids and Bouafles, near the outlet (Poses). Finally, an estuarine site on a harbour dock at Rouen (EST) integrates both direct estuarine sources with the upstream river

sources, particularly from major chemical industries, including P fertiliser production from phosphogypsum. The atmospheric inputs of contaminants are greater at the core sites URB to EST [58].

Floodplain sites store the fraction of the sediment flux circulating during floods, usually during winter, and act as a temporary sediment trap. The SPM transported by high-flow and flood events in the Seine River, i.e. 10% of the time, was estimated for the Marne River (12,900 km^2) at 60% of the total annual flux [30], and samples from floodplains are therefore interesting candidates for the estimation of yearly fluxes.

At the Bassée floodplain, downstream of Troyes (URB), the sedimentation rate 35 m from the river channel is 10 mm $year^{-1}$ on average (5 mm $year^{-1}$ at 70 m), and the flooding frequency is 1.6 $year^{-1}$ [30]. The Bouafles site, at the basin outlet (OUT_1), underwent particular study, and 12 cores were retrieved between 2003 and 2010. It was clear in the field that the site was under active accretion, with large trees buried above their roots. Unfortunately, in 2016, the inundated prairie was ploughed, evidencing the issue of on-site conservation of the archives in strongly anthropic basins.

The sampling strategy is completed by the assessment of the natural backgrounds. For organic pollutants, the PRI core can be considered. For metals, a prehistoric reference sediment (REF_1) is considered (Marne–Seine Rivers confluence at Paris-Bercy, dated 5,000 years BP). It is similar to another prehistoric estuarine sediment (REF_2) [59].

3.4 Core Dating

With the notable exception of the cores from Chatou for which no age model could be drawn, the cores exhibiting a clear chronology based on the measurement of radioactive tracers, ^{137}Cs and ^{210}Pb (and ^{238}Pu for EST), were selected. All cores were checked for a regular and fine-grain sedimentation profile. Core analyses are detailed in the published papers quoted here.

4 The Evolution of the Seine River Basin Contamination (1910–2015) Unveiled from Sediment Cores

4.1 Temporal Evolution of Persistent Organic Pollutants at the Basin Outlet and in the Estuary

4.1.1 Evolution of the River Basin at Its Outlet

The vertical profiles are represented for a set of different pollutants in Fig. 3. Since analyses provide multielemental contents for a given chemical family for each sample, the individual results are often clustered and given as "sum of". As the
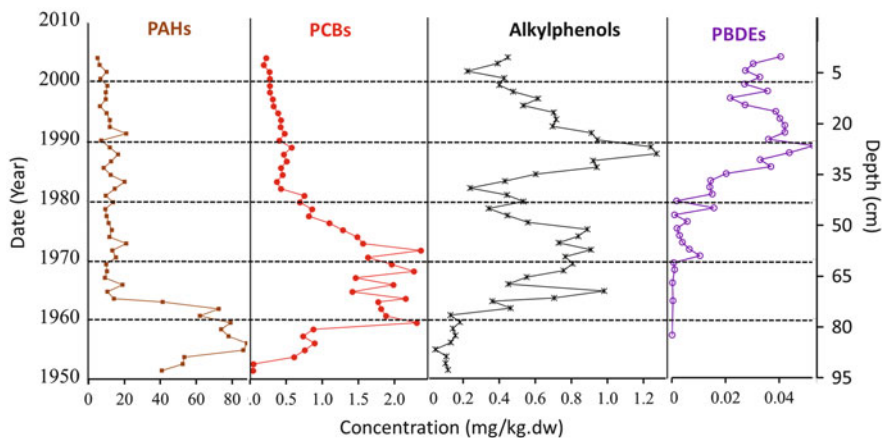
Fig. 3 POP contamination profiles (mg kg^{-1} dry weight) for PAHs, PCBs, alkyl phenols and polybromodiphenylethers (PBDEs) in the Seine River floodplain core OUT$_1$ [15]

conventions may have evolved over the last 50 years, for instance, for the sum of PCBs, one must check if the same basket of molecules is considered.

Four organic pollutant (POPs) families, namely, 13 polycyclic aromatic hydrocarbons (PAHs), 15 polychlorinated biphenyls (PCBs), 3 alkylphenols (APs) and 8 polybromodiphenylethers (PBDEs), were analysed on the Seine River outlet core [15]. The core provides detailed contamination records at a nearly annual scale. Each of them exhibits a specific trend, which can be described by several metrics: (1) the state of contamination at the core bottom, for those already featuring a contamination in 1945, (2) the emergence date (T_e), (3) the period of maximum (T_{max}), (4) the multimodal or monoclinic evolution and (5) the recent – last 10 years – trend and its slope.

The PAH profile reflects an old contamination associated with the use of coal for domestic heating, industries and power plants in the Seine River basin, which had never been impacted by coal mining. Most particularly, the post-war reconstruction period, from 1952 to 1960, is clearly marked with the highest levels (up to 90 mg kg^{-1} for $\sum$PAHs). A marked decrease of PAHs is observed between 1962 and 1965, by a factor of 10. The present-day level is fairly stable at 10 mg g^{-1}. This very rapid decrease of PAHs in flood sediments after 1960 cannot be explained solely by changes in coal consumption for domestic heating but also reflects a major industrial transformation with the closure of several gas plants based on the distillation of coal and the reduction of coal use in the steel industry. After 1968, coal was gradually replaced by fossil fuel in power plants, and contaminated sewage SPM was reduced by urban water management at the basin scale. Pre-industrial levels of PAHs – originating from fires – are not established; they should be a few mg kg^{-1} in the sediments [67].

