

Interactive comment on “Metal contamination budget at the river basin scale: a critical analysis based on the Seine River” by L. Lestel et al.

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Referee #1

Specific comments 1. The F2A methodology and vocabulary: The different meaning between flux and flow may be clarified earlier than line 3-4 page 1799 in order to better understand the F2A methodology. If it is common, please reference it. You can also better enhance that the F2A methodology is an original procedure. Why do you reference Steffen et al (2004) in the conclusion: do they already use this method ? The paragraph on the definition of Fluxes and Flows has been rewritten in order to be more explicit and put earlier in the text (from page 1799 to the introduction). We changed the title in order to make clearer the originality of our own procedure (the F2A approach). We changed the first sentence of the conclusion in order to be clearer. Steffen et al.

(2004) spoke about local to global scale and not about combined fluxes/flows analysis.

2. The contamination budget of Cd, Cu, Cr, Pb and Zn The description of metal transfer is done with different indicators, one metal with one parameter, another metal with another parameter. It appears difficult to obtain a clear image of Cd, Cu, Cr, Pb and Zn transfers, specially without figures. You may describe one or two distinct metals. The aim of this work was not to focus on one metal as usually done in the literature, but to have a global approach of the link between the anthropogenic and the environmental eras whatever the metal considered. We thus chose illustrative data rather than data dedicated at one distinct metal.

3. Calculations How is the total metal output calculated from natural sources (line 26 page 1805) ? Calculations of the per capita excess; and leakage ratio; indicators should be more detailed than line 24-29 page 1806. Can you make clearer how you calculate the percentage of unknown sources (Fig. 6) ? We developed the explanations related to Fig. 6 in the text, in order to better explain how we deduced the percentage of unknown sources;. (i) better explanation of natural sources: The contribution of soil erosion was calculated on the basis of an average river SPM yield of 10.8 t km⁻² y⁻¹ and a basin average metal content for pristine SPM. (ii) better explanation of industrial sources: The industrial sources result both from voluntary industrial declaration procedure and from partially measured and extrapolated fluxes (see Table 3, L3A/L5 calculated or measured by different ways). Details and estimated uncertainties ranges are given in Thévenot et al. (2007). (iii) better explanation for the unknown sources: The difference between the sum of natural, domestic and industrial sources and the measured total metal output represents the unknown output;, ranging from 83% of the total output for Hg to 30% for Pb

Editorial / Technical comments Page 1796 line 21-22 : “considered”
twice

Page 1797 line 14 :give references for ECA studies Done

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Page 1801 line 5 : why is METOX unusable for MFA ? We add a sentence: this aggregated indicator is obviously unusable for MFA, where data specific for each metal are required.

Page 1803 line 22 : replace $\&\#8220$;approaches $\&\#8221$; by $\&\#8220$;studies Done

Page 1803 line 28 : what does PbEt4 mean ? tetraethyl lead (PbEt4)

Page 1804 line 23 : why do you use parameters such as T1A, T1F, P7... in the text and not in the figure 4. Values, cited in the text, appear difficult to find in the figure 4. We changed Fig.4 to add the parameters.

Page 1805 line 21 : aeolian and not Aeolian Done

Page 1806 line 1-6 : put all the explanations about domestic inputs to page 1805 line 27 We changed the whole paragraph, to add explanations about natural sources, as well as industrial sources (see above).

Figure 2 : why do you not use only S for storage ? what do the different arrows mean ? What is the difference between SR3 and SR4 ? what does $\&\#8220$;scrap $\&\#8221$; mean ? Identify the anthroposphere S is for Storage, SR is for Storage in River. The arrows are a representation of fluxes and flows. There is no real difference, except that one was upstream and the other one downstream of the city. We suppressed SR4. The scrap industry is the recycling industry. This is a specific term. We added an explanation in Table 1. We put a circle around the anthroposphere.

Figure 4 : why are the boxes different from Figure 2 ? Fig. 2 is more general than Fig.4, which is specific to Zn. However, fluxes and flows have the same meaning.

Figure 5 :This figure is already published in Thevenot et al, 2007 in the exact same way so you can delete it in this article Done

Figure 6 : delete $\&\#8220$;sources $\&\#8221$; in the legend Now Fig. 5 Done.

Figure 7 : add the symbols $\&\#184$;T and $\&\#9830$; in the legends for A and B. A readings

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on the left or on the right ? Now Fig. 6. We suppressed the arrows on the figure and added symbols in the legend, in order to be clearer.

