10.1071/EN24041
Environmental Chemistry

Supplementary Material

Solubility and dissolution kinetics of particle-bound metals in a surrogate lung fluid

Sara D'Aronco[A], *Valerio Di Marco*[B], *Alberto Gambalunga*[C], *Federica Chiara*[D], *Andrea Trevisan*[C] and *Chiara Giorio*[A,*]

[A]Yusuf Hamied Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge, CB2 1 EW, UK

[B]Dipartimento di Scienze Chimiche, Università degli Studi di Padova, Via Marzolo 1, I-35131 Padova, Italy

[C]Department of Cardiac Thoracic Vascular Sciences and Public Health, Università degli Studi di Padova, Via Giustiniani 2, I-35128 Padova, Italy

[D]Department of Surgery, Oncology and Gastroenterology, Università degli Studi di Padova, Via Giustiniani 2, I-35124 Padova, Italy

*Correspondence to: Email: chiara.giorio@atm.ch.cam.ac.uk

Optimisation of SELF cleaning with Chelex 100

For optimisation of the cleaning procedure as well as identification of which ingredients contributed the most to any background contamination of metal and metalloid ions, we prepared one test solution for each individual component of the SELF. The concentration of each compound in these "test" solutions was the same as that in final SELF solution (e.g. NaCl 6020 mg L^{-1} in SELF, NaCl 6020 mg L^{-1} in the test solution). The test solutions were then analysed for trace metal quantification (Table S1).

Table S1. Background contamination of trace metals in each component of the SELF.

Test solution	Co (ppb)	Cu (ppb)	Fe (ppb)	Mn (ppb)	Pb (ppb)	Sn (ppb)	Zn (ppb)
$MgCl_2$	n.d.	n.d.	0.75	0.62	n.d.	0.20	n.d.
$CaCl_2$	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
$NaHCO_3$	n.d.	n.d.	0.86	1.83	n.d.	2.55	n.d.
NaCl	n.d.	n.d.	1.29	0.40	n.d.	0.15	n.d.
Na_2HPO_4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Na_2SO_4	n.d.	n.d.	1.30	0.51	n.d.	0.07	n.d.
KCl	n.d.	n.d.	0.88	0.49	n.d.	n.d.	n.d.
Ascorbic acid	n.d.	0.49	1.68	0.51	n.d.	1.17	n.d.
Uric acid	n.d.	n.d.	0.75	0.47	n.d.	0.81	n.d.
Cysteine	n.d.	n.d.	1.73	0.55	n.d.	n.d.	0.30
Glycine	n.d.	n.d.	1.49	0.47	n.d.	0.37	n.d.
Albumin	n.d.	0.53	2.12	0.58	1.69	1.03	7.68
Mucin	n.d.	15.81	64.5	4.03	n.d.	0.52	49.44
Glutathione	n.d.	4.64	35.69	6.90	n.d.	n.d.	25.71

n.d., not detected

Each test solution was cleaned with Chelex 100 using the "batch method" as detailed in the instruction manual (Bio-Rad Laboratories[1]). Briefly, 5 g of resin were added directly to 100 mL of test solution. The solutions were then left stirring at room temperature for 1 h. The resin was then decanted and the surnatant was collected for analysis of trace metal concentrations (Table S2).

[1] Chelex 100 and Chelex 20 Chelating Ion Exchange Resin Instruction Manual, see https://www.bio-rad.com/webroot/web/pdf/lsr/literature/LIT200.pdf

Table S2. Background contamination of trace metals in each component of the SELF after cleaning with Chelex 100.

Test solution	Co (ppb)	Cu (ppb)	Fe (ppb)	Mn (ppb)	Pb (ppb)	Sn (ppb)	Zn (ppb)
$MgCl_2$	n.d.	n.d.	n.d.	0.38	n.d.	n.d.	n.d.
$CaCl_2$	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
$NaHCO_3$	n.d.	n.d.	n.d.	0.43	n.d.	n.d.	n.d.
NaCl	n.d.	n.d.	n.d.	0.49	n.d.	n.d.	1.07
Na_2HPO_4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Na_2SO_4	n.d.	n.d.	n.d.	0.58	n.d.	n.d.	n.d.
KCl	n.d.	n.d.	n.d.	0.64	n.d.	n.d.	n.d.
Ascorbic acid	n.d.	n.d.	n.d.	0.68	n.d.	n.d.	n.d.
Uric acid	n.d.	n.d.	n.d.	0.55	n.d.	n.d.	n.d.
Cysteine	n.d.	n.d.	n.d.	0.61	n.d.	n.d.	n.d.
Glycine	n.d.	n.d.	n.d.	0.35	n.d.	n.d.	n.d.
Albumin	n.d.	n.d.	n.d.	0.66	n.d.	n.d.	n.d.
Mucin	n.d.	8.64	n.d.	0.75	n.d.	n.d.	n.d.
Glutathione	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

n.d., not detected

Cleaning the test solutions of inorganic salts with Chelex reduced the amount of Fe and Sn but it was not effective for Mn. However, the inorganic salts appeared to have a low background contamination from metals to start with. Among the organic components mucin was the one found to contain the higher amounts of metals (Cu, Zn, Fe, and Mn). Cleaning with Chelex treatment reduced the amount of Cu (from 15.81 to 8.64 ppb), Zn (from 49.44 to below detection limit), Fe (from 64.51 to below detection limit). Other organics showed the presence of Fe, but its concentration fell below detection limit after cleaning with Chelex.