PCBs, PBDEs and APs have different contamination profiles, reflecting the pattern of use and regulations specific to each family. The starting points (T_e) of

contamination correspond to the dates of their first industrial uses: the 1950s for PCBs, 1960s for APs and 1970s for PBDEs. PCBs and PBDEs present a sharp increase, while AP increases are more gradual. The Tmax occurred between 1960 and 1972 for PCBs, between 1965–1975 and 1985–1995 for APs and after 1990 for PBDEs. Finally, each of these peaks is followed by a period of sharp decrease, according to the regulations and the means of available substitution products. For example, a rapid decay of PCBs from 1972 to the 1980s was observed, followed by a slower period of decay. This trend is further discussed in Sect. 8. For APs, a rapid decrease has been observed since 1992 following the gradual restriction of their use, initiated by the OSPAR convention on pollutant inputs into the Atlantic Ocean and due to their substitution in detergents by other compounds such as PBDEs, which show a plateau pattern over the 1995–2005 period.

4.1.2 Evolution of Contamination in the Estuary

The EST core was sampled in a harbour dock that can be considered a perfect sediment trap when considering the ^{137}Cs profile used for dating and the sub-annual definition (1 cm year^{-1}) of the analysed layers, which provides a highly detailed record of the temporal variations of 95 potentially hazardous compounds (Fig. 4) [26]. The contamination pattern is similar to those observed in the OUT$_1$ core for PCBs, with maximum contents in 1970–1975. The comparison of actual PCB levels

Fig. 4 Evolution of POP contamination in the Seine estuary sediment core EST (1970–2008) [26]
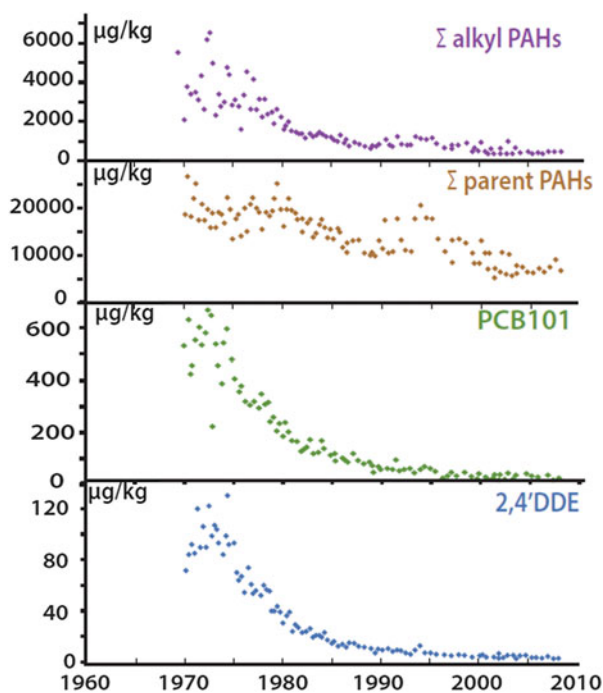
between the two cores is difficult because the analyses do not consider the same compounds. The content of dichlorodiphenyldichloroethane (DDE), a degradation product of dichlorodiphenyltrichloroethane (DDT), a chlorinated insecticide used until 1970, drops sharply after its total ban in the early 1970s.

4.2 Metals (Ag, Cd, Cr, Cu, Hg, Ni, Pb, Sb, Zn) and Arsenic (As) at the Seine River Outlet (1935–2005)

Considering the OUT_1 trends, the maximum metal EF occurred before 1965 (Fig. 5). Metal regulation started in the 1970s (Circulaire July 4, 1972, limiting the metal discharges from the surface treatment industry) so that the decontamination started ~10 years before the social responses to contamination. This pattern is first due to the industrial delocalisation outside of the centre of Paris and then from its suburbs; second, to changes in industrial practices, which include the gradual recycling of metals (e.g. in plating, in the 1980s); and third, to gradual use restrictions and then bans on Cd (its use as pigment was banned in 1995), Hg (in thermometers in 1999) and leaded gasoline (2000) [36].
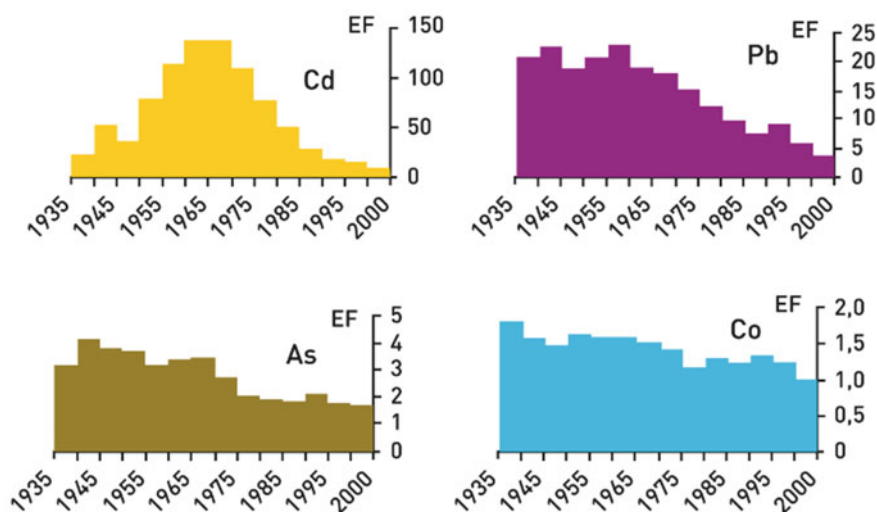

Fig. 5 Evolution of metal enrichment factors – 5-year averages – in the Seine River sediment archives at the basin outlet OUT[1] (1935–2005) [25]

