Impact of dam flushing operations on sediment dynamics and quality in the upper Rhône River, France

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Abstract: The Rhône River (France) has been used for energy production for decades and 21 dams have been built. To avoid problems due to sediment storage, dam flushing operations are periodically organized. The impacts of such operations on suspended particulate matter (SPM) dynamics (resuspension and fluxes) and quality (physico-chemical characteristics and contamination), were investigated during a flushing operation performed in June 2012 on 3 major dams from the Upper Rhône River. The concentrations of major hydrophobic organic contaminants (polychlorinated biphenyls, polycyclic aromatic hydrocarbons - PAHs, bis(2-ethylhexyl)phthalate [DEHP] and 4-n-nonylphenol), trace metal elements, particulate organic carbon (POC) and particle size distribution were measured on SPM samples collected during this event as well as on those obtained from 2011 to 2016 on a permanent monitoring station (150 km downstream). This allows to compare the SPM and contaminant concentrations and fluxes during the 2012 dam flushing operations with those during flood events and baseflow regime. At equal water discharge, mean SPM concentrations during flushing were on average 6-8 times higher than during flood events recorded from 2011 to 2016. While of short duration (19 days), the flushing operations led to the resuspension of SPM and contributed to a third of the mean annual SPM flux. The SPM contamination was generally lower during flushing than during baseflow or flood, probably due to the fact that flushing transport SPM only issued from resuspended sediment, with no autochtonous particles nor eroded soil. The only exception are PAHs and DEHP with higher concentrations during flushing, which must be issued from the resuspension of legacy-contaminated sediments stored behind the dams before the implementation of emission regulation. During flushing, the variations of POC and contaminant concentrations are also mostly driven by particle size. Finally, we propose a list of recommendations for the design of an adequate monitoring network to evaluate the impact of dam flushing operations on large river systems.

Title page

Impact of dam flushing operations on sediment dynamics and quality in the upper Rhône River, France

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*Cover Letter

Cadarache, 8 November 2019

Dear Editor,

Please find enclosed the revised version of the manuscript entitled "Impact of dam flushing operations on sediment dynamics and quality in the upper Rhône River, France" by H. Lepage and co-authors.

This manuscript was corrected according to the reviewers recommendations. Answers to their remarks were compiled in a document enclosed with the manuscript. Finally, the total length of the article was also reduced by 10% (from 6752 words to 6078 words excluding references).

Kindly acknowledge the receipt of the same.

Yours sincerely,

On behalf of the authors,

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*Response to Reviewers

Comment from reviewers	Answer from corresponding author	Correction in the manuscript
Reviewer 1		
Highlights	corrected	Sediment dynamic differed during flushing operations and flood events
Line 15	corrected	The Rhône River (France) has been used for energy production for decades and 21 dams have been built.
Line 29 – In general it is not advisable to start a sentence with an acronym, rather right it out in full	corrected	The SPM contamination was generally lower than during baseflow and flood regimes.
Line 131	corrected	More than 3 x 10^6 m^3 of SPM were thus stored in the Verbois reservoir between 2003 and 2012 (Services industriels de Genève, 2014).
Line 133	corrected	[...] which transports ~0.7 x 10^6 m^3 of SPM per year (approx. 1 to 3 tons dry weight) [...]
Line 309-314 – This section reads as introduction or as part of the discussion	According to reviewer 4, this section was moved in the Materials and Method part.	**2.7. SPM and contaminant fluxes calculation** One important question regarding the impact of flushing operations is to determine their relative contribution to SPM transport compared to flood events. Such a comparison requires an estimation of the influence of both types of events over a longer time scale. The Jons station allows an estimation of the annual SPM fluxes from 2011 to 2016 (based on hydrological years, i.e. from September to August).
Reviewer 4		
[...] the manuscript need a deep re-organization in order to better address the main aim. In particular, both methodological and research aspects are mixed in the text, and this creates some confusion in the reader. For example, to my opinion comparison between results obtained	According to reviewer 1, the paper is well written and logically organized. However, the comparison between CFC and PT was indeed confusing and a part of this was moved to the supplementary materials (Supplementary Material #1.1)	

with CFC and PT sampling methods is redundant and may be moved to Supplementary material. As well, the final recommendations for an adequate monitoring network of flushing operations doesn't add any relevant information to the previous text.	Regarding the recommendations, we thought that it is interesting to share our feedback on flushing operations monitoring as we did not find any similar study in the literature with a long term monitoring. We also noticed that recommendations are used in scientific papers, even in Journal of Environmental Management (ex : https://doi.org/10.1016/j.jenvman.2019.06.092 https://doi.org/10.1016/j.jenvman.2015.08.014 https://doi.org/10.1016/j.jenvman.2019.109405 Since the reviewer 1 did not complain on that point and since specific areas of interest of JEM includes: "Development of methods for environmental quality management (new procedures, characterization techniques, **monitoring methods**)", we really think that it is relevant to keep such recommendations in the paper.	
By contrary, some scientific aspects should be more deepened and explained. In the present form, results are mainly presented as a report regarding this particular case-study. The case-study of the Rhone River has been widely studied before in many aspects, as proved by the publications reported in the reference section (e.g. Peter et al. 2014 regards the same flushing event, with samples collected in different sampling stations upstream Jons). Thus, results may be shortened, while in the discussion section a wider generalization and comparison with other	The Rhône River has been indeed studied for decades by many research fields, including sedimentary aspect. However, studies investigating the impact of flushing operations on SPM quality remain rare contrary to what is said by reviewer 4. Furthermore, studies on particulate contaminant behaviors during flushing operations are also very rare at a worldwide scale, as described in the introduction of our paper. Reviewer 4 indicates that Peter et al. 2014 worked on the same event. This is right (and we cite this work in our manuscript) but their approach was	

published cases should be added.	really different with a specific focus on the impact of the flushing operations on benthic invertebrates. Also, their investigation on the water quality was complementary to our study as they conducted measurements of dissolved and particulate metals (Al, Co, Cr, Cu, Fe, Mn, Ni, Pb) while we measured particulate organic contaminants (PCBs, PAHs, DEHP and 4-n-nonylphenols) in addition to particulate metals (Cd, Cu, Pb, Zn, Hg). Finally, the impact of the flushing operations on SPM and associated contaminant over a long term monitoring was not studied by Peter et al. 2014. Therefore, we think that both studies are complementary.	
Moreover, a better explanation of the different behavior of contaminants may be addressed.	Additional information were added to the manuscript according to the comments below (chapter 4.2 and 4.3).	
Abstract: methods are missing. Line 29: what do you mean with "origin"? Lines 30-31 seem to contradict lines 29. Maybe this part should be re-written.	The abstract was entirely rewritten to fit these comments.	
the English text should be revised (e.g. highlights contain some mistakes)	The manuscript was proof-corrected by a native English (certificate enclosed in the built pdf) and Reviewer 1 said that the paper was well written. However, special attention was given in re-writing the manuscript.	
line 84: "fixed" doesn't seem correct: maybe "adsorbed" or "carried by"	This sentence was mixed with another to shorten the manuscript.	Additionally, we still need a better understanding of contaminants remobilization and transport processes under such conditions (Hauer et al., 2018; SedNet, 2014), as floods and dam flushing operations are major events able to transport a large fraction of contaminant fluxes (Poulier et al.,

		2019).
In Figure 1 many other smaller dams are present between Genissiat dam and the sampling station at Jons: do they have some influence on your results (e.g. flux calculations or contaminant concentrations?) Are there contaminated sediments stored in those smaller reservoirs which may be remobilized during the flushing events?	The text was corrected in order to include the specific management of these dams. Unfortunately we have no data on contamination levels in these smaller reservoirs.	Five small dams located between Génissiat and Jons (Figure 1) were also opened during this period and managed in order to prevent both sediment deposition and resuspension. The by-passed sections of the Rhône River (Old Rhône reaches) were disconnected and the whole SPM flux transiting through the reservoirs and tailrace canals. The levels of the reservoirs were lowered as much as possible to prevent deposition and ensure a quick transfer of water and SPM.
lines 100-114: this part seems a summary of the research: the final aim of the research should be more addressed	Additional information was added to clarify the aim of the research.	This study aimed at characterizing the impact of flushing operations on SPM dynamics and quality in a large river system: the Rhône River basin. Thanks to a specific monitoring, the fluxes and mass balances of SPM and associated contaminants triggered by flushing operations in 2012 and 2016 were estimated at different time scales. The variations in contaminant concentrations were related to the characteristics of SPM (particle size distribution, organic carbon content) as well as to their origin in the watershed (eroded soil versus resuspended sediment).
line 119: 95600 km2 is the watershed area?	Corrected.	The Rhône River (95 600 km^2 watershed area, mean water discharge of ~1700 $m^3.s^{-1}$ at the outlet station of Beaucaire)
lines 152-153: add "2016" in the dates	Corrected	(from May 20[th], 2016 at 12:00 to May 31[st], 2016 at 12:00).
lines 155-158: some results are based on water discharge values: maybe a short description of this hydrodynamical model performance/validation should be mentioned	Additionnal information was added	There is no hydrometric station close to Jons and hourly water discharge was calculated using the 1-D hydrodynamical model MAGE (Irstea, France) and discharge inputs from upstream hydrometric stations (Lagnieu), Bourbre (Tignieu-Jameyzieu),