Considering the initial background contamination level of each component and the effectiveness of the Chelex treatment we decided: (1) clean the organic solution with Chelex (before adding the DPPC that requires a specific procedure to be able to remain in suspension, see the 'SELF preparation' in the main paper for details), (2) not to clean the inorganic solution.

Data on kinetics profiles for each element

Table S3. Data of the dissolution kinetics in SELF for Cu reported as concentrations in air (ng/m^3).

Sample ID	24 B	26 B	28 B	16 +18 B	32 B	22 B	20 B	30 B	12+14 B	38 + 40 B
Time (min)										
0	4.9	11.3	7.3	0.80	6.2	9.6	6.6	5.8	0.38	0.98
1	7.9	13.2	8.7	0.80	3.4	10.8	7.6	6.6	1.2	1.8
2	6.8	13.5	11.0	1.1	2.3	11.7	8.2	7.1	1.6	2.2
3	8.7	14.3	10.5	1.4	2.8	11.4	8.8	7.9	1.6	2.6
4	8.3	13.8	9.7	3.0	4.0	13.2	8.0	8.1	1.5	2.1
5	11.0	15.5	12.8	2.1	3.5	11.9	8.0	8.4	1.8	2.2
6	8.4	13.7	16.4		4.9	14.4	7.9	8.2	2.2	1.8
8	9.5	13.6	11.0	3.2	3.3	11.9	8.4	8.4	2.0	2.0
10	8.6	14.9	11.1	3.4		14.3	8.7	8.1	2.7	1.9
12	9.4	16.0	11.8	3.6	5.0	12.5	8.4	8.6	2.5	2.3
14	10.3	14.9	12.2	2.9	5.4	11.6	8.4	8.5	2.4	1.9
16	9.3	16.5	12.8	3.1	6.2	12.3	8.7	9.1	3.4	2.0
20	8.9	14.7	11.5		4.8	13.3	9.9	8.6	2.4	2.0
24	8.3	14.4	12.5	2.6	5.6	14.4	9.5	8.9	2.3	2.0
28	11.5	14.3	11.7	3.1	4.7	13.9	8.2	9.2	2.4	1.9
32	8.2	15.4	15.0	3.2	5.1	12.5	8.7	9.5	3.2	2.0
60	8.3	15.9	12.9	3.1	5.1	12.7	9.0	9.5	2.6	1.8
120	8.9	15.5	13.1	3.8	5.7	13.3		9.3	2.9	1.8
180	7.8	15.6	14.1	3.9	5.3	13.0	9.6	8.8	2.7	2.1
240	8.5	16.2	13.6	4.0	4.7	13.8	10.1	9.2	2.7	1.7

Table S4. Data of the dissolution kinetics in SELF for Fe reported as concentrations in air (ng/m³).

Sample ID	24 B	26 B	28 B	16 +18 B	32 B	22 B	20 B	30 B	12+14 B	38 + 40 B
Time (min)										
0	36.7	40.6	26.1	0.00	28.1	40.6	20.9	20.3	2.0	0.00
1	41.2	49.2	25.4	0.00	30.8	34.3	17.8	19.9	3.4	1.1
2	41.8	46.5	19.4	1.4	20.8	43.6	20.9	22.7	2.7	2.0
3	50.7	43.9	30.9	0.8	24.6	45.3	17.5	21.6	2.8	3.2
4	46.4	53.8	24.3	9.6	30.5	44.6	23.4	18.7	2.6	3.3
5	52.4	50.4	29.2	4.5	25.3	46.0	18.3	26.6	4.3	1.3
6	45.2	53.4	31.5	9.1	30.5	53.2	18.8	19.3	4.7	1.6
8	60.1	61.6	30.9	7.0	22.3	46.5	26.0	22.5		1.2
10	53.8	60.8	31.1	6.9		40.7	23.6	21.5	4.9	2.1
12	38.4	63.1	30.8	9.9	33.7	45.8	23.9	24.3	5.0	2.2
14	43.8	42.4	31.6	9.6	41.6	45.1	22.0	20.3		2.1
16	61.1	56.1	31.8	9.8	27.4	47.5	22.9	24.7		1.7
20	47.3	55.5	26.7	9.2	33.1	50.3	24.2	23.5	4.5	1.6
24	47.7	48.3	33.4	6.5	37.4	48.2	22.3	22.4	4.5	0.58
28	41.0	53.1	32.1	9.5	35.5	48.8	20.3	24.4	3.7	1.3
32	49.9	69.0	35.4	10.2	34.0	45.1	21.4	24.0	5.1	1.3
60	43.9	55.5	35.1	6.5	34.8	51.4	20.2	27.4	5.8	0.5
120	37.4	41.5	34.7	7.7	25.4	36.1	17.6	17.6	4.4	0.6
180	23.0	40.9	26.1	7.7	18.0	32.0	13.2	12.2	2.1	0.6
240	19.9	29.2	26.6	8.9	33.7	29.7	15.6	11.1	1.5	0.5

Table S5. Data of the dissolution kinetics in SELF for Mn reported as concentrations in air (ng/m³).