4.3   Antibiotics at the Seine River Outlet (1954–2004)

Several antibiotics were analysed on the Seine River outlet OUT_1 core for a period of 40 years (1954–2004) [19]. The temporal evolution of norfloxacin, flumequine, oxolinic acid, sulfamethoxazole and nalidixic acid is specific to each compound (Fig. 6). Norfloxacin was detected below its quantification limit in two slices out of 45. Flumequine, oxolinic acid and nalidixic acid were accurately recorded: (1) their emergence date in the record is 2–3 years after or before their assumed first use; (2) they present a general maximum near 1988 for flumequine, oxolinic acid and nalidixic acid. The progressive decrease could be due to (1) changes in the
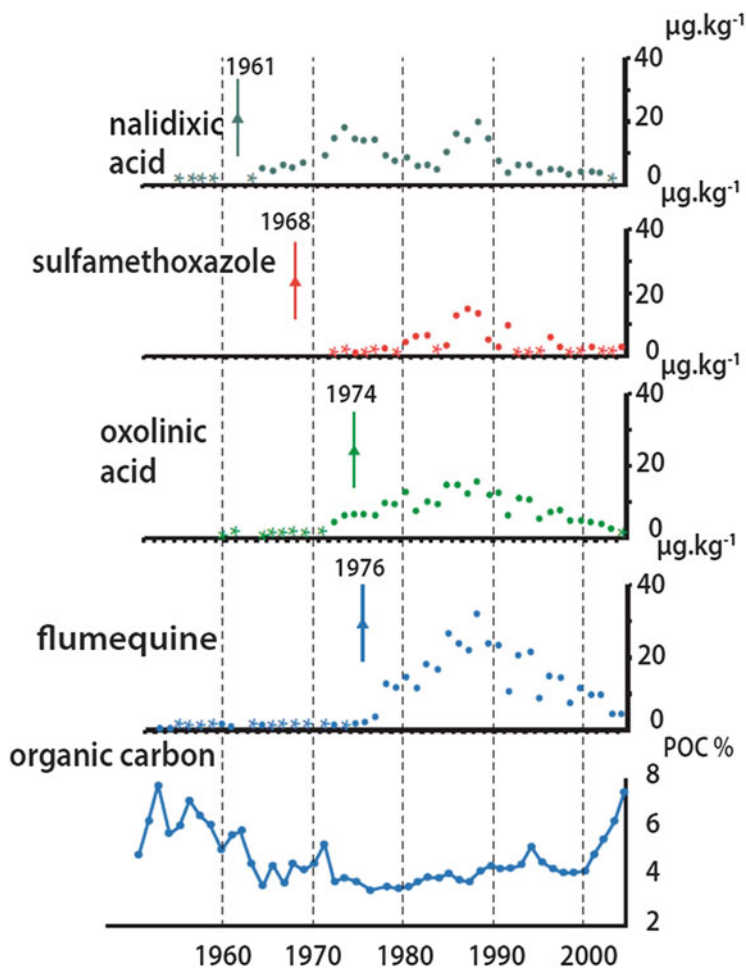

Fig. 6 Evolution of antibiotic contents in sediment archives of the Seine River at its outlet (OUT_1), from [19]. Vertical bars indicate the date of marketing authorisation

prescription strategies of practitioners, (2) a general decrease in antimicrobial consumption between 2000 and 2005 and (3) constant improvements made in the collection and treatment of wastewater coming from Paris and its suburbs during the last 40 years. Sulfamethoxazole has the least regular record and its detection lags 12 years behind its assumed first use. The regularity and the coherence of these profiles are encouraging: this is the first demonstration that an antibiotic's history can be recorded in sedimentary archives, extending this approach to other types of environmental indicators (e.g. [20]).

5 Intra-basin Comparisons in the Seine River Basin Reveal Contrasted Trends in Subbasins

5.1 Compared Metal Contamination History: The Cadmium Example

Cadmium is an iconic contaminant in the Seine River basin. No fewer than seven contamination profiles are compared in Fig. 7 (Step E, Fig. 1).

All enrichment factors were normalised to the thorium content in shale (12.3 mg kg^{-1}) and to the local background element content of Cd (0.2 mg kg^{-1}), except for the PRI core, located in the Morvan region, a crystalline shield where the background levels are Cd = 0.5 and Th = 18.4 mg kg^{-1} [68]. For the URB and IND cores, the Cd EFs are very low, compared to those observed downstream of Paris and in the estuary (OUT and EST). They clearly evidence that the maximum Cd contamination occurred in 1960–1965 at the outlet [25]. The high Cd levels found in the Lower Seine River (OUT and EST compared to PRI, URB, IND) result from the emissions of numerous plating workshops and industries within Paris, which have been greatly reduced in the last few decades.
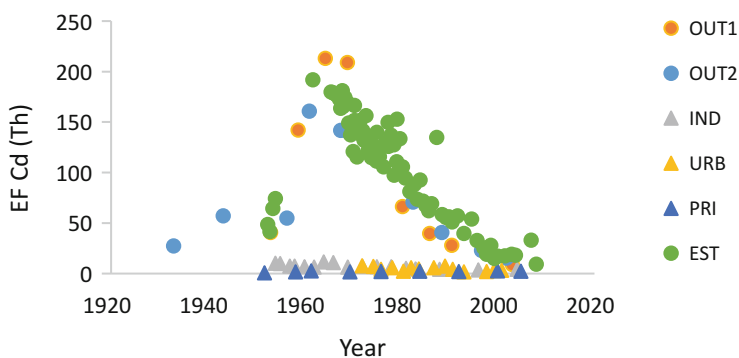

Fig. 7 Comparison of cadmium contamination trends in the Seine River sediment archives, upstream (PRI, URB, IND) and downstream of Paris (OUT$_1$, OUT$_2$, EST)