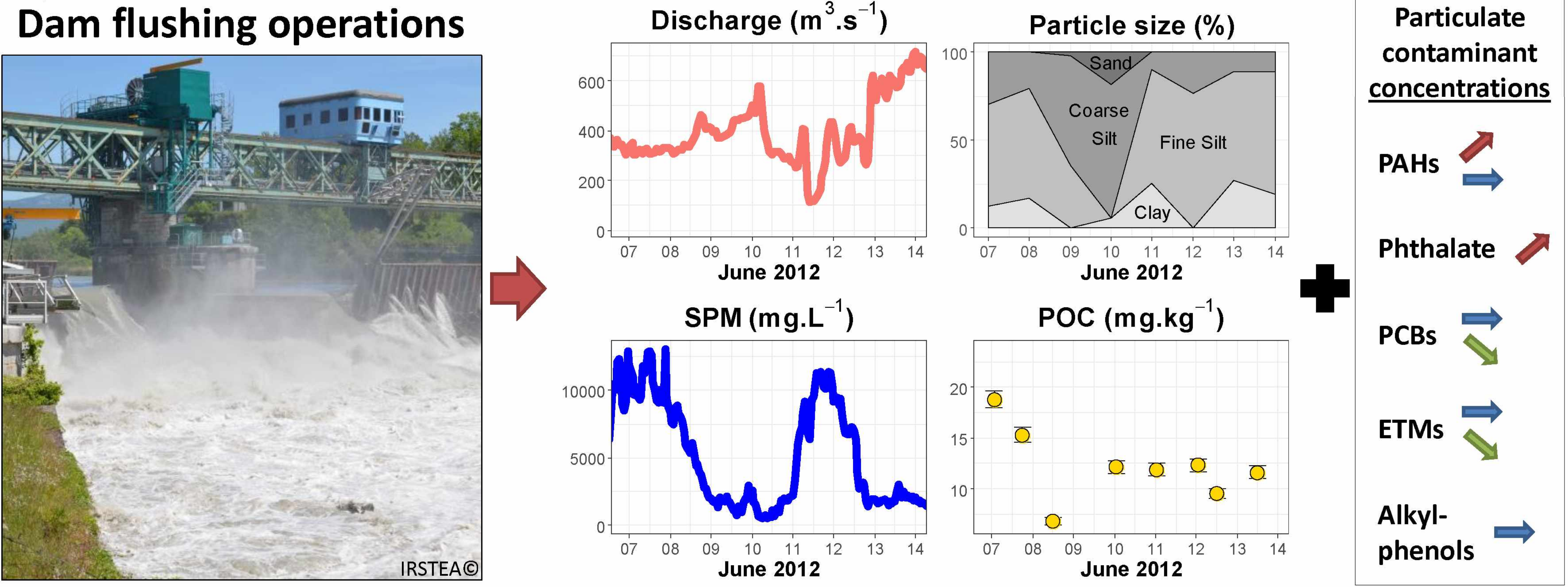
		and Ain (Port-Galland) Rivers (Figure 1) (Dugué et al., 2015; Launay et al., 2015). This model is calibrated for the whole Rhône River and outputs were evaluated against measured water levels, with a maximal accepted difference of 10 cm.
lines 205-206 should be moved to discussion or supplementary material	Part 2.4 including lines 205-206 was moved to supplementary materials to reduce manuscript length.	
- A "Data analysis" paragraph is missing. Maybe lines 309-314 could be part of this paragraph. As well, the calculation of fluxes should be reported	Lines 309-314 were moved to the Materials and Method part (chapter 2.7). Flux calculation was already explained in this chapter.	
lines 292-308 are partly a repetition of paragraph 3.3, so this part can be moved to paragraph 3.3 and shortened	Lines 292 to 308 describe the variation of the SPM parameters (POC, particles size distribution) and contaminants concentrations during the flushing operations, while part 3.3 describe the variation between the different hydrological conditions (flood+baseflow vs flushing). There is thus no repetition and we propose to keep both parts separately since reviewer 1 noted that the paper is "logically organized".	
lines 301-302: how do you explain this contrasting behavior?	This sentence in the "results" section was written to describe the observations detailed in the following lines. The explanation is given in the discussion section. However the sentence was deleted to clarify the paragraph.	
lines 370-374: how do you explain a different behavior of congeners of organic compounds (PAHs, PBCs)?	PAHs molecules and PCBs congeners are indeed two families of organic compounds, and it is well know that their sources are different (e.g. for PAHs: https://doi.org/10.1002/etc.5620160212). In the Rhône River, we highlighted such differences in a report of the OSR programm and concluded that the different congeners of PAHs have different origins, mainly road traffic and	For the different behavior of the PAHs congeners (Table S4), preliminary results on PAHs concentration in SPM collected in the upper Rhône River demonstrate that the different congeners of PAHs have different origins, mainly road traffic and domestic heating (Botha et al., 2014, Poulier et al., 2018).

	domestic heating. Moreover, the absence of relation between PAHs and PCBs demonstrate that PCBs mainly result from industrial releases. http://www.graie.org/osr/IMG/pdf/2018.01_livrable_iv2_vdiffusable_mc_gp_drm.pdf Furthermore, the explanation about these differences is given in the "discussion" section at chapters 4.2 and 4.3 but more details were added to the manuscript.	
Discussion should not report citations of figures. Sub-titles in discussion are not needed	References to the figures in the discussion should help the reader to find which part of the results is related to this discussion. Also, sub-titles presented as questions were proposed in order to clarify the discussion. We are surprised by this reviewer comment, because almost all papers refer to figures in the discussion when necessary (see for examples: : https://doi.org/10.1016/j.jenvman.2019.109479 or https://doi.org/10.1016/j.jenvman.2019.109391 or https://doi.org/10.1016/j.jenvman.2019.109405). Based on the comment of reviewer 1 ("the paper is logically organized. I found hard to criticize the different sections"), we prefer to keep these citations and sub-titles.	
line 412: the day is missing in the date: "06/0/2012"?	Corrected	[...]from 06/11-14/2012 [...]
lines 419-422: any idea of contamination levels in sediments stored in the reservoirs?	Unfortunately investigations on contamination levels in the studied reservoirs are very sparse.	

	Concentration of TME, PAHs and PCBs measured on sediment cores collected in the studied dams were previously reported by the Forel Institute in 2009. However, each sediment cores were fully mixed and only one measure of contaminants were conducted on each mixed sediment cores. Therefore as our results demonstrate that the particle size affect the concentrations as well as the deepness of the sediment flushed, it is difficult to compare the results of the mixed sediment cores and our samples of SPM.	
lines 424 and 431: what do you mean with "origin"?	The meaning is explained in chapter 4.2 and 4.3. Origin means the source of particles that include soil of the catchments, sediments of the different tributaries, or anthropic sources (for example road traffic and domestic heating for PAHs).	
Paragraph 5: these are conclusions	We do not agree with this comment. Indeed, this section presents recommendations for scientists or stakeholders to improve monitoring of the consequences of flushing events in fluvial systems. These recommendations underline specifically how the monitoring of such events should differ from the one usually performed (e.g. regulatory monitoring) and could be implemented for all types of rivers.	
lines 482-458: this part of text is redundant. In particular: lines 482-521 could be moved to supplementary material, lines 522-548 could be deleted as they summarize results	We partly disagree with this comment. We rely on the reviewer1 comments to support this position. However, we shortened the text to avoid redundancy as much as possible.	The full manuscript was shortened as much as possible and some sections have also been moved to a new supplementary material section.
lines 496-500: also a pressure analysis in the watershed could drive the choice of contaminants of interest	Right. We add this point in the text.	Selection criteria include: priority pollutants according to regulations, performance of the analytical methods (limit of quantification, uncertainty) and emergent pollution, watershed

		pressure;
Fig. 1: the city of Geneva is missing in the map	Corrected	
Figure 5: maybe both events should be plotted in the same graph (even if some points will be hidden behind the others): in the present form black points represent both normal conditions and one flushing event, and this creates some confusion	Corrected	
Tab. 1: why is the number of samples (n) different in each column of the same row?	This table was moved to the supplementary materials (Table S4) to shorten the manuscript. The number of samples was corrected and is now the same for each row of particle size.	

Graphical Abstracts



Highlights

- Dam flushing operations were monitored on the Upper Rhône River (France)
- Sediment dynamic differed during flushing operations and flood events
- About 0.6 Mt of SPM were stored in the Upper Rhône from 2011 to 2016
- 21 to 37% of the mean annual SPM flux transited during the 2012 flushing operations
- Particulate contaminant concentrations were driven by particle size and SPM origin

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Abstract

The Rhône River (France) has been used for energy production for decades and 21 dams have been built. To avoid problems due to sediment storage, dam flushing operations are periodically organized. The impacts of such operations on suspended particulate matter (SPM) dynamics (resuspension and fluxes) and quality (physico-chemical characteristics and contamination), were investigated during a flushing operation performed in June 2012 on 3 major dams from the Upper Rhône River. The concentrations of major hydrophobic organic contaminants (polychlorinated biphenyls, polycyclic aromatic hydrocarbons - PAHs, bis(2-

ethylhexyl)phthalate [DEHP] and 4-n-nonylphenol), trace metal elements, particulate organic carbon (POC) and particle size distribution were measured on SPM samples collected during this event as well as on those obtained from 2011 to 2016 on a permanent monitoring station (150 km downstream). This allows to compare the SPM and contaminant concentrations and fluxes during the 2012 dam flushing operations with those during flood events and baseflow regime. At equal water discharge, mean SPM concentrations during flushing were on average 6-8 times higher than during flood events recorded from 2011 to 2016. While of short duration (19 days), the flushing operations led to the resuspension of SPM and contributed to a third of the mean annual SPM flux. The SPM contamination was generally lower during flushing than during baseflow or flood, probably due to the fact that flushing transport SPM only issued from resuspended sediment, with no autochtonous particles nor eroded soil. The only exception are PAHs and DEHP with higher concentrations during flushing, which must be issued from the resuspension of legacy-contaminated sediments stored behind the dams before the implementation of emission regulation. During flushing, the variations of POC and contaminant concentrations are also mostly driven by particle size. Finally, we propose a list of recommendations for the design of an adequate monitoring network to evaluate the impact of dam flushing operations on large river systems.

Key words

Trace metals, PAHs, suspended particulate matters, pollution monitoring, sediment flux

1. Introduction

Investigating suspended particulate matter (SPM) transport in river catchments is a key component of the environmental monitoring of large rivers as SPM may cause important environmental impacts (Frings and Ten Brinke, 2018; Le Bissonnais *et al.,* 2005; Walling, 2005). Suspended particulate matter transport can result in increasing water turbidity, degradation of fish habitat (Grimardias *et al.*, 2017), siltation of reservoirs (Owens *et al.,* 2005), and transport of contaminants such as polychlorinated biphenyls (PCBs), trace metal elements (TME), radionuclides or nutrients (Dumas *et al.,* 2015; Evrard *et al.,* 2015; Horowitz, 2008; Taylor and Owens, 2009). The monitoring of SPM and associated particulate contaminant fluxes in rivers is therefore necessary, though practically challenging.