Sample ID	24 B	26 B	28 B	16 +18 B	32 B	22 B	20 B	30 B	12+14 B	38 + 40 B
Time (min)										
0	3.4	6.1	7.7	1.3	2.6	6.3	5.5	2.7	1.3	0.39
1	6.6	8.3	7.2	1.4	2.5	4.7	6.6	2.6	1.4	0.67
2	5.2	8.7	3.7	2.6	1.7	6.0	6.9	3.0	1.9	0.76
3	5.6	8.3	9.1	2.5	2.1	5.6	5.4	2.9	1.2	0.87
4	6.6	9.2	7.1	2.2	3.4	4.4	5.7	3.2	1.2	0.63
5	7.3	7.8	2.2	3.4	3.2	6.5	6.0	2.8	2.0	0.58
6	7.3	8.5	7.7	3.2	3.1	7.8	5.6	2.8	1.8	0.45
8	8.6	8.9	6.4	2.7	2.3	5.4	6.3	2.6	1.2	0.62
10	6.7	10.2	8.6	3.0		2.5	6.3	4.3	1.9	0.50
12	4.2	10.0	8.1	2.8	4.5	5.4	6.6	3.5	1.3	0.54
14	5.9	7.0	7.2	3.1	3.6	6.7	6.8	2.2	1.5	0.57
16	6.7	8.1	8.2	3.3	3.4	5.7	5.8	3.4	1.1	0.63
20	6.6	9.3	5.3	2.9	2.4	6.3	6.4	3.0	1.7	0.46
24	5.6	9.1	8.1	2.9	3.6	6.2	6.6	2.8	1.5	0.20
28	5.6	8.9	9.1	2.8	3.0	6.8	6.2	3.2	1.6	0.27
32	6.5	9.2	7.9	3.3	3.6	5.8	5.8	3.1	1.6	0.26
60	4.4	6.9	6.9	1.8	3.1	5.9	5.7	3.0	1.0	0.17
120	2.0	2.9	2.3	0.88	1.2	2.1	2.0	0.98	0.48	0.16
180	2.0	2.9	2.3	0.88	1.2	1.9	2.0	0.98	0.48	0.16
240	2.0	2.9	2.3	0.88	1.3	1.9	2.0	0.98	0.48	0.16

Table S6. Data of the dissolution kinetics in SELF for Pb reported as concentrations in air (ng/m^3).

Sample ID	24 B	26 B	28 B	16 +18 B	32 B	22 B	20 B	30 B	12+14 B	38 + 40 B
Time (min)										
0	1.1	4.1	2.6	0.00	1.3	5.0	2.2	2.2	0.71	0.15
1	1.8	3.1	2.7	0.16	0.80	3.9	2.5	2.7	0.76	0.33
2	1.9	4.6	1.7	0.45	1.1	5.4	2.7	3.0	0.89	0.57
3	2.7	4.4	3.6	0.67	1.7	5.6	2.6	3.9	0.92	0.73
4	3.8	6.0	3.1	1.3	2.5	5.1	3.2	4.4	0.87	0.72
5	4.3	5.6	1.5	1.4	3.2	7.4	3.2	4.7	1.7	0.71
6	4.8	7.4	4.2	1.4	4.3	8.3	3.6	5.0	1.1	0.77
8	6.9	8.9	3.5	1.4	3.7	7.7	4.6	5.3	1.2	0.88
10	5.9	9.4	4.3	1.7		4.4	4.3	4.9	1.6	0.83
12	4.2	9.9	5.4	1.7	6.9	8.0	4.7	6.3	1.2	0.80
14	5.6	7.5	4.8	1.8	5.9	9.6	4.6	4.1	1.4	0.80
16	6.4	8.1	4.6	2.2	5.1	9.8	4.7	6.3		0.92
20	6.6	9.0	3.8	2.0	4.3	10.1	5.9	6.0	1.4	0.77
24	6.5	8.6	5.0	2.2	6.6	10.9	5.4	5.5	1.5	0.65
28	7.2	9.5	5.8	1.9	5.5	11.1	5.4	6.1	1.4	0.47
32	6.5	9.6	5.4	2.4	6.2	10.2	5.4	6.4	1.5	0.57
60	5.6	7.1	4.8	1.5	5.9	9.8	5.9	6.5	1.1	0.71
120	3.7	4.6	3.0	1.1	3.4	5.1	2.7	2.9	0.74	0.37
180	2.6	4.7	2.7	0.8	2.6	4.6	2.3	2.4	0.57	0.32
240	2.1	4.1	2.2	0.7	3.3	4.3	2.1	2.3	0.50	0.23

Table S7. Data of the dissolution kinetics in SELF for Sn reported as concentrations in air (ng/m^3).

Sample ID	24 B