In the estuary site, the main Cd peak is observed around 1970 and a second one is observed in the late 1970s. A third very sharp peak is observed in 1987. These peaks are not observed in the OUT_1 core, suggesting that the 1970, 1975–1980 and 1987 estuary contamination episodes are due to very local events as the direct release of phosphogypsum wastes from a fertiliser industry using Cd-rich ore. There has been continuous dumping of "phosphogypsum residues" from the phosphoric acid industry, treating phosphorus ores at several locations in the estuary. Such waste materials were first released in the upper estuary, over several decades, then in the middle estuary and finally by barges in the outer estuary (Baie de Seine) with substantial overspills. These dumpings were on the order of magnitude of the Seine River sediment load (0.7 Mt. year^{-1} on average): 1974 (0.4 Mt. year^{-1}) to 1987 (0.4 Mt. year^{-1}) with a maximum between 1980 and 1984 (1.6–2 Mt. year^{-1}) [60]. The EST core reveals this long-term impact on many metals and phosphorus (P content up to 5,000 mg kg^{-1} in EST sediment vs 800 mg kg^{-1} for the basin reference) and for Y, La and lanthanides, U, Cd, As, Cr, Mo, Th, Ba, etc. [26]. All show a marked increase of their contents in the estuarine core with the same temporality as phosphorus. This contamination source is estimated to account for 50–70% of these elements' contamination, reaching 90% for Y. This type of generalised contamination by a mineral chemistry industry has rarely been documented.

These contaminations were superimposed to those originating from the upstream river basin: the EFs recorded in the estuarine sediments were extreme in the 1970s, from 100 to 1,000 for many elements (Hg, Pb, Ag, Cd), making this waterbody one of the most polluted in Europe [69].

5.2 Compared POP Contamination History: The PAH Example

The comparison of the PAH contamination trends at five coring stations is illustrated in Fig. 8 (Step E, Fig.1). The sub-pristine site (PRI, Fig. 2) is considered to be representative of the non-impacted levels. Particle size analysis of the sediment core reveals that the core bottom layers (dated between 1952 and 1955) correspond to a mixture of soil and sediment, which is consistent with the reservoir's impoundment date (1949). Therefore, the analyses performed on layers deposited after 1955 are considered as representative of the contamination trend of this area of the basin where direct industrial and urban sources of contamination are minimal.

In the 1950s, the PRI PAH levels were below 0.2 mg kg^{-1}. Then PAHs increased, possibly due to atmospheric inputs. The maximum levels reached 10 mg kg^{-1} ($\sum$13 PAHs). In 1992, the contamination was still present at 2 mg kg^{-1}, which is five times less than at OUT_1 at the same time but remains high for this type of basin slightly impacted by anthropogenic pressure.

The PAH contamination at the Seine River basin outlet was assessed in three cores taken 10 km apart (two cores at OUT_1 and one core at OUT_2). This allows for a
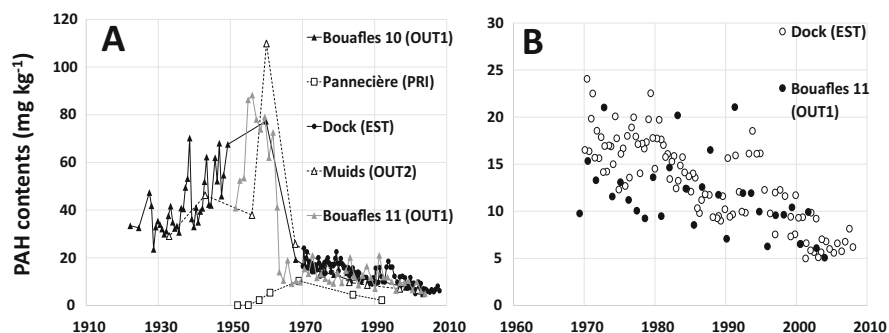
Fig. 8 (**a**) Comparison between the sum of PAH contents (13 PAHs, mg kg^{-1}), in the Seine River sediment archives, downstream of Paris (Bouafles and Muids sediment (black, grey and open triangles)) with the upstream sub-pristine reference (Pannecière reservoir sediment, open squares). (**b**) Enlargement of the 1970–2010 period for basin outlet (Bouafles) and estuary (Dock). These contents are also compared with estuary sediment (dark circles)

validation of the sediment archive approach: there is no significant PAH source between Muids and Bouafles and the two OUT$_1$ cores should be identical. Actually, the contamination pattern is very similar in the three cores (date of maximum contamination, slope of contamination and decontamination), which proves the reliability of the general trend. Small differences (less than 30%) in peak values may stem from local patterns of sedimentation or sediment reworking.

Focusing on the 1970–2000 period, similar PAH decontamination rates are observed in the outlet cores (OUT$_1$ and the EST), but second-order peaks in 1997 and 2004 showed by the EST core suggest a direct PAH source in the estuary. Also, the decontamination rates for OUT$_1$ (3.0 $\pm$ 0.3 mg kg^{-1} year^{-1}) and for the upstream site (PRI) (2.8 $\pm$ 0.2 mg kg^{-1} year^{-1}) are very close, representative of the general decrease of PAH emissions at the scale of the Seine River basin, probably linked to the decrease in coal consumption over this period (see Sect. 4.1).

6   Interbasin Comparison of Metal Contamination Trends in Western European Rivers Using Sedimentary Archives

Interbasin comparisons can be performed at basin outlets which integrate the whole catchment area (Fig. 1, F). The evolution of metals in nine rivers in Europe (Rhine Volga and Danube, Mersey, Scheldt and Garonne) and in the Mississippi Delta shows various levels of contamination, highly controlled by the river SPM contamination level [70]: the Mississippi had the lowest EFs and the highest dilution power

by erosion-derived SPM, while the Rhine and Seine Rivers show opposite characteristics.

The level of contamination of river sediment strongly depends on the ratio of the pressure over the dilution power (sediment load derived from erosion). The Seine River at its mouth has a much higher ratio than the Rhône or the Mississippi. Similar control is observed for PAH contamination when comparing the Orge River, an urban sub-catchment of the Seine River, and other worldwide urban catchments [71].