One of the challenges is to evaluate the role of dam reservoirs in terms of both sink and source of SPM (Hauer *et al.*, 2018). Indeed, dams are among the main structures that impact SPM transport by reducing flow velocity and increasing sediment deposition (Syvitski *et al.,* 2005). Such storage leads to a decrease in reservoir capacity and triggers economic issues (White, 2001), hence several methods are used to limit reservoir filling (Kondolf *et al.,* 2014). Dam flushing operations are regularly organized to remove sediments stored in reservoirs and avoid problematic consequences such as siltation and enhanced flood hazard (Brown, 1944; Peteuil *et al.,* 2013). These operations consist in increasing water velocity and shear stress by lowering the water level to erode material deposited in the reservoir (Brandt, 2000; Di Silvio, 2001; Kondolf *et al.,* 2014). However, flushing operations can have significant ecological impacts: increased stress or mechanical damage of organisms, loss of habitats, and decrease in dissolved oxygen concentrations due to increased SPM concentrations (Espa *et al.*, 2016; Grimardias *et al.*, 2017; Hauer *et al.*, 2018; Kemp *et al.*, 2011). Besides these direct consequences, contaminants associated with deposited sediments (and legacy contamination) may also be resuspended and disseminated (Peter *et al.,* 2014) or transferred to the dissolved phase (Bretier *et al.*, 2019; Kalnejais *et al.*, 2010), increasing their bioavailability (e.g. Dong *et al.,* 2018 for TME). On the contrary, resuspension of less contaminated sediments may result in the dilution of particulate contamination (Ferrand *et al.,* 2012). While difficult to evaluate, especially in large river systems, these impacts should however be better addressed in order to minimize the costs under consideration of ecological requirements and to improve sediment management in terms of hydropower use (Hauer *et al.*, 2018).

The Rhône River is France's number one river in terms of water discharge and hydroelectricity production. Downstream of Lake Geneva in Switzerland, the Rhône River is highly artificialized and characterized by the presence of 21 hydropower plants. A large proportion of SPM is stored each year in this river, mostly in dam reservoirs (Bravard and Clémens, 2008; Provansal *et al.*, 2014). As several industries are implemented between Lake Geneva and the Rhône River Delta, a large range of contaminants is delivered to the river (e.g., Ollivier *et al.,* 2011) and a fraction is also trapped in dam reservoirs. Dam flushing operations are regularly conducted on the upper Rhône River to prevent bed aggradation in the Verbois reservoir (Figure 1), and thus urban flooding in the city of Geneva (Peteuil *et al.,* 2013). From 1945 to 2003, flushing operations were performed every 3 years and the quantity of SPM exported was similar to the quantity of sediment stored since the previous flush (Diouf, 2013). Due to the associated increase of turbidity and potential consequences on fish populations (Grimardias *et al.*, 2017), such operations are now strictly monitored in order to reduce their environmental impact. However, the survey of SPM transport on a large scale remains difficult and the impact of such operations compared to flood events is still a matter of debate due to the lack of data. Additionally, we still need a better understanding of contaminants remobilization and transport processes under such conditions (Hauer *et al.*, 2018; SedNet, 2014), as floods and dam flushing operations are major events able to transport a large fraction of contaminant fluxes (Poulier *et al.*, 2019). The behavior and dynamics of sediment (and associated contaminants) and dissolved/labile metals have already been studied on the Rhône River during floods and flushing operations, respectively (Antonelli *et al.*, 2008; Ollivier *et al.*, 2011; Sicre *et al.*, 2008; Bretier *et al.*, 2019; Peter *et al.*, 2014). However, there is a lack of information regarding the impact of flushing operations on particulate contaminants.

This study aimed at characterizing the impact of flushing operations on SPM dynamics and quality in a large river system: the Rhône River basin. Thanks to a specific monitoring, the fluxes and mass balances of SPM and associated contaminants triggered by flushing operations in 2012 and 2016 were estimated at different time scales. The variations in contaminant concentrations were related to the characteristics of SPM (particle size distribution, organic carbon content) as well as to their origin in the watershed (eroded soil versus resuspended sediment). Finally, the SPM monitoring performed during a second flushing operation in 2016 is also used to compare and evaluate the role of such events on a pluriannual scale.

2. Materials and Method

2.1. Location of the dams and description of the dam flushing operations

The Rhône River (95 600 km² watershed area, mean water discharge of ~1700 $m^3.s^{-1}$ at the outlet station of Beaucaire) constitutes the main SPM input to the Western Mediterranean Sea, with a mean SPM flux of 6.5 $Mt.year^{-1}$ from 2000 to 2015 according to Poulier *et al.* (2019). The Rhône Sediment Observatory operates two permanent monitoring stations along this river and several others on its main tributaries (Poulier *et al.*, 2019; Thollet *et al.*, 2018). We used data collected at the permanent station of Jons (45.8121N, 5.0896E – see Fig. 1), near the city of Lyon, to investigate the dynamics of sediments in the upper Rhône River and notably the impact of dam flushing operations conducted downstream of Lake Geneva.

This study focuses on flushing operations conducted in June 2012 in three reservoirs located downstream of Lake Geneva (Figure 1): Verbois, Chancy-Pougny, and Génissiat. Flushing operations used to be performed in this area every 3 years from 1945 to 2003, but the Swiss authorities later decided to consider alternative methods to manage SPM and dams. More than 3 x 10^6 m^3 of SPM were thus stored in the Verbois reservoir between 2003 and 2012 (Services industriels de Genève, 2014). They mainly originated from the Arve River, a tributary located upstream of the dam (Figure 1), which transports ~0.7 x 10^6 m^3 of SPM per year (approx. 1 to 3 tons dry weight), half of this amount being stored behind the dam (Guertault *et al.*, 2014; Launay *et al.*, 2019). This accumulation induced a change in the water level of the reservoir during floods that could impact the security of local population. Since no alternative management solutions were validated, the Swiss and French authorities agreed to conduct a new flushing operation in June 2012 on the three successive dams (Figure 1). The flushing operations were conducted by lowering the water level following the drawdown flushing method (Fruchard and Camenen, 2012) in order to remove stored SPM in the three dams (Figure 2). The large-volume Génissiat reservoir located downstream of the two other dams (Figure 1) was flushed first (4-12 June) in order to compensate for the deposition of the SPM released from the Chancy-Pougny (9-16 June) and Verbois dams (9-22 June, Figure 2; Diouf, 2013). Five small dams located between Génissiat and Jons (Figure 1) were also opened during this period and managed in order to prevent both sediment deposition and resuspension. The by-passed sections of the Rhône River (Old Rhône reaches) were disconnected and the whole SPM flux transiting through the reservoirs and tailrace canals. The levels of the reservoirs were lowered as much as possible to prevent deposition and ensure a quick transfer of water and SPM. It should also be noted that floods occurred in two upstream tributaries, Ain and Fier, during the 2012 flushing operations.

Another flushing operation, also monitored at Jons, was conducted on the same dams in May 2016 (Diouf, 2017). Compared to 2012, the 2016 flushing operation was only partial, with a smaller decrease of the water level in Verbois reservoir: 361 m a.s.l. in 2016 vs 352 m a.s.l. in 2012 (Diouf, 2017). Contrary to 2012, the 3 dams were flushed at the same time and during 11 days (from May 20th, 2016 at 12:00 to May 31st, 2016 at 12:00).

2.2. Water discharge

There is no hydrometric station close to Jons and hourly water discharge was calculated using the 1-D hydrodynamical model MAGE (Irstea, France) and discharge inputs from upstream hydrometric stations (Lagnieu), Bourbre (Tignieu-Jameyzieu), and Ain (Port-Galland) Rivers (Figure 1) (Dugué *et al.*, 2015; Launay *et al.*, 2015). This model is calibrated for the whole Rhône River and outputs were evaluated against measured water levels, with a maximal accepted difference of 10 cm.

2.3. Concentration of SPM

To evaluate the SPM concentration at Jons, turbidity measurements were conducted continuously every 15 min using a Hach Lange SC200 turbidity probe. The relation between turbidity (in NTU) and SPM concentration was calibrated as follows: SPM concentration was measured in water samples collected during various hydrological conditions (baseflow, flood, and flushing operations) in order to cover the entire range of discharges. Those water samples were collected manually or using a portable automatic water sampler (ISCO or SIGMA – 1 sample every 4 hours) due to logistic constrains. Suspended particulate matter concentration was measured after filtration of the samples through pre-weighed filters (Whatman GF/F, 0.7 µm). The limit of quantification (LQ) was 2 mg.L^{-1}.

2.4. SPM sampling for chemical analyses

In order to collect a sufficient amount of SPM material for subsequent physicochemical analyses, two sampling methods were used at Jons, Continuous Flow Centrifugation (CFC) and Particle Trap (PT) (Masson *et al.* 2018), as described in details in the supplementary materials (Supplementary Material #1.1). The CFC samples were used to characterize the concentration of TME (Cd, Cu, Pb, Zn), Hg and PCBs. The PT was used to collect a sufficient amount of SPM for chemical analysis of polycyclic aromatic hydrocarbons (PAHs), bis(2-ethylhexyl)phthalate (DEHP) and 4-n-nonylphenol. During the 2012 flushing operations, samples were collected every day from June 4 to June 16 with the CFC and during 2-3 day periods from June 5 to June 16 with the PT (supplementary material #2 Table S1).

At the Jons station, the end of the pumping pipe is installed approximately 5 m downstream of the PT in order to side step potential heterogeneity of SPM throughout the river cross-section. Prior to chemical analysis, SPM collected with the two sampling techniques were transferred to clean brown glass bottles (250 mL), deep-frozen (-18°C), freeze-dried, and finally homogenized by grinding in an agate mortar. SPM samples were stored in the dark and at ambient temperature before analysis.

2.5.Grain-size distribution and particulate organic carbon analysis

Volumetric grain size distribution was analyzed on a CILAS 1190 laser particle size analyzer (range of values: 1 – 2 500 µm) according to the ISO 13320 standard method (AFNOR, 2009). Recent investigation on Particle Size Distributions (PSD) in the Rhône River concluded that the usual percentiles d10, d50 or d90 might not be sufficient to describe grain size due to a multimodal shape (Masson *et al.,* 2018). This shape can be modeled as a combination of several homogeneous subpopulations of mixed particles, each following a log-normal distribution. Therefore, each subpopulation was modeled using the R software (version R3.2.0) to extract the mass proportion of each subpopulation, which were used to compare the different PSD. Subpopulations were clustered in five classes according to their modal diameter: < 4 µm (clay), 4-15 µm (fine silt), 15 – 63 µm (coarse silt), 63 - 125 µm (very fine sand) and > 125 µm (sand).

