Annual budgets of suspended particulate matter and associated pollutants in the Rhone River from Lake Geneva to the Mediterranean Sea

Bilans annuels de matières en suspension et de polluants associés sur le bassin du Rhône, du lac Léman à la mer Méditerranée

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RÉSUMÉ

En 1975 la convention de Barcelone a mis en lumière l'importance de surveiller les rejets de matières en suspension (MES) et de polluants associés dans la mer Méditerranée. Dans cette zone, le Rhône constitue le principal apport d'eau et de sédiments. Le but de notre étude était de calculer des flux de MES et de polluants associés à l'échelle du bassin versant, afin d'identifier les principaux affluents contributeurs, et appréhender les possibles variations temporelles. Pour cela nous avons acquis un large jeu de données basé sur des mesures quasi-continues de débit et de concentrations en MES et polluants particulaires, dans le Rhône et ses principaux affluents. Nous avons obtenu de nouvelles relations entre le débit et la concentration en MES sur les affluents. La relation obtenue pour le Rhône à Arles a été améliorée par rapport aux études précédentes grâce à l'acquisition et l'incorporation d'une gamme de donnée élargie. Le flux moyen annuel de MES exporté par le Rhône a été estimé à 6.4 Mt pour la période 2000-2015. Les contributions des affluents s'échelonnent comme suit : Isère (26 %), Durance (21 %), Haut-Rhône (11 %), Saône (5 %). Les flux moyens annuels de mercure et PCB 180 ont été estimés à 680 et 15 kg/an respectivement sur la période 2000-2015. Notre étude apporte des informations nouvelles et récentes sur les flux de MES et de contaminants à l'échelle du bassin du Rhône, basées sur plusieurs années de mesure et un échantillonnage haute fréquence. Nos résultats soulignent l'importance des mesures à long terme pour un calcul plus fiable et plus robuste des flux de MES et de polluants associés.

ABSTRACT

In 1975 the Barcelona convention highlighted the importance of monitoring SPM fluxes and related pollutant outputs to the Mediterranean Sea, of which the Rhône is the main supplier of water and SPM. The aim of our study was to calculate representative fluxes of suspended particulate matter (SPM) and associated pollutants at the entire basin scale, for a better understanding of spatial distribution and temporal trends over the entire Rhône watershed. We used an original dataset based on quasi continuous monitoring of discharge, SPM and pollutants concentrations in the Rhone River and its main tributaries. New relationships between SPM concentrations and water discharges were obtained for tributaries. For the Rhône River (at the Arles station) the relationship was improved compared to previous studies, thanks to the collection of a wider range of data. The annual mean of SPM loads exported by the Rhône to the Mediterranean Sea was 6.4 Mt for the period 2000-2015. Contributions of tributaries ranged as follows: Isère (26 %), Durance (21 %), Upper Rhône (11 %), Saône (5 %). Concentrations of mercury and polychlorinated biphenyls (PCB) showed a decreasing trend with increasing discharge, which could be due to the presence of coarser material during floods. Mean annual loads of PCB 180 and mercury at the outlet of the Rhône river were 15 kg/yr and 680 kg/yr respectively, for the period 2000-2015. Our work provides new insights in SPM and pollutant annual loads over the Rhône watershed, based on long term monitoring and high resolution data. These results highlight the need for continuous measurement for accurate determination of annual loads of SPM and associated pollutants.

MOTS CLES

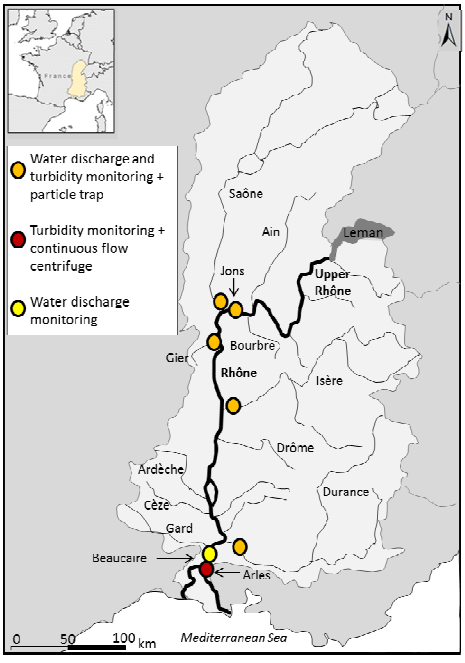
Suspended particulate matter, annual fluxes, mercury, PCB, Rhone River, rating curves