Compared to the rivers cited above, the Seine River outlet exhibits the highest metal concentrations for the 1940–1960 period, but small rivers impacted by big cities present even higher contamination levels [35]. This is illustrated (Fig. 9) when comparing the metal contamination trends in cores from the Zenne River floodplain (Brussels), a Lambro reservoir downstream of Milan (data courtesy of Luigi Vigano, CNR), the Quentzee Lake on the Havel-Spree River (Berlin) and the Seine River outlet [72]. The maximum contamination reached in the Seine River core is always much lower than for the other rivers (Table 2A). The city impact ranking for the general metal contamination is as follows: Havel/Spree > Lambro > Zenne > Seine rivers. Note that if the CSO core (Seine River), whose sampling site is more similar
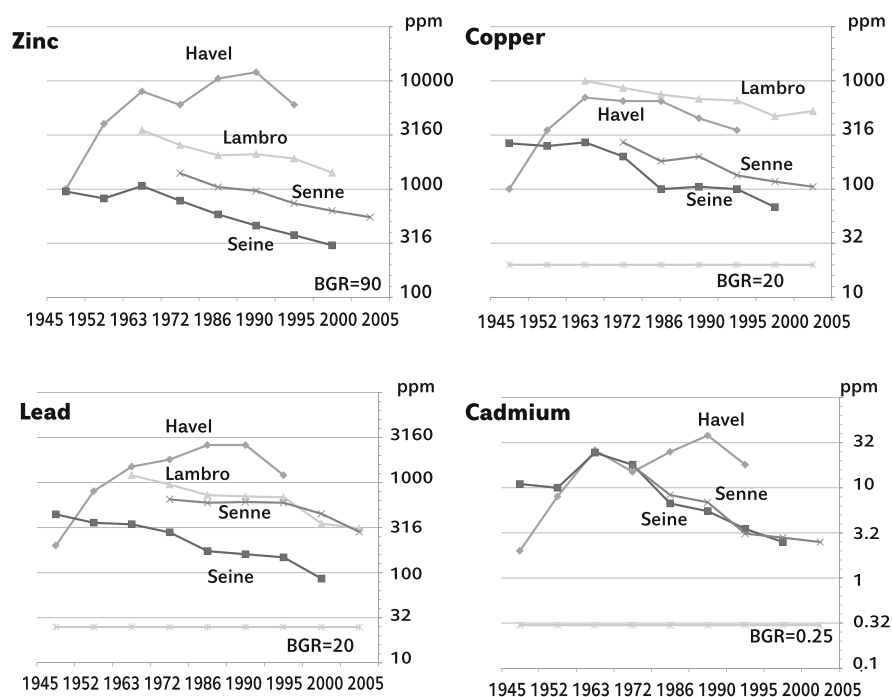

Fig. 9 Comparison of metal contamination trends, resulting from sedimentary archives (ppm or mg kg^{-1}, log scales), in four rivers impacted by major European cities, the Havel-Spree River and Berlin, the Lambro River and Milan, the Zenne River and Brussels and the Seine River and Paris, and background concentration values (BGR) (from [72])

Table 2 Comparisons of metal contaminations in world rivers from highly industrialised countries, using sedimentary archives at their peak contamination stages

	Cd	Hg	Pb	Zn	Basin area (km^2)	Population (10^6 cap.)
A. Maximum contamination ($mg\ kg^{-1}$) observed at urban site (year)						
Lambro (Milan) (1)	n.d.	n.d.	1,400 (1964)	3,500 (1963)	2,747	4.2
Zenne (Brussels) (1)	15.5 (1974)		700 (1979)	1,300 (1974)	1,160	1.3
Spree/Havel (Berlin) (1)	38 (1990)		2,600 (1986)	12,000 (1990)	10,105	3.5
Seine (Paris) (1)	28 (1960)		450 (1945)	1,150 (1967)	65,000	10
European and US urban sewage PM (1970–1980) (1)	140		410	3,000		
European rivers background (6)	0.25	0.04	20	90		
B. Per capita in French rivers at outlet positions ($g^{-1}\ cap^{-1}\ year^{-1}$)						
Seine (2)	3.4	0.6	50	160	75,000	15
Loire (3)	0.5	0.1	10	40	117,800	7.7
Garonne (4)	1.9	n.d.	50	170	56,198	3.25
Rhône (5)	1.5	0.5	100	190	97,800	14
C. Per capita in other basins ($g^{-1}\ cap^{-1}\ year^{-1}$)						
Western European rivers (6) (7)	2.8	0.74	33.7	175	0.53	123
Mississippi (8)	0.58	0.175	30	47.5	3.2	67
Danube (9)	0.6	–	9.5	70	0.8	81
Montreal (10)	0.23	0.05	7.6	64	–	–

(1) [72] Median of 21 analyses, adjusted to Al = 75,000 mg kg^{-1}, (2) [25], (3) derived from [16], (4) derived from [11], (5) derived from [18], (6) derived from [70]. For Montreal: per capita release into St Lawrence River based on sewer analysis. (7) Elbe, Meuse, Garonne, Rhine, Scheldt and Seine river basins [70], (8) [5], (9) [77], (10) [78]
n.d. not determined

to the Spree/Havel, Lambro and Zenne river sampling sites, had been usable as a dated archive, the ranking would have been very different. The Cd content in the CSO core was up to 100 mg kg^{-1} [66]. The decontamination of the Lambro, Zenne and Seine rivers developed around 1960, but the decontamination of the Havel-Spree did not occur before 1990, after the German reunification. Water quality studies in the River Elbe since the German reunification in 1990 revealed the improvement of water quality due to the reduction of industrial wastewater emissions since 1990 [73].