The determination of particulate organic carbon (POC) in SPM samples was performed using a carbon analyzer (Thermo Electron, CHN Flash 2000) at the INRA laboratory (Arras, France). Decarbonatation was performed using hydrochloric acid according to the NF ISO 10694 standard method (AFNOR, 1995). The LQ was estimated to be 50 $mg.kg^{-1}$ by using a reference material (Aglae, 15 M9.1; 40 $g.kg^{-1}$) and the analytical uncertainty varied between ~3% and ~6% (k=2), depending on the POC concentration.

2.6. Chemical analysis of SPM

Concentrations of total Cd, Cu, Pb, and Zn were determined after a microwave acid digestion of the SPM as described in the supplementary materials (Supplementary Material #1.2). The LQ was 0.05 $\mu g.kg^{-1}$. The mean difference between measured and certified values were, for STSD-3 (n=6) and MESS-4 (n=3), respectively: 20 and 3% for Pb; 7 and 6% for Cu; 5 and 2% for Zn; 14 and 7% for Cd.

The determination of total Hg in SPM was performed using an automated atomic absorption spectrophotometer, DMA 80 (Milestone), according to EPA method 7473 (US EPA, 2007). The LQ was 10 $\mu g.kg^{-1}$. Blanks and certified reference materials (IAEA 433, marine sediment; LGC 6187, river sediment) were systematically used to check analytical accuracy (94%) and uncertainty (14%; k=2).

Indicator PCBs (PCB 28, 52, 101, 118, 138, 153, and 180) were analyzed using capillary gas chromatography coupled to an electron capture detector (GC-ECD), as detailed in Masson *et al.* (2018). Limits of quantification ranged from 0.5 to 1 $\mu g.kg^{-1}$. Only results for compounds with a quantification frequency higher than 60% (Helsel, 2006) were considered here, i.e. PCBs 101, 138, 153, and 180.

The 16 priority PAHs (Keith, 2015) were analyzed by the Laboratory of Hygiene and Environment (Rouen, France), using capillary gas chromatography coupled to a mass spectrometer (GC-MS) and according to the XP X33-012 Standard (AFNOR, 2000). Limits of quantification ranged from 1 to 2 $\mu g.kg^{-1}$ for most of the PAHs, except for dibenz[a,h]anthracene and fluorene (up to 5 µg.kg^{-1}) and acenaphtylene (up to 20 µg.kg^{-1}). In this paper, the 16 PAHs are presented and discussed as their quantification frequency was higher than 60%.

Alkylphenols (4-n-nonylphenol, octylphenol, para-tert-octylphenol, tert-butylphenol) and DEHP were analyzed by the La Drôme laboratory (Valence, France), by GC-MS. Limits of quantification were 10 µg.kg^{-1} for alkylphenols and 100 µg.kg^{-1} for DEHP. In this paper, only substances with a quantification frequency higher than 60% are presented, i.e 4-n-nonylphenol and DEHP. Analytical uncertainties of these organic contaminants were estimated at 60% (k=2) for concentrations lower than 3-times the LQ and 30% (k=2) for concentrations higher than 3-times the LQ. For concentrations higher than 3-times the LQ, this value was confirmed by interlaboratory trials (Charpentier, 2016).

2.7. SPM and contaminant fluxes calculation

One important question regarding the impact of flushing operations is to determine their relative contribution to SPM transport compared to flood events. Such a comparison requires an estimation of the influence of both types of events over a longer time scale. The Jons station allows an estimation of the annual SPM fluxes from 2011 to 2016 (based on hydrological years, i.e. from September to August). The SPM fluxes were calculated from the mean hourly discharge and SPM concentration time series and cumulated over periods of flood events, flushing operations, or years (Poulier *et al.*, 2019). Contaminant fluxes were calculated by multiplying the hourly SPM flux by the particulate contaminant concentrations. Station-specific median particulate contaminant concentrations were used for the non-monitored periods to fill gaps in the measured time series (Poulier *et al.*, 2019). The BDOH tool was used to do the calculation of SPM and contaminant fluxes (Branger *et al.*, 2014), and data are available online (Thollet *et al.*, 2018).

2.8. Classification according to hydrological conditions

In order to investigate the impact of flushing operations, results were compared to baseflow and flood conditions using data collected at Jons from 2011 to 2016. Samples classified as "flushing period" refer to those collected between 06/04/2012 00:00 and 06/22/2012 15:00. Samples were classified as "flood samples" if the corresponding discharge was higher than a 800 $m^3.s^{-1}$ discharge threshold (i.e., half the 2-year flood discharge (Launay, 2014)). For time-integrative samples collected with a PT, classification was based on the SPM proportion related to the different hydrological states. For each PT sample, the corresponding SPM flux was calculated using turbidity values for the entire period of collection (total SPM flux) and for the period with water discharge higher than the flood threshold (SPM flux during flood). The PT samples were classified as flood samples if the SPM flux during the flood event represented more than 50% of the total SPM flux. The other samples were classified as "baseflow samples".

3. Results

3.1. Variation of water discharge, SPM concentration and quality during flushing operations

At Jons, the discharge ranged from 343 to 1600 $m^3.s^{-1}$ (maximum reached on June 14) during the 2012 flushing operations, while SPM concentration ranged from 10 to 905 $mg.L^{-1}$ (maximum reached on June 13) (Figure 3). The POC concentration was 13.5 $\pm$ 3.8 $mg.kg^{-1}$

(mean ± standard deviation) but varied depending on the sampling method (see supplementary material #2 Table S1).

The PSD were mostly characterized by silt (fine silt (from 30 to 82%), coarse silt (5%-70%)) and clay (0-34%, supplementary material #2 Table S1) depending on the sampling method (see supplementary materials #1.3). Sand particles were only observed in two samples and the proportion never exceeded 26% (supplementary material #2 Table S1).

The TME concentrations remained fairly stable during all the flushing period, while a decrease of the levels of Hg and PCBs was observed in the last collected sample (Figure 4 – supplementary material #2 Table S2). For all the PAHs and 4-n-nonylphenol, the concentration of the first samples (collected from June 7 to 11) were at least 1.2 times higher (up to 6 times) than the two other samples (collected from June 11 to 16) (Figure 4 – supplementary material #2 Table S3). For DEHP, the only clear trend is a lower value (by a factor ten) found in the first sample.

3.2. Specificity of SPM characteristics during flushing events

3.2.1. Water discharge and SPM concentration

The water discharge measured at Jons during the 2012 flushing operations was similar to that of several floods occuring that year (Figure 3). Peaks of SPM concentration were concomitant to those of water discharge (June 13th and 14th), and the highest one during the flushing operation was almost 25% higher than during flood events (Figure 3). Due to sediment resuspension in reservoirs, for equivalent water discharge at Jons, mean SPM concentrations during the flushing operations of June 2012 were 8 times higher than during floods (Figure 5). In 2016, the discharge was lower than in 2012 (Figure 5) due to a different management of the flushing operations (Diouf, 2017). However, the mean SPM concentration was 6 times higher than during flood events at equivalent water discharge.

3.2.2. SPM Fluxes

From 2011 to 2016, the annual SPM fluxes ("output" fluxes) at Jons remained fairly constant with values ranging from 0.71 to 0.96 $Mt.y^{-1}$. Flood events, representing 16-37% of the year duration, contributed to the major part of annual SPM fluxes and transported 48-89% of the annual flux (Figure 6). While representing only 3-5% of the year (Figure 6), the contributions of 2012 and 2016 flushing operations represented 37% and 23% of the total annual SPM flux, respectively. However, the contribution in 2012 might have been overestimated due to a flood event occurring in two upstream tributaries (Ain and Fier) during the flushing operation of Verbois dam. Thus, the contribution of the 2012 event should range from 0.15 to 0.26 Mt (21-37% of the annual flux) (Launay, 2014). Lastly, SPM fluxes during baseflow conditions remained quite constant over the years and contributed to less than 20% of the total annual fluxes.

The water discharges and SPM concentrations measured at other monitoring stations located upstream, on the main tributaries, were used to estimate the "input" SPM fluxes at Jons (Poulier *et al.*, 2019) (Figure 7). The two hydrological years with flushing operations (2012 and 2016) were characterized by higher output SPM fluxes compared to estimated input ones, with an export of about 0.1-0.2 Mt. In contrast, sediment storage of approximately 0.2 $Mt.y^{-1}$ was observed during years without flushing operations and despite the occurrence of flood events (outputs < inputs). From September 2012 to August 2016 (i.e. the period starting from the end of a flushing operation to the end of the next one), about 0.6 Mt of SPM were thus stored in the Upper Rhône according to this estimation.

3.3. Relative contribution of flushing events to SPM and contaminant concentrations and fluxes

Contaminant concentrations, PSD and POC of samples collected during the 2012 flushing operations were compared to 2011-2016 data during other hydrological conditions (baseflow and flood) (Thollet *et al.*, 2018). Regardless of the sampling period or sampling method used, SPM were mostly silt sized with a non-negligible proportion of clay (Table S4). Presence of sand remained very sparse. The POC concentrations were almost twice lower in samples collected during flushing operations than for the other periods ($p < 0.01$, Table S4).

Different trends were observed for the contaminants of interest. Concentrations were not statistically different during flushing operations, baseflow, and flood conditions for Cd ,4-n-nonylphenol (Figure 8), PCB118 and PCB153 (supplementary material #2 Table S5). For the other TME (Cu, Pb, Zn) and for PCB101 and PCB138, concentrations during flushing operations were significantly lower ($p < 0.05$) than under baseflow regime but similar to values measured during flood events (Figure 8, Table S5). For Hg, the mean concentration during flushing operations (39 ± 6 $\mu g.kg^{-1}$) was significantly lower than for other hydrological conditions (58 ± 15 $\mu g.kg^{-1}$ for baseflow and 50 ± 12 $\mu g.kg^{-1}$ for floods). PCB180 exhibited a similar behavior (Table S5). In contrast, SPM sampled during flushing operations were more contaminated by PAHs and DEHP than SPM sampled during baseflow or flood conditions (Figure 8). Due to a low number of samples (n=3-4), this increase was only significant ($p < 0.05$) for some of the PAHs including fluorene (Figure 8, Table S5).

