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SUPPLEMENTARY MATERIAL

Antimony in the Lot-Garonne river system: a 14-year record of solid-liquid partitioning and fluxes

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Figure S1. Distribution frequency histograms of dissolved Sb (µg L^{-1}) and particulate Sb (mg kg^{-1}) concentrations, with average daily river discharge (m^3 s^{-1}) and suspended particulate matter (mg L^{-1}), for the whole database at La Réole (from 2003 to 2016): with outliers (a - d) and without outliers (e – h). Corresponding expected Gaussian distributions overlapping each histogram are also shown.

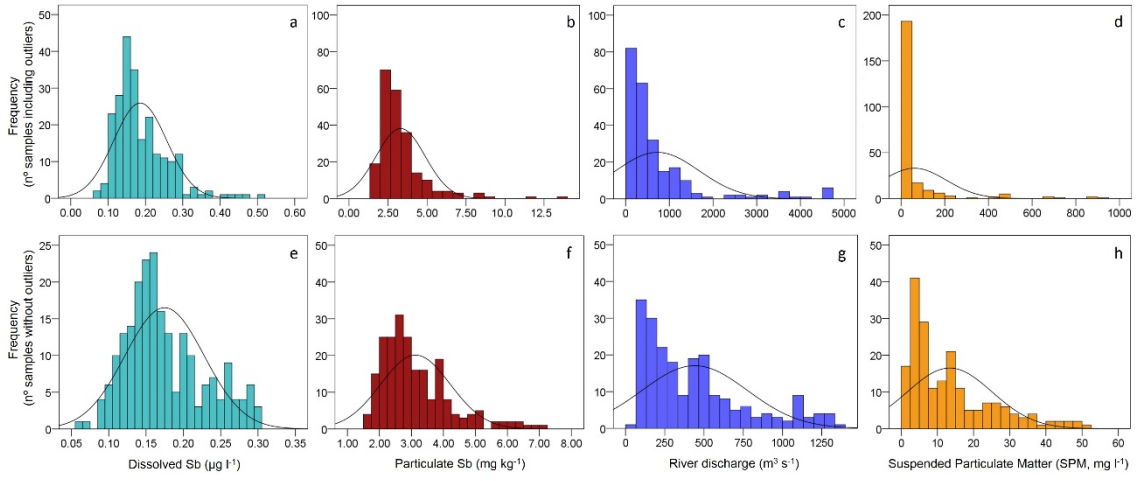

Figure S2. Temporal trend of the modified geoaccumulation index (I'_{geo}) for the five sampled sites (a: LR and PSM, b: T, BP and RM). Colour code correspond to I'_{geo} classification: class 1 (0–1, unpolluted to moderately polluted, blue), class 2 (1–2, moderately polluted, green), class 3 (2–3, moderately to strongly polluted, yellow), class 4 (3–4, strongly polluted, pink), and class 5 (4–5, strongly to very strongly polluted, red).