7 Circulation of Material Within Basins and Its Impact on River Fluxes

In a fully sustainable society, the products and goods containing potentially harmful substances should circulate without any leak into the environment (air, soil, water, coastal zone). The previous sections have shown that this is not the case and that river SPMs are a very good indicator of the way a society is handling these environmental issues. Sedimentary archives add a precious time-depth to this question, provided that additional information on the general circulation of targeted materials within the anthroposystem are available. This approach has been applied to establish the metal circulation patterns in Western European rivers (Steps G–K, Fig. 1).

7.1 Leakage Ratio of Metals Within the Seine River Basin (1950–2005)

The national demand for Cd, Cu, Hg, Pb and Zn has been reconstructed since 1900 on the basis of the national consumption of all metals and products containing metals. The metal demand in 1900 is the reference value and is set at 1.0 (Fig. 10, left). The metal demands greatly increased (2.5- to 5-fold) after World War II, and the present-day consumption (year 2000) is much higher than in 1950, except for Hg. This increase is faster than the population increase in the Seine River basin, indicating that the per capita consumption has greatly increased, except for Hg after 1975 and Cd after 1985. The Hg demand has dropped by nearly two orders of magnitude since 1975, and Cd demand has dropped since 1995, both as a result of regulations (gradual Hg ban since the 1970s and Cd use limitation beginning in 1995).

The leakage ratio (in %) is the ratio of exported river pollutant fluxes over circulation of the related material within the intercepted basin. It can be regarded
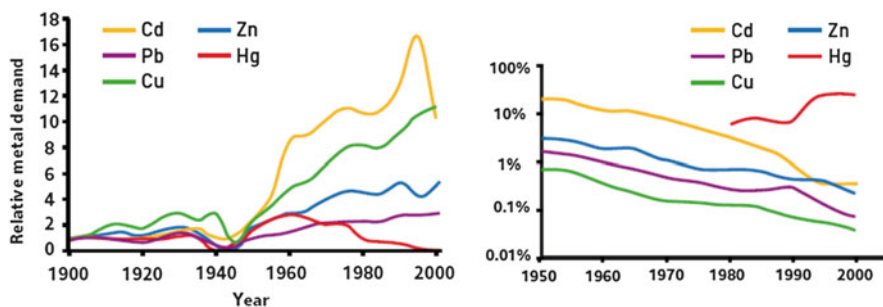

Fig. 10 Left circulation of metals in France, normalised to its value in 1900. Right: leakage rates in % estimated for the Seine River basin [25]

as an efficiency indicator of the capacity of a given society to recycle its metals. It has dramatically decreased for all metals since 1950 (e.g. from 0.8 to 0.05% for Cu), except for Hg, which remains at a high ratio, around 10% of leaks. The metal exported fluxes calculated from core data are therefore powerful tools to observe the general decontamination trend in Europe, which can be linked to the local type of economy and environmental policies, defining the trajectory of a given environmental issue [74]. In the Seine River basin, the location of recycling facilities is a key control factor in contamination by metals [33]. The recycling process (collection of used metal-containing devices and their retreatment outside of the Seine River basin and often outside France, i.e. a kind of pollution delocalisation [36]) seems to have been effective mostly after 1980.

As pointed out in Sect. 2, this leakage estimation is based on several hypotheses: (1) all river particles have the same age; (2) their age is very recent – a few years on average – so that the average export at time t $\pm$ a few years can be compared to the circulation data for the same period. When there is evidence that the river particles have extended age ranges or that they have greater ages, these hypotheses are not valid. In the Seine River, the floodplain deposits are relatively fine (silt), so that their average transit time throughout the river network ranges from a few weeks (large floods) to a few years. The occurrence of antibiotics in core sediment (Fig. 6) within 1 year after first arriving on the market and the decline of DDT metabolite, which started with the ban of this insecticide in France, suggest a rapid reaction of Seine River fine sediment (median grain size in the silt fraction) to source changes. In contrast, coarser material (1 cm) travels in the Seine River at secular rates: gravels contaminated by iron smelting during the eighteenth century have actually been used to study the very slow bedload movement in the Upper Marne (see [75], box 1).

7.2 Intercomparison of River Contaminant Fluxes in Relation to Their Population in Western European Rivers

The excess load of metals and the load of micropollutants can be rated by the population of river basins, to generate the per capita excess load (ELcap), an indicator also used for major ions or nutrients to compare and scale these per capita loads to economic indicators, such as per capita energy consumption [51, 76]. For metals, the comparison of the per capita loads for Western European rivers, the St Lawrence, Mississippi and Danube rivers, at the period of their maximum contamination level near 1970 (Table 2A) showed similar orders of magnitude for each metal, suggesting the existence of a metal metabolism common to these old industrial countries for similar levels of development and environmental regulations [70, 72]. Based on the metal content, the general pressure ranking is $Zn \gg Pb \geq Cu > Cr > Ni > Cd = As > Hg$, but their maximum EFs in river particulates are completely reversed: $Hg > Cd > Pb > Zn$. When looking in greater detail, each
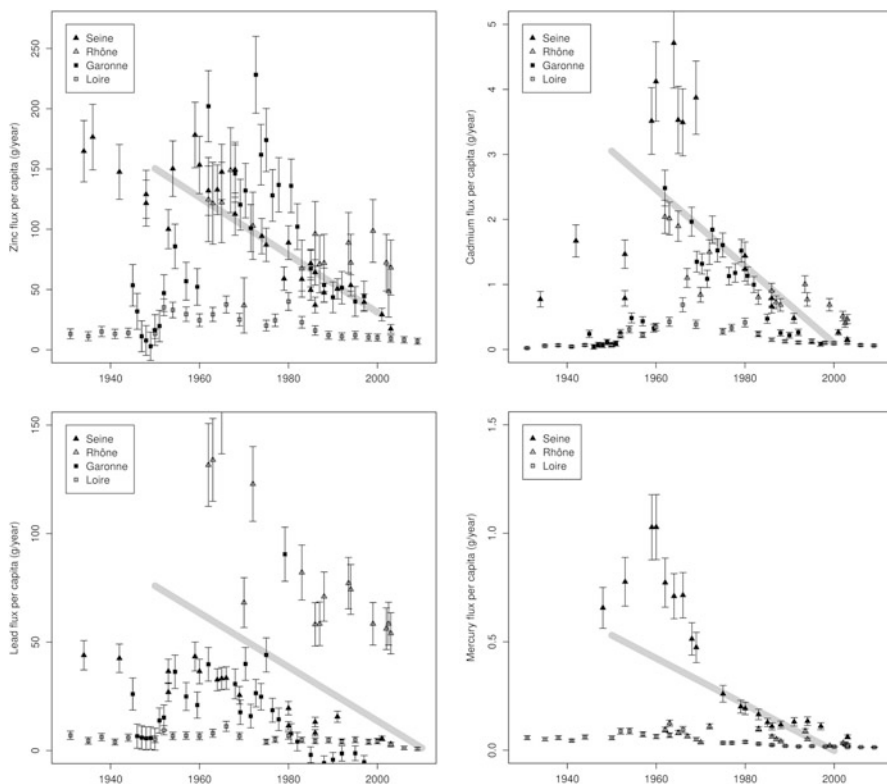
Fig. 11 Long-term evolution of metal fluxes (in $g^{-1}\ cap^{-1}\ year^{-1}$) carried by French rivers (Rhône, Seine, Loire and Garonne Rivers). The thick grey line is the per capita linear regression for the French rivers (population-weighted)