The fluxes of particulate contaminants during the June 2012 flushing operations were estimated based on discharge and SPM concentrations (Table 1). For all contaminants, the flushing operations transported about one third of the total annual flux.

4. Discussion

4.1. *Why are SPM concentrations higher during flushing operations than during flood events at equivalent water discharge?*

Our results demonstrate that the hydrological processes that occur during flushing operations are different from those occurring during flood events (Figures 5-7). In fact, the operations conducted on the dams by drawdown flushing to erode buried materials (Fruchard and Camenen, 2012) resuspended sediments that cannot be affected by flood events. This is why SPM concentrations were higher during flushing operations than flood events at equivalent water discharge. Thus, the use of a power relationship between discharge and SPM as described in Sadaoui *et al.* (2016) and Poulier *et al.* (2019) would lead to the underestimation of the SPM concentrations and fluxes transiting during flushing operations (Figure 5). For example, the total SPM fluxes during the 2012 and 2016 flushing operations would be 0.07 Mt and 0.05 Mt, respectively, when using the equation proposed by Poulier *et al.* (2019), i.e., almost 4 times lower than the measured fluxes (0.26 Mt and 0.19 Mt, respectively – Figure 7).

The excess of annual SPM flux measured at Jons during hydrological years with flushing operations is related to the resuspension of sediment stored during years without flushing (Figure 7). The flushing operations led to a substantial resuspension of silt-sized particles that were stored during other flood events (Table S4). However, this estimation indicates a disequilibrium in the SPM budget over the six years studied on the Upper Rhône River (2011 – 2016 – Figure 7). Therefore, part of the stored sediments was not sufficiently resuspended during flushing operations to reach Jons, and an excess of 0.6 Mt of sediment was trapped in this area despite the 2012 and 2016 flushing operations. Additional investigation on the reservoirs of this area would be necessary to characterise the proportion of SPM stored and resuspended during extreme events.

4.2. *Which parameters influence particulate contaminant concentrations?*

Variations of particulate contaminant concentrations were observed both during the flushing operations and during the different hydrological conditions.

First, the variations observed during the flushing operations for POC and several contaminant concentrations (Hg, PCBs; PAHs, 4-n-nonylphenol and DEHP) are mostly related to the PSD. The lowest concentrations of POC, PAHs and 4-n-nonylphenol were observed on samples collected from 06/11-14/2012 and characterized by at least two third of particles coarser than 15 µm (coarse silt and sand). Also, the highest concentration of POC (22.1 ± 0.9 $mg.kg^{-1}$) was observed in the sample collected from 06/07-09/2012 with the second highest proportion of clay (17.5%). The increase in PAHs concentrations observed in the sample collected from the 06/09-11/2012 (Figure 4) was related to the resuspension of buried materials in the two most upstream dams as the operations started on 06/09/2012 at Verbois and Chancy-Pougny (Figure 2). This will be further discussed in the next section.

Second, the differences of POC and contaminants observed between the various hydrological conditions (Figure 8, Table S5) are related both to the particle size and the origin of the SPM. The decrease of concentrations observed during the flushing operations for most TME, Hg, PCB101, PCB138, and PCB150 is related to the PSD as such contaminants are mainly adsorbed onto finest particles (Delle Site, 2001; Luoma and Rainbow, 2008; Steen *et al.*, 1978). In fact, coarser particles were transported during the flushing operations than during flood events or baseflow (Table S4). Decrease of POC due to PSD was observed both on CFC and PT samples. However, the presence of different trends depending on the contaminant demonstrates that other parameters such as the origin of the particles or local hot spots might also be involved. In this area, input by tributaries might be characterized by different geochemical signature as the concentrations observed for TME and Hg were associated with the geological backgrounds of their watersheds (Poulier *et al.*, 2019; Thollet *et al.*, 2018). For example, the mean concentration of Hg measured from 2011 to 2016 varied by a factor 5 between the Arve River (main SPM contributor – 0.026 ± 0.008 $mg.kg^{-1}$, n = 16) and the Bourbre River (highest Hg concentration - 0.136 ± 0.040 $mg.kg^{-1}$, n = 6). For the different behavior of the PAHs congeners (Table S5), preliminary results on PAHs concentration in SPM collected in the upper Rhône River demonstrate that the different congeners of PAHs have different origins, mainly road traffic and domestic heating (Botha *et al.*, 2014, Poulier *et al.*, 2018).

Furthermore, preliminary work on modeling and fingerprinting conducted on SPM at Jons during the 2012 flushing operations confirmed that SPM mostly originated from the resuspension of SPM deposited in the dams (i.e. mainly from the Arve River) with a low contribution of particles freshly eroded from the other tributaries (Begorre *et al.*, 2018).

Finally, our results confirmed that despite the observed variation of concentrations on the Upper Rhône River, contaminant fluxes transported during dam flushing operations were mostly driven by an increase of SPM concentration, rather than by a change in contaminant contents.

4.3. *Can we explain the increase of some particulate contaminant concentrations?*

The observed increase of PAHs and DEHP concentrations during the flushing operations compared to other hydrological conditions (Figure 8 – supplementary material #2 Table S5) may be related to the resuspension of legacy-contaminated sediments stored behind the dams, due to the drawdown flushing method used. In fact, the SPM resuspended and collected during the flushing operations were stored since 2003 (the last previous flushing operation), i.e. before the implementation of emission regulation of these pollutants (EC, 2008). On the other hand, SPM collected at Jons during baseflow and flood events started in 2011, i.e. after the implementation of the various regulations. PAHs are known to be produced mainly by incomplete combustion (pyrogenic processes) in domestic heating systems using wood (Hedberg *et al.*, 2002; Sicre *et al.*, 2008; Zhang and Tao, 2009). This induces higher PAH concentrations in urban areas and in winter. As reducing PAH emissions by 30% was a priority in the 2nd French National Health and Environment Plan (PNSE 2, 2009), the use of wood was gradually replaced by other more efficient heating systems. This was especially the case in the Arve basin (Figure 1), with a decrease of PAH emissions observed since 2008 (Atmo Auvergne-Rhône-Alpes, 2018). In fact, concentrations of Benzo[a]pyrene measured in SPM downstream of Chancy-Pougny from 2003 to 2008 (NAIADES, 2019) ranged from 10 to 120 $\mu g.kg^{-1}$ (48 ± 34 $\mu g.kg^{-1}$, n=24). In 2009, concentrations dropped and ranged from 16 to 67 $\mu g.kg^{-1}$ (38 ± 25 $\mu g.kg^{-1}$, n=4). During the 2012 flushing operations, the highest concentration (143 $\mu g.kg^{-1}$) was measured on the SPM sample collected at the beginning of the operations on the most upstream dam (06/09/2012 - Figure 1), and is therefore probably related to sediments stored before the regulation.

Finally, the increase of DEHP can also likely be explained by a change in regulations. In fact, DEHP is listed as a priority substance under the European Water Framework Directive (WFD; CEC, 2000). Its emissions are therefore regulated since 2008. DEHP was also included in 2011 in the Annex XIV of REACH by Regulation (EU, 2011), inducing the need of authorizations (and restrictions) for its specific uses.

5. Feedback for future studies on sediment dynamics and quality during dam flushing operations

Our study demonstrates that SPM dynamics on the Upper Rhône River differed between flushing operations and natural floods, and confirms the necessity to monitor both discharge and SPM concentrations during such events. Also, the physico-chemical characteristics of SPM and associated contaminants might help to understand the dynamics and quality of the sediments. In this part, we focus on the best methodology to study sediment dynamics and quality during dam flushing operations.

i) Design the monitoring to assess SPM concentrations and associated fluxes

Both aspects (concentrations and fluxes) are important for assessing the ecological impact of dam reservoir flushing operations:

- For computing fluxes and mass budgets, continuous records of discharge and SPM concentration are required using gauging stations and turbidity records calibrated with frequent water samples. General relationships between SPM and discharge should not be used during flushing operations (see Figure 5).
- Monitor the entire flushing period for discharge and calibrated turbidity time series. We recommend assessing the type of flushing operations conducted (e.g.,

pressure flushing or drawdown flushing (Fruchard and Camenen, 2012; Kondolf *et al.,* 2014)).

ii) Select the contaminants and other physico-chemical parameters to assess SPM quality

- Optimise the selection of contaminants based on the results of previous monitoring (baseflow, flood, dam flushing operations or on sediments stored behind dam reservoirs), including the screening of new contaminants that were not sought or detected before;
- Selection criteria include: priority pollutants according to regulations, performance of the analytical methods (limit of quantification, uncertainty) and emergent pollution, watershed anthropic pressure;
- Monitor SPM quality over extended periods of time. Time integrative sampling systems like passive particle traps are appropriate (Masson *et al.*, 2018).

iii) Extend the observations beyond the dam flushing event (temporally and spatially)

Comparison with other periods of time and various hydrological conditions (average, baseflow, floods, other dam flushing events) are necessary to assess the relative impact of a dam flushing operation:

- It is useful to have at least one monitoring station with multi-year records of discharge, SPM concentration and physico-chemical parameters (POC and PSD);
- It is also important to monitor the discharge and SPM load of the main tributaries in the river system; additional information such as physico-chemical parameters

of the tributaries might also help understand the results observed at the monitoring station.

- Store SPM samples in proper conditions and with associated documentation to allow for future analyses, in case suspicious results are obtained for a given contaminant, or for future re-analysis of past events, for other contaminant and/or using better analytical methods.

4. Conclusions

Variations of SPM and associated contaminant concentrations and fluxes were investigated from 2011 to 2016, including two dam flushing operations, at a permanent monitoring station located on the Upper Rhône River, France.

Despite the fact that flood events contributed to most of the annual flux of SPM, fluxes triggered by flushing operations were substantial (21-37% of the total annual SPM) despite their low annual duration (less than twenty days). These novel results demonstrate the necessity to have a long-term monitoring station located downstream to evaluate the impact of these operations.