Table 1 : what is a miscellaneous source ? why do you use P8B ? P8A exists in figure 2 but it is not explained. Why do you use P2 and P3, it seems similar ? In the figure 2, P1A + P1B exist but they are not explained in Table 1 It means that fluxes were calculated from several kinds of data, for example car traffic (INSEE) and evolution of lead concentration in gasoline (academic publications). Data for P8B are given by the SIAAP but we have no clear data for P8A. P8A corresponds to sewage sludge which is not sent in cultivated area and is stocked in landfills. P2 corresponds to metal import to the Basin and P3 to metal export from the Basin. P1A referred to ores and P1B to ingots. We should have to simplify Fig. 2.

Table 2 : Grosbois instead of Grobois.

In figure 2, T2F, T2A, T1U exist but they are not explained in Table 2 T1 are fallout on forests (T1F), cultivated areas (T1A) and urban areas (T1U).

Table 3: L3A/L5 seem to be different data with similar symbols : why ? L3A and L5 are metal outputs from the industry. Whatever the data providers, these outputs are separated into direct release to rivers (L5) or release to the sewage system (L3A).

Referee #2

Specific comments:

1. It would be helpful to further explain briefly the exact difference (including the units) of the concepts of flux and flow. The paragraph on the definition of Fluxes and Flows has been rewritten in order to be more explicit and put earlier in the text (from page 1799 to the introduction). Flows and fluxes units are generally t/y.

2. To what extent do peak flows have contributed to the total transport of pollutants in the basin, and how can we be sure that these large fluxes that occur during short periods of time have been included in the estimates? This has been already discussed

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in several papers of Meybeck et al., for example in Grosbois, 2006. Annual flood deposits collected after winter high-flow events correspond to more than 50% of the total annual fluxes. This approach has been compared with daily fluxes determined on the basis of the metal analysis on suspended particulate material (SPM) from 12 to 24 times per year, and the daily knowledge of the river TSS fluxes. We are making the hypothesis that SPM composition is constant around the sampling day for SPM analysis. This hypothesis has been tested by Idlafkih, 1995. It was concluded that the sediment associated trace element temporal trends observed at the mouth of the Seine River Basin at Poses, closely match the trends determined from bimonthly SPM samples.

Idlafkih, Z, Cossa D, Meybeck M. Comportement des contaminants en trace dissous et particulaires (As, Cd, Cu, Hg, Pb, Zn) dans la Seine. *Hydroécol Appl* 1995;7:127-150 [in French]. Grosbois, in reference list.

3. There seems a risk in circular reasoning when expressing output per capita after the output has been rescaled proportionally to the number of inhabitants. I presume this has not happened. Numerous data needed for the determination of Excess metal load are not linked to the population (natural erosion, industrial outputs, sewage data, etc.). There is no reason for circular reasoning.

4. The meaning of 'recycling' (p1808, l. 26): is it truly re-using of the material, or does it also include storage of waste or contaminated sewage sludge, such that it does no longer leach into the environment. It includes all the ways metal leakages are avoided. The last sentence in the conclusion should be corrected.

5. The authors mention that similar studies should be undertaken for other rivers, such as the Rhine, Elbe, Scheldt or Humber (p1808, l.25). At least they could give reference to studies on those rivers that have attempted to reconstruct their pollution history. Although these do not give estimates of the indicators of the present study, it is interesting to compare the pollution trends among rivers. International references about metal fluxes circulation in the environment have been added in the introduction

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and a comparison of the Seine River with other European rivers has been added as a new paragraph (4.4) just before the conclusion. The Seine river is among the most contaminated rivers, but a general decline in metal content is observed in the 1960 as for the other rivers. A figure has been added to illustrate this point for Zn.

Editorial / technical comments: 1. Methods section (chpt 3, p 1799 -): use past tense (were, instead of 'is'; or 'have been'), unless other tense are truly appropriate. Done

2. p1800, l.15 replace 'as examples'; by 'for example'; Done

3. p1800, l. 24 'estimated from (i) nominal'; and do not repeat 'from'; in subsequent items Done

4. p1801 l. 16,17: use: 'such as'; Done

5. p1801 l.18: replace 'prorated to the proportion of'; by 'scaled proportionally to'; Done

6. p1805, l. 17: place ';&'; at the end of this line Done

7. p1806, l. 23: replace 'the proposition of'; by 'proposing'; Done

Interactive comment on Hydrol. Earth Syst. Sci. Discuss., 4, 1795, 2007.

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