1 INTRODUCTION

In the Mediterranean Sea area, the Rhône is the main supplier of water and SPM, together with various organic and inorganic pollutants that can have significant impact on marine environments by altering water quality. Thus it is important to calculate representative fluxes of suspended particulate matter (SPM) and associated pollutants at the outlet of the Rhône River, and also on its tributaries for a better understanding of spatial distribution and temporal trends over the entire watershed. To this end, data on water discharge, SPM concentrations and pollutants levels in the river and its main tributaries are required. SPM concentrations (Cs) are traditionally assessed *via* empirical power laws relating Cs values to the flow discharge (Q). These relationships depend on hydrological and climatic conditions and can vary from one year to another or even from one flood event to another. Robust Cs-Q relationships require long term and high resolution SPM data, in order to capture exceptional flood events and balance temporal variations. Unfortunately, previous works devoted to SPM monitoring in the Rhone River were either long-term studies with low temporal resolution, or studies with high sampling frequency but short timescales. Concentrations of pollutants in SPM are also required for calculating annual pollutant discharges of rivers, but most studies are challenged by the lack of representative data. In this study, our main objectives were the following:

- To estimate Cs-Q relationships, both for the Rhône River and its main tributaries, using long term (2000-2015) and high resolution data in order to predict SPM concentrations for unmonitored periods and to fill gaps in measured time-series,
- To compute annual discharges of SPM over the whole Rhône watershed between Lake Geneva and the Mediterranean Sea, in order to assess the spatial variability of the contributions of the main tributaries,
- To provide new insights in PCB and mercury particulate annual loads based on long term monitoring and time-integrative measurements accounting for baseflow discharges and flood events.

2 MATERIAL AND METHODS



The monitoring network is based on the Rhône Sediment Observatory (OSR) program and includes 7 main sampling stations, covering 75863 km² *i.e.* 79 % of the entire basin. Three stations are located on the Rhône River. The other 4 stations are located on the main tributaries (Saone, Isère, Durance) and on the smaller Gier River, just a few km upstream the confluence with the Rhône River (Figure 1). Water discharge and turbidity have been measured hourly. SPM concentrations were assessed *via* site-specific turbidity-SPM rating curves at all stations except Arles, where SPM concentrations are daily or hourly measured from time-integrative filtered water samples. The monitoring periods extend from 3 years to 11 years depending on the station. Analysis of pollutant concentrations were carried out on SPM collected using particle traps exposed from 15 days to 1 month, or on monthly continuous flow centrifuge samples (Arles station). Depending on the station, the monitoring period for particulate pollutants extends from 2 years to 5 years (between 2011 and 2015).

Figure 1: Location of the 7 permanent monitoring stations of water discharge, suspended particulate matter and pollutants on the Rhône watershed (Rhone Sediment Observatory network)

3 RESULTS AND DISCUSSION

3.1 Annual SPM fluxes

Specific 2-segments Cs-Q relationships were obtained for all stations by using pairs of hourly SPM concentrations and water discharge, and were used to predict SPM concentrations for unmonitored periods and to fill gaps in the measured time-series.