Additional measurements of water discharge and SPM concentration along the Upper Rhône River and the main tributaries were used to investigate the spatial variation and to estimate the input/output SPM fluxes. The output fluxes measured at Jons were lower than the input fluxes during hydrological years without flushing operations and otherwise higher, attesting of a removal of the stored SPM. Flushing operations triggered the resuspension of sediments stored during flood events in reservoirs located along the Upper Rhône River. It is therefore crucial to continue such monitoring to investigate the fate of this excess of stored sediment. Moreover, the composition of SPM changed during flushing operations as demonstrated by the analysis of several parameters and contaminants. Although the variations of POC and contaminants during the flushing operations were mostly related to changes in particle size, the origin of particles (e.g., resuspension of deeply stored sediments) was also important. Finally, we shared a general methodology to conduct similar monitoring.

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Figure caption

Figure 1 – Location of the studied dams in the Upper Rhône River catchment with the main tributaries and the permanent monitoring station at Jons.

Figure 2 – Water level variation in the three dams studied on the Upper Rhône River during the 2012 dam flushing operations (modified from CNR, 2014).

Figure 3 – Variation of water discharge and the SPM concentration measured at Jons during the hydrological year of the studied period (September 2011 to August 2012). Flood threshold was estimated at 800 $m^3.s^{-1}$. The gray area represents the period of the 2012 flushing operations.

Figure 4 – Particulate contaminant concentrations (± analytical uncertainty) in SPM samples collected in the Rhône River at Jons during the flushing operations in June 2012.

Figure 5 – Relation between discharge and SPM concentration measured in the Rhône River at Jons from September 2011 to August 2016 (black circles). Data for the flushing operations of June 2012 (purple circle) and June 2016 (cyan circle) are shown. Flood threshold was estimated at 800 $m^3.s^{-1}$. The pink line represents the relationship between hourly water discharges and hourly SPM concentration at the station of Jons according to Poulier et al. (2019).

Figure 6 - Proportion of annual SPM fluxes of the Rhône at Jons for the different hydrological conditions (baseflow, flood and flushing event) against the annual duration of the events

Figure 7 – Annual SPM fluxes of the Rhône River at Jons from 2011 to 2016 (hydrological years) for the different hydrological conditions (baseflow, flood and flushing events of 2012 and 2016); total output SPM fluxes (measured at Jons) and input SPM fluxes (computed from data on tributaries) are indicated for comparison.

Figure 8 – Particulate contaminant concentrations in SPM samples collected in the Rhône River at Jons manually, by CFC or by PT between hydrological conditions (baseflow, flood and flushing). For Cd, one outlier is not displayed for baseflow (value = 2.41 $mg.kg^{-1}$). Only the significant differences are displayed with * for p value < 0.05, ** for p value < 0.01, *** for p value < 0.001 and **** for p value < 0.0001. Box plots represent the median and quartile values, Black circles and red diamonds represent outliers and mean values, respectively. n = number of samples.

Figure 1

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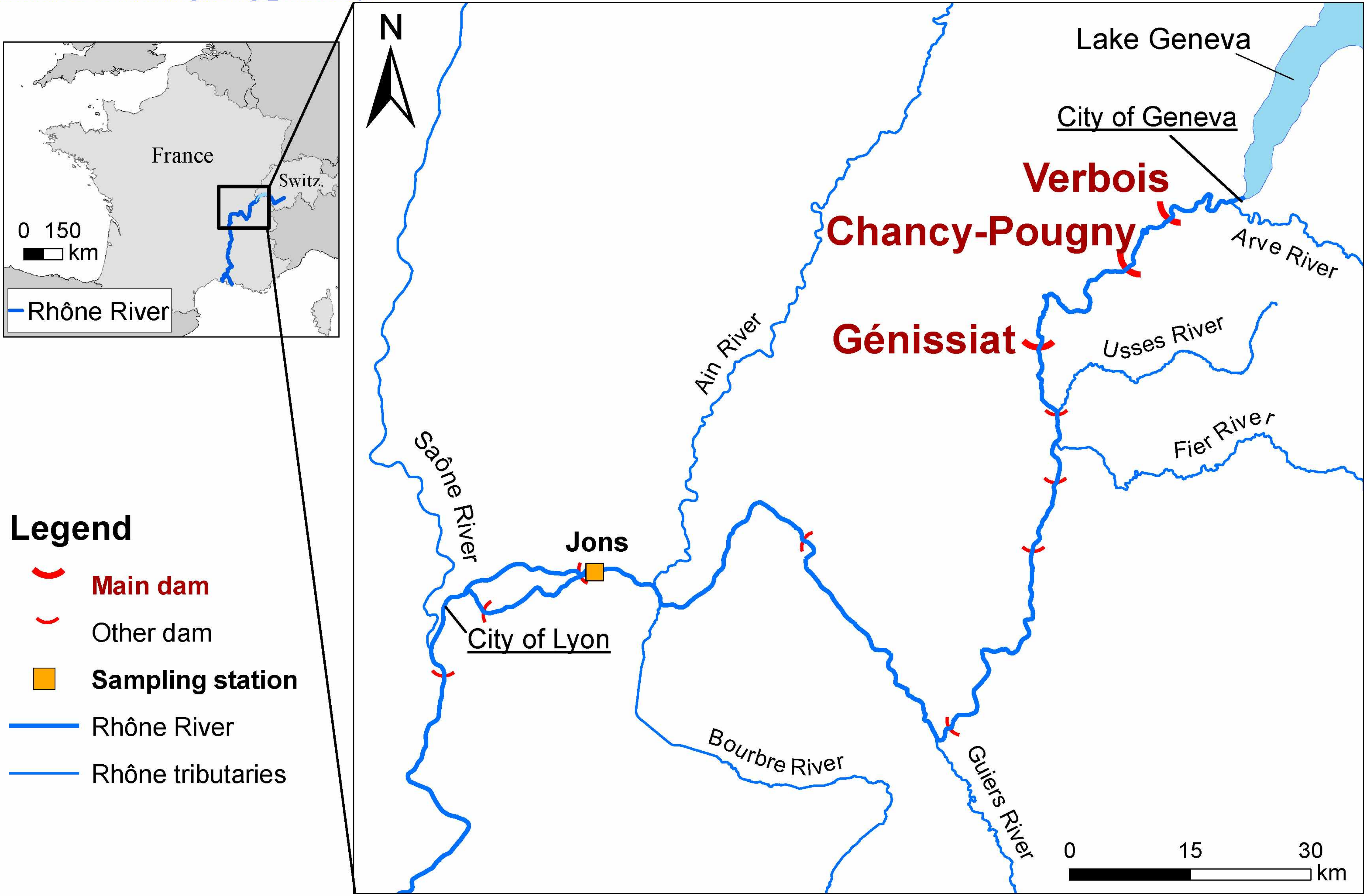

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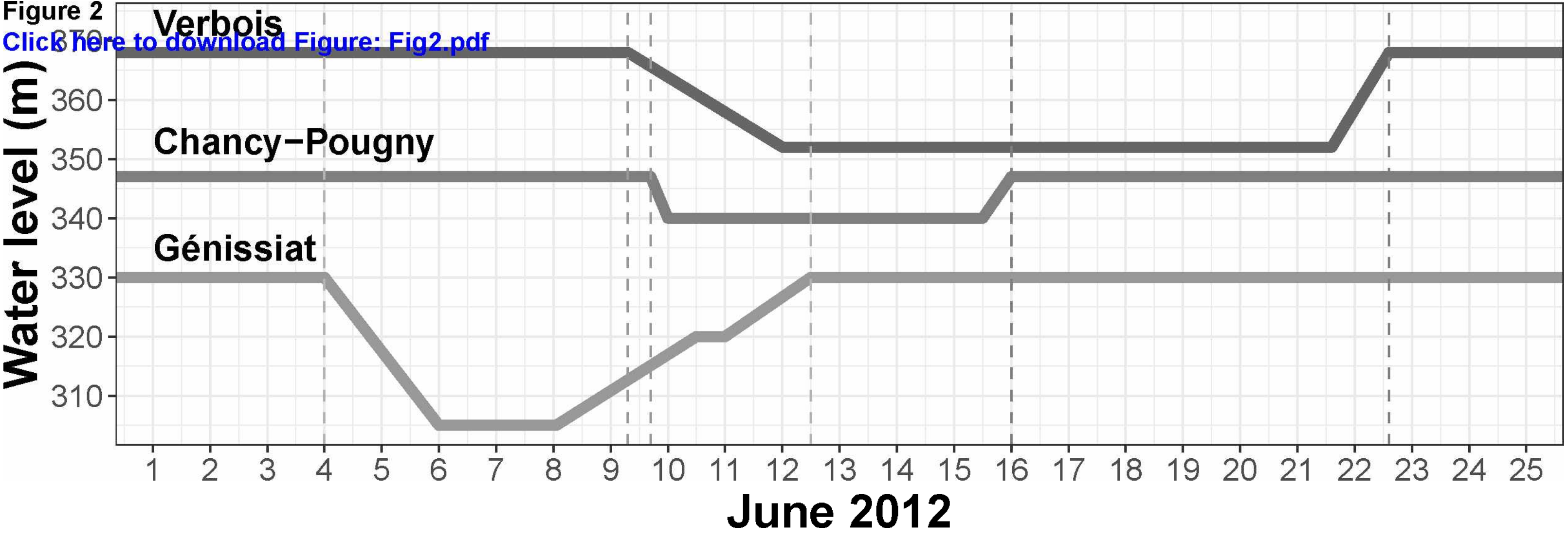