basin may have its own maximum period, depending on local environmental and political history [72].

ELcap trends were calculated for Cd, Hg, Pb and Zn based on the sediment archives of the four main French rivers that were studied: the Seine [25], Loire [16], Garonne [11] and Rhône [18] (Fig. 11). All per capita loads in French rivers show a regular decrease after 1960–1965, while the metal demand shows an inverse trend (Fig. 10). This is an outstanding indication of the improved environmental efficiency for metal emissions over the long term in France. It could not have been demonstrated on the basis of the regulatory monitoring of metal contamination in these basins, which is less than 25 years old, and sometimes not relevant for the river contaminant fluxes (e.g. survey of the contamination of aquatic mosses by the Rhône basin authority).

The levels of per capita loads reveal a totally different ranking of the four basins, with regard to their actual river SPM contamination (Table 2B). While the levels of

metal concentrations are always lower in the Rhône River and higher in the Seine River, i.e. inversely to their suspended solid loads, the per capita loads of the Seine River are the greatest for Hg and Cd, the highest per capita Zn fluxes are observed in the Garonne River, while the lowest fluxes of all metals are in the Loire River. The top ranking of the Seine for Cd, although well-known mine tailings have also contaminated the Garonne for decades [79], is due to the numerous plating workshops and industries in Paris in the 1960s until the 1980s. The particular pattern of Pb in the Rhone River suggests a shift in reference values. In this highly impounded basin whose solid discharge was considerably reduced during the last century, it is possible that a catchment with a high Pb background has become a higher contributor to the Pb background in the last few decades.

These per capita figures for French rivers are compared to those of other industrial countries, at the peak contamination, i.e. the pre-regulation stage (Table 2C). They present a remarkable similarity.

8 Trajectories of the Environmental Issues Affecting River Basins and Society: Example of PCB Contamination in the Seine River Basin

The trajectory of the water quality situation (Fig. 1) includes the following components [74]: (1) the evolution of a state indicator measured during the targeted period; (2) the relevant pressure or driver indicator; (3) the first awareness of a deterioration of water quality, its social recognition, environmental monitoring and assessment and reporting; and (4) the technical and/or regulatory responses provided. Here, the trajectory is completed by a brief analysis of how scientific knowledge of this issue has evolved, in particular its controlling factors. For micropollutants, the state indicator might not always be derived from the regulatory survey (see Sect. 3) but from the sedimentary archives. Historical archives are used to reconstruct other components of trajectories: the scientific knowledge of the inhabitants' perception of the problem and the interactions between key actors, including controversies. This approach is therefore typically interdisciplinary. It has been tested in the Seine River basin for trace metals and requires a large amount of information to complete these components and the circulation of the contaminant within the anthroposphere, as for metals [36].

Figure 12 is an attempt to describe the trajectory of PCBs in the Seine River basin. The state indicator is the total PCB content in the dated core profile from [15], taken at the river outlet (65,000 km^2).

PCBs are one of the first industrial products to be regulated and then banned. Following the alert made by Rachel Carson (Silent Spring, 1962), PCBs and other organochlorines such as DDT and lindane were an early concern of some French scientists [80]. In 1972 these compounds were included on the blacklist of UNESCO's International Oceanographic Commission and, in turn, on the list of
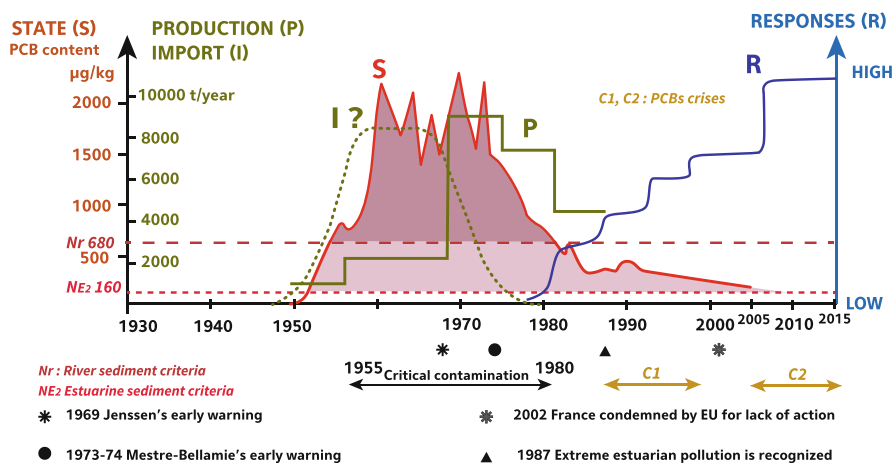
Fig. 12 Schematic trajectory of the PCBs dumped into the Seine River, based on the data recorded at the OUT1 coring site [15]

products to be carefully monitored in French estuaries and river basin outlets [51]. Marchand [81] stated that the Seine estuary sediments had the highest contamination in PCBs with regard to all other French study sites.