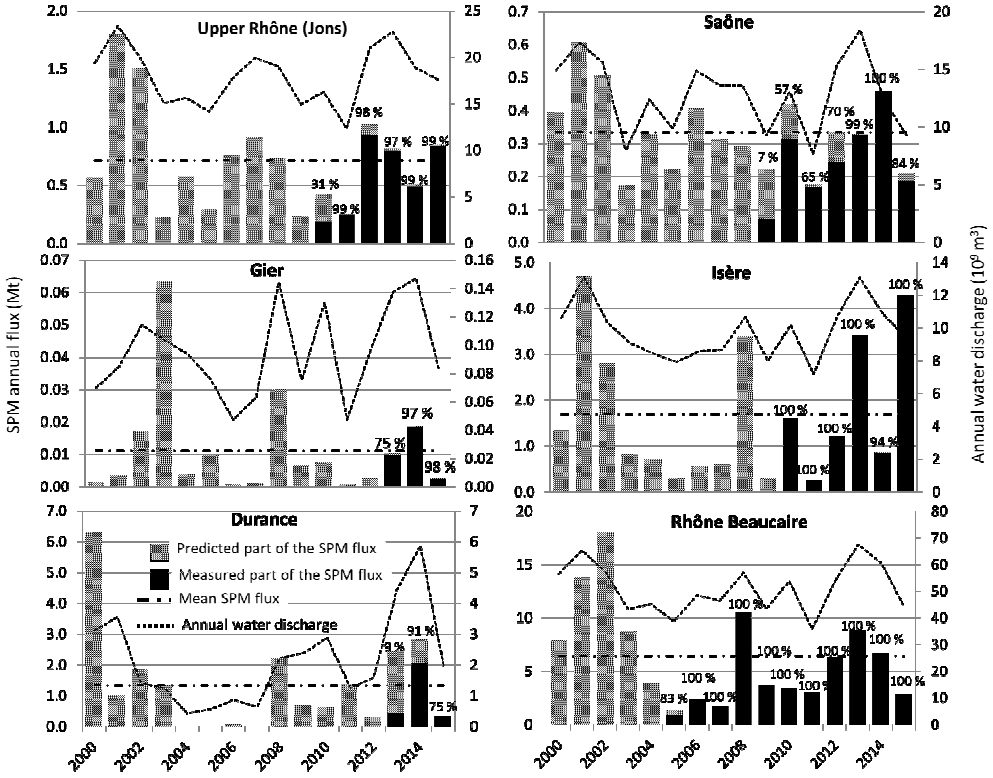


Figure 1 Annual suspended particulate matter masses (bars) transported in the Rhône River and its main tributaries during the 2000-2015 period. The predicted part of the SPM flux is highlighted in grey color. Numbers above the bars stand for the percentage of measured hourly SPM concentrations within a year, as opposed to the predicted ones.

The mean annual SPM flux at the outlet of the Rhône River (Beaucaire station) was estimated to be 6.4 Mt/yr for the 2000-2015 period. The maximum occured in 2002 (18 Mt) and was related to the exceptional floods of October and November (Q=9650 $m^3 s^{-1}$, return period > 40 years). Mean annual contributions of the main tributaries ranged as follow: Isère (26 %), Durance (21 %), Upper Rhône (11 %), Saône (5 %).

3.2 Annual fluxes of mercury and PCB

Concentrations of mercury and PCB 180 exhibited a clear spatial pattern over the Rhone watershed, with values on the Gier and Saone Rivers between 4 (Hg) and 30 fold (PCB 180) higher than on the Durance River, the less contaminated tributary. Over the entire 2000-2015 period, we estimated annual fluxes of particulate Hg and PCB 180 to be about 680 kg/yr and 15 kg/yr at the outlet of the basin, respectively. Among the studied tributaries, the Isère River is the main supplier of particulate pollutants with contributions of 18 % (Hg) and 24 % (PCB 180). For Hg, annual contributions of other tributaries range as follows: Durance (10 %), Saône (9 %), Upper Rhône (6 %) and Gier (< 1%). For PCB 180, contributions range as follows: Upper Rhône (7 %), Saône (4 %), Durance (4 %) and Gier (2 %). This order differs greatly from the one observed for annual solid discharge. Hence, the strong spatial pattern observed over the basin in terms of pollutant levels can lead to a noticeable reordering of the contributions of tributaries to annual fluxes. Annual budgets were not in equilibrium. This could be due to inputs of pollutants from downstream tributaries that were not included in our study, since Mourier *et al.* (2014) found high concentrations of PCBs in sediment cores collected in the Rhône River, downstream of Lyon and upstream of Arles, which were related to local sources.

LIST OF REFERENCES

Mourier, B., M. Desmet, P. C. Van Metre, B. J. Mahler, Y. Perrodin, G. Roux, J.-P. Bedell, I. Lefèvre et M. Babut (2014). "Historical records, sources, and spatial trends of PCBs along the Rhône River (France)." Science of The Total Environment 476–477: 568-576.