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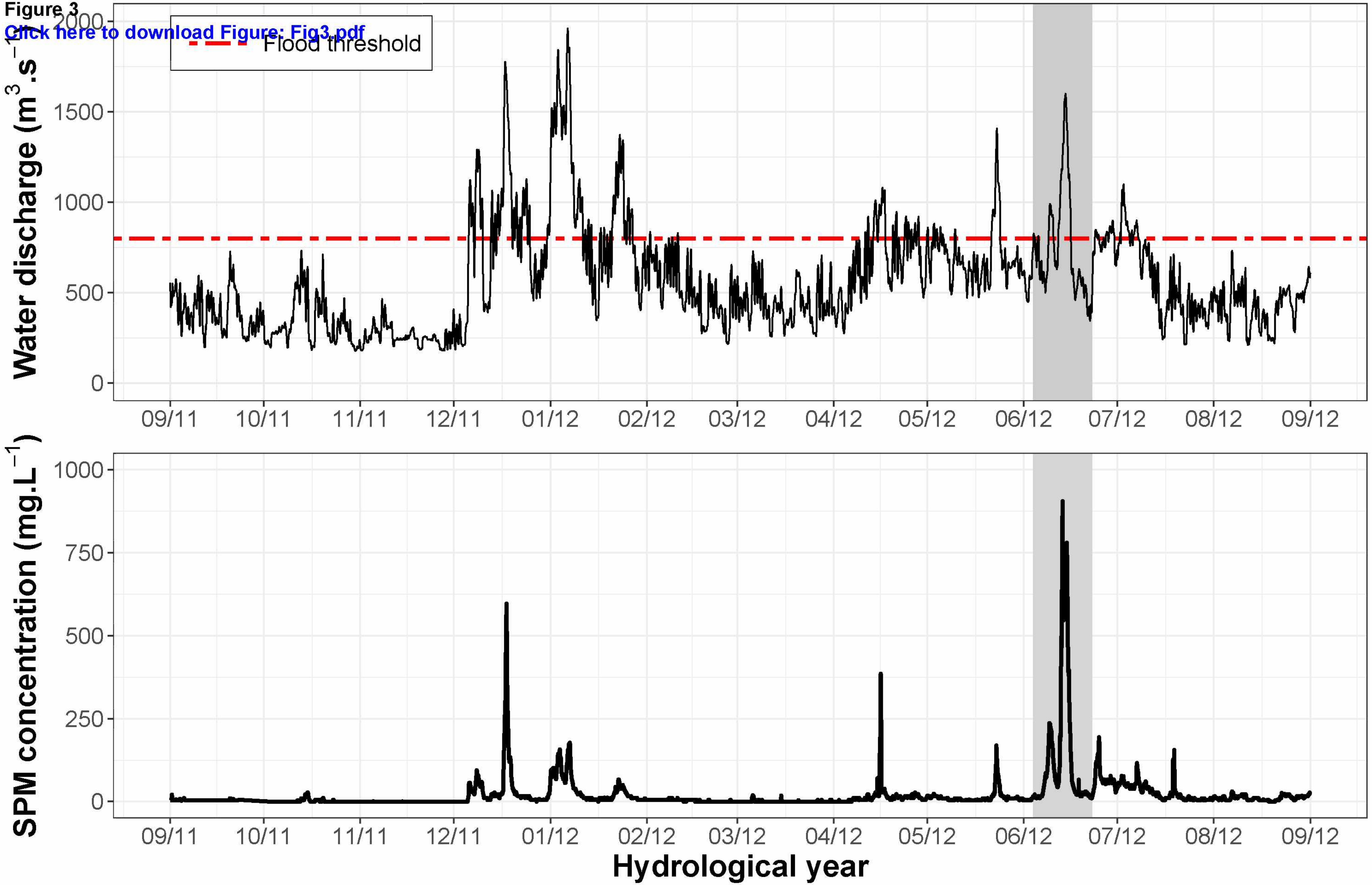

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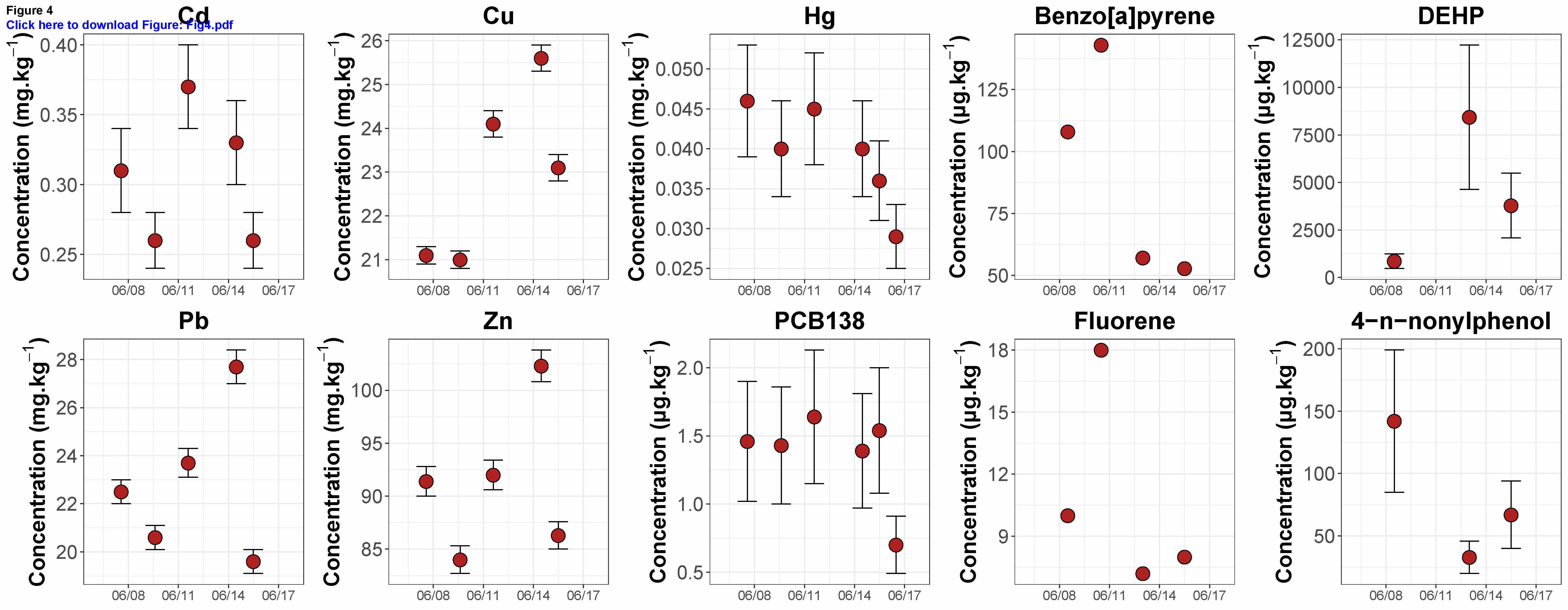

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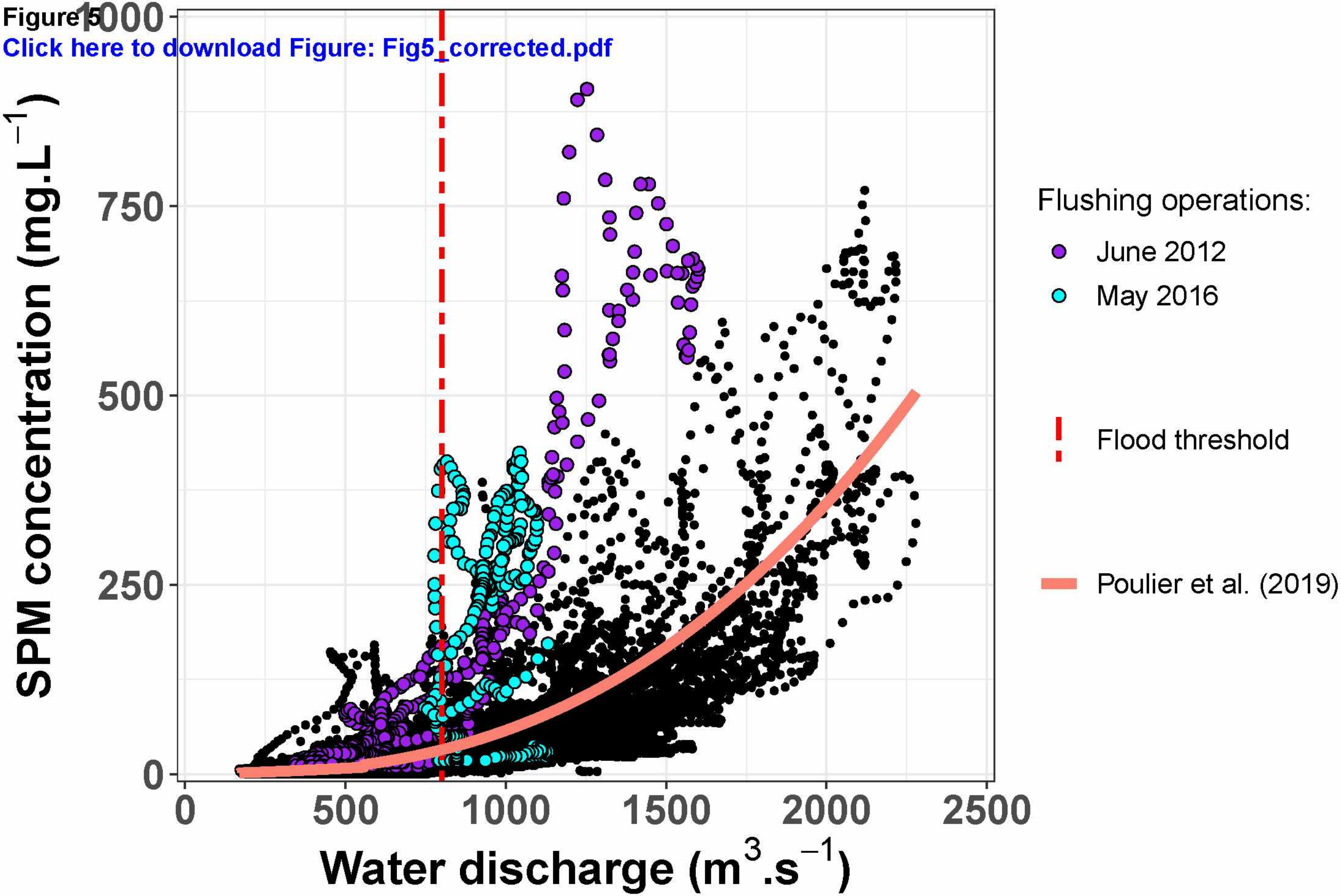