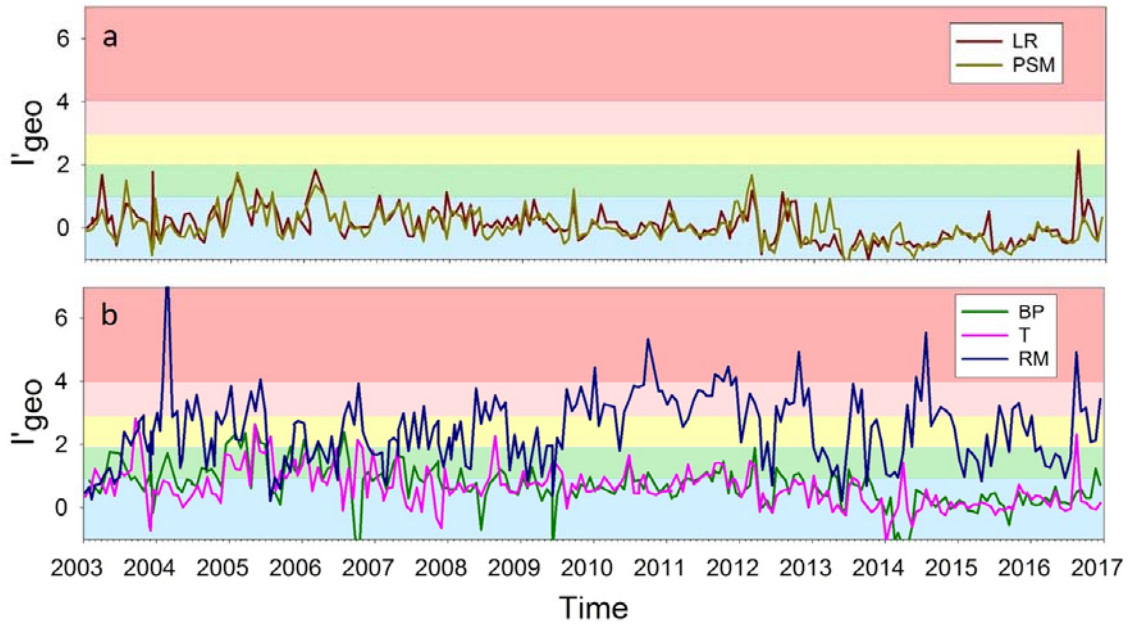

Figure S3. Seasonal variation of Sb $\log_{10} K_d$ (L kg^{-1}) at La Réole, Temple and Port-Sainte-Marie for 2003-2016.

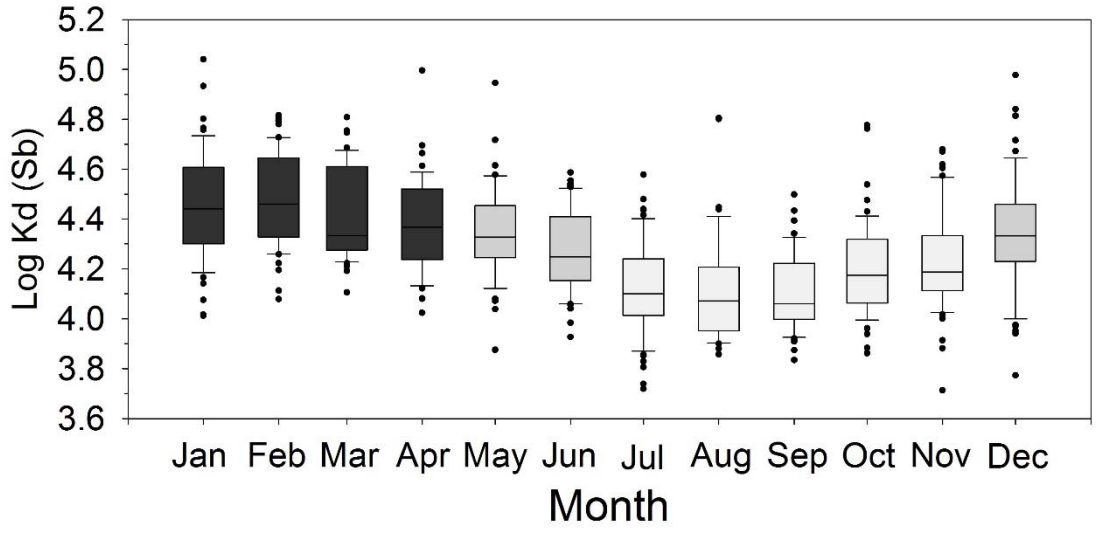

Figure S4. Temporal trends (2004 – 2016) of annual water (Q, in km^3 $year^{-1}$, cyan solid line) and suspended particulate matter fluxes (SPM, in Mt $year^{-1}$, grey area) with annual Sb_d (t $year^{-1}$, solid lines) and Sb_p discharged-weighted fluxes (t $year^{-1}$, filled symbols) at all sampled sites: La Réole (LR, a), Port-Sainte-Marie (PSM, b), Temple (T, c), Boisse Penchot (BP, d) and Riou Mort (RM, e).