PCBs have been commonly used in France since 1950 in transformers and condensers. They were first imported; then in the late 1950s, they were manufactured in France by two chemical companies, outside the Seine River basin. In 1979, Colas [82] estimated that the total consumption of PCBs at that period was between 40,000 and 50,000 t, i.e. about 12,000–15,000 t for the Seine River basin, considering the proportion of industries in the Seine River basin compared to other river basins. The societal response to the PCB issues (Step K, Fig. 1) lags far behind the maximum contamination period, itself unnoticed until the first analysis of PCBs in the core.

The preliminary analysis of PCBs in Seine River basin fish revealed fish contamination [42]. The mussel watch established since 1979 in the estuary by Ifremer confirmed an extreme contamination by PCBs, when compared to other French and foreign estuaries [81]. However, this issue could not be handled at the level of a single river basin. At the national level, the banning of the further sale and use of PCB-containing devices was an answer to the first socio-political PCB crisis (C1, Fig. 12). The second national crisis (C2) in 2005 was triggered by a health regulation when PCB values exceeding the WHO criteria for edible fish were detected in bream fished in the Rhône River, near Lyon [83]. As a result, all fish from the Rhône basin were officially declared as non-edible, causing great concern on the overall quality in this basin. This event, in which fish were used as environmental indicators, triggered a national PCB plan (PCB 2008–2012) which forced the Seine River authorities to undertake specific assessments and accelerated the elimination of all former industrial devices still containing PCBs.

The current evolution of the contamination of the Seine River and the estuary is directly provided by the core analysis of the main PCB congeners ($n = 7$). The OUT$_1$

river outlet core [15] presents the following pattern (Fig. 12): (1) an emergence of contamination in 1950, as in the Rhône River [17], (2) a sharp increase until 1960, (3) then a 15-year plateau between 1,500 and 2,200 µg kg^{-1} until 1975 followed by (4) an exponential decrease to 200 µg kg^{-1} in 2005. The Lower Seine River was under critical contamination from 1955 to 1980 when applying the criteria for dredged sediments in rivers (Nr = 680 µg kg^{-1}) and from 1952 to 2005, when using those for estuaries (Ne$_2$ = 160 µg kg^{-1}) [84]. When the PCB content exceeds these levels, the sediment should be treated or appropriately stored [83]. This trend is confirmed by that observed in the estuarine Rouen core [26], which covers the period from 1968 to 2008: the maximum contamination is noted in 1972, as in the OUT$_1$core, with total PCBs at 4150 µg kg^{-1}, and then a regular 30-fold decrease to 140 µg kg^{-1}. The higher contents in the estuary can be attributed to direct sources and to finer sediments in this core (Al contents at 4% vs 3% in the Seine River SPM). Highly polluted estuarine sediments and agricultural soils in the Seine River flood-plain are a long-term contamination heritage. When the second crisis was triggered in 2005, the PCB contamination was already reduced by a factor of 20–30 compared to the 1970s: at that time it was probably considerably above the current WHO criteria that could have resulted in health problems for regular fish and seafood consumers, but this issue was not detected. Sedimentary records unveil this past environmental history.

9 Conclusions

The emergence of new water quality issues is continuous as scientific knowledge progresses and analytical capacities are developed to detect new deleterious substances at reduced costs. Environmental regulations are evolving as well, but with a time-lapse of several years or decades between the start of use of a compound and its recognition as a potentially harmful agent to the environment or health. For the micropollutants studied in the PIREN-Seine programme, the medium time lapse is at least 30 years. Furthermore, the environmental release of many of these xenobiotics is not regulated, such as for antibiotics. It is common knowledge that resources are lacking to implement surveys dedicated to unregulated compounds. Finally, as in the case of PCBs, the regulatory survey which followed the water compartment was not adequate, missing the target media: biota and SPM. If the biota is now included as a target media in the WFD, SPM guidelines are still lacking.

Sedimentary archives make it possible to document the long-term state of the river quality for many issues. Water quality trajectories should then be complemented by substantial research on environmental history: (1) definition and recognition of the issue, with effects, causes, control factors, indicators, metrics, criteria and/or scales of water quality; (2) implementation of sustainable observatories for long-term monitoring and surveillance on proper media, at proper stations and frequencies, with appropriate analytical tools; (3) assessment of water quality and its declaration by relevant authorities; (4) identification of the origin(s) of the

issue; (5) social consensus on the issue's importance, origins and solutions; and (6) funding and implementation of appropriate solutions.

None of the long-term contaminant trajectories in the Seine River – nor in any other French river – was available before the use of sediment archives, since the monitoring of these pollutants is very recent and/or has been inefficient, due to lack of appropriate analysed media and analytical difficulties. The studies presented here demonstrate the effectiveness of sedimentary archives, which give a comprehensive overview of the contamination in the Seine River basin. The results presented here are in agreement with previous studies dedicated to other European rivers and have highlighted for the first time the specificities of the Seine River basin compared to other large French and European basins. Sediment archives are still present for all European rivers, waiting for multidisciplinary teams, aware of the sedimentary, hydrological, chemical and historical issues, who will be able to elucidate their contamination history.

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