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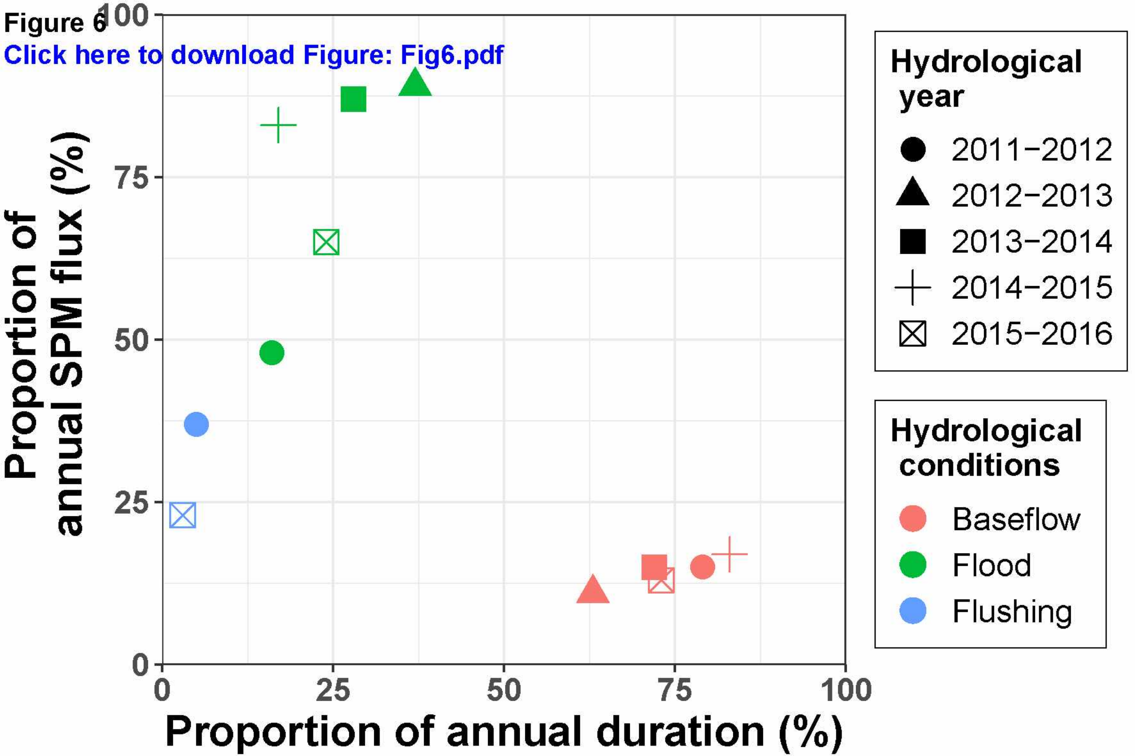

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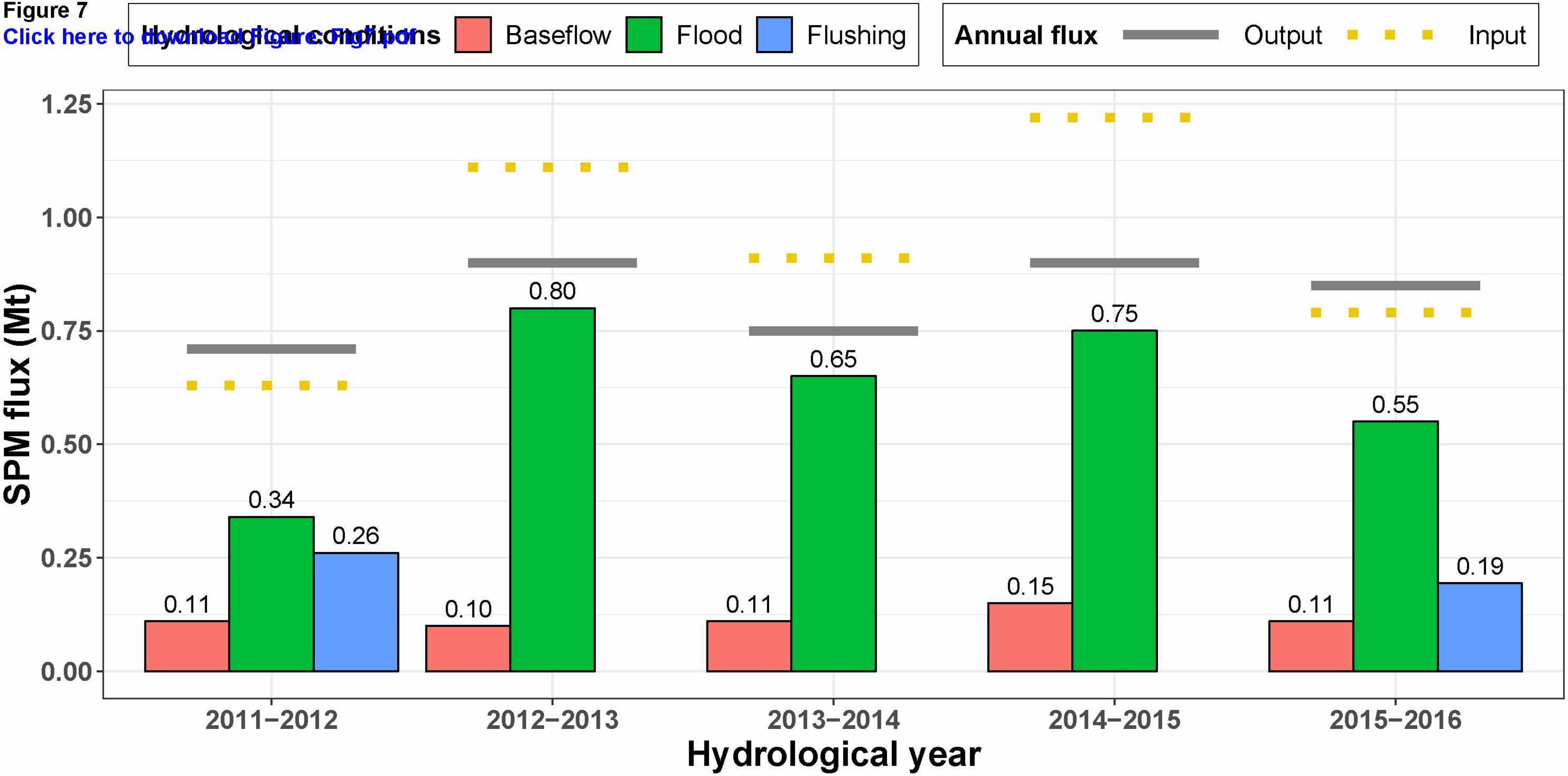

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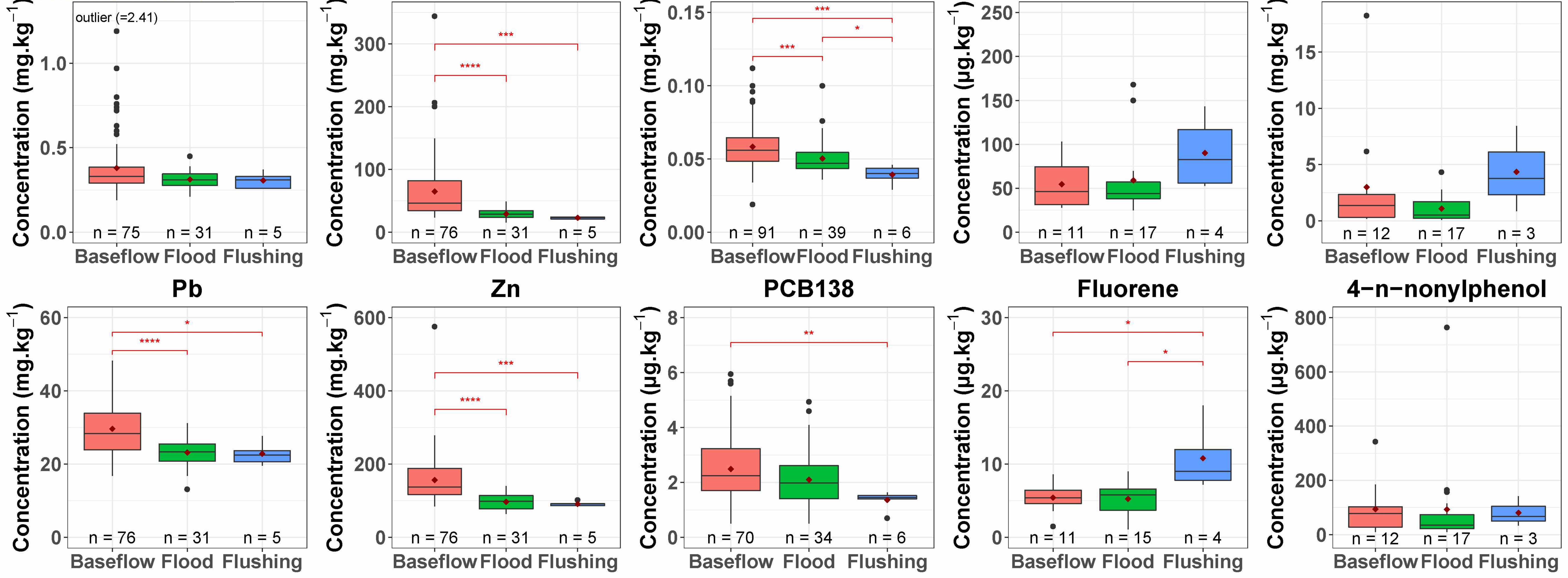

Table
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Table 1 – Proportions (%) of annual SPM and particulate contaminant fluxes in the Rhône River at Jons for the different hydrological conditions (baseflow, flood, flushing event) from September 2011 to August 2012.

Hydrological condition	SPM	Cd	Cu	Pb	Zn	Hg	PCB138
Baseflow	15.5	15.7	15.1	17.1	16.6	14.5	17.5
Flood	47.9	48.9	53.1	49.6	50.7	52.7	55.8
2012 Flushing	36.6	35.4	31.8	33.3	32.8	32.8	26.7

Supplementary materials
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*Declaration of Interest Statement

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Jérôme Le Coz: Conceptualization, Methodology, Resources, Writing - Original Draft

Hélène Angot: Conceptualization, Methodology, Investigation, Writing - Review & Editing

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Stéphanie Gairoard: Conceptualization, Methodology

Olivier Radakovitch: Conceptualization, Methodology, Resources, Writing - Review & Editing

Marina Coquery: Conceptualization, Methodology, Writing - Original Draft, Supervision