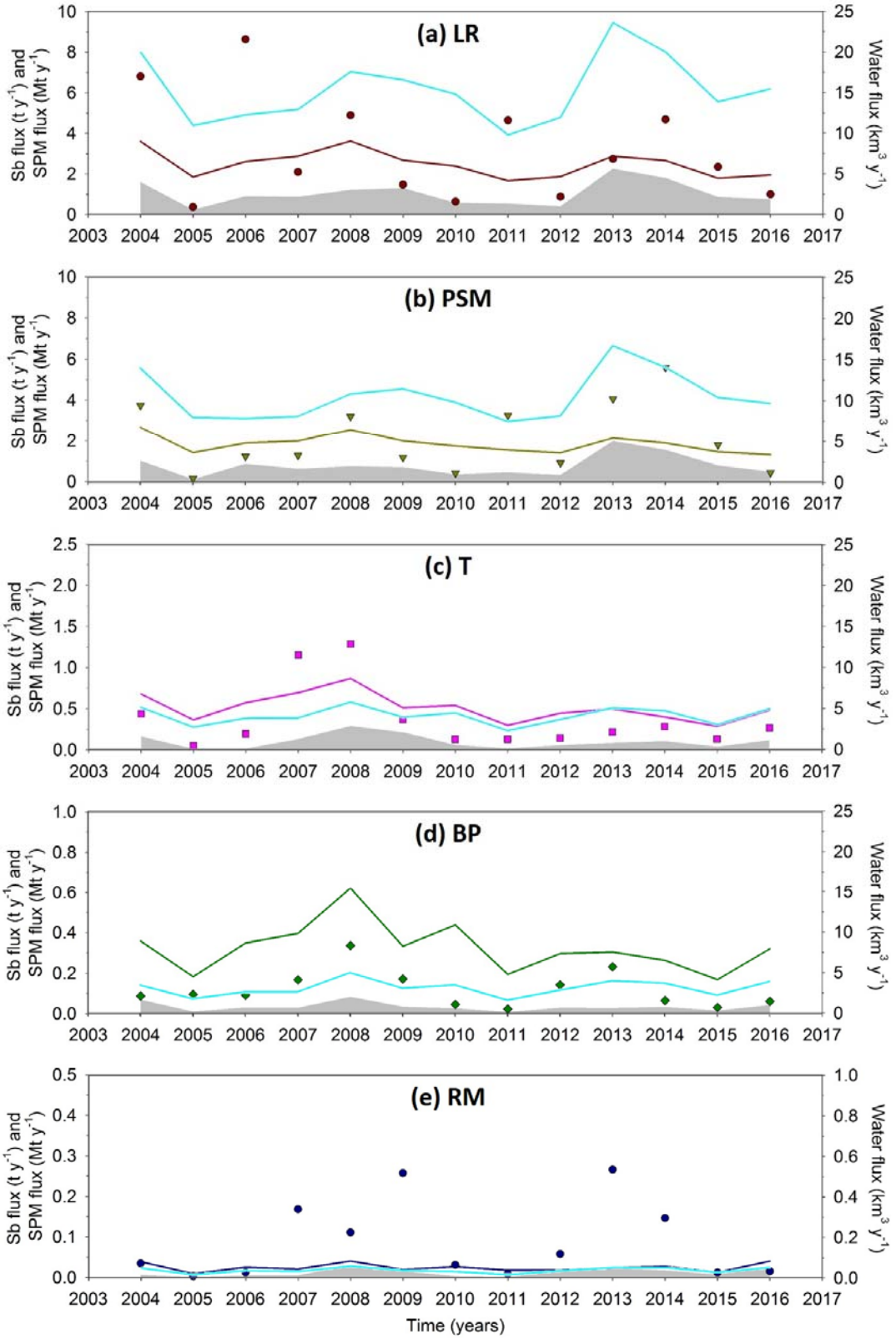
(a) LR
(b) PSM
(c) T
(d) BP
(e) RM
Sb flux (t y-1) and SPM flux (Mt y-1)
Water flux (km3 y-1)
Time (years)
2003 2004 2005 2006 2007 2008 2009 2010 2011 2012 2013 2014 2015 2016 2017

Table S1. Record of Certified Reference Materials (CRM) used during the 2003 to 2016 period for validation of particulate (PACS-1, IAEA-433, NCS DC 70311, NCS DC 70317, NIST SRM 8704) and dissolved (TMRAIN-95, TMRAIN-04, SLRS-4, SLRS-5, SLRS-6) Sb analyses.

Matrix	CRM	Units	Certified	Measured	
			Mean ± SD	Mean ± SD	Replicates
Marine sediment	PACS-1	mg kg^{-1}	171 ± 14	182 ± 19	N = 9
Marine sediment	IAEA-433	mg kg^{-1}	1.96 ± 0.18	1.71 ± 0.08	N = 15
Rock powder	NCS DC 70311	mg kg^{-1}	13.8 ± 0.8	15.1 ± 1.2	N = 28
Rock powder	NCS DC 70317	mg kg^{-1}	4.44 ± 0.44	5.00 ± 0.51	N = 38
River sediment	NIST SRM 8704	mg kg^{-1}	3.07 ± 0.32	2.74 ± 0.28	N = 25
Rain water	TMRAIN-95	µg L^{-1}	0.35 ± 0.10	0.40 ± 0.03	N = 33
Rain water	TMRAIN-04	µg L^{-1}	0.35 ± 0.07	0.32 ± 0.03	N = 84
River water	SLRS-4	µg L^{-1}	0.23 ± 0.04	0.28 ±0.03	N = 159
River water	SLRS-5	µg L^{-1}	0.3*	0.28 ± 0.04	N = 165
River water	SLRS-6	µg L^{-1}	0.34 ± 0.01	0.32 ± 0.02	N = 20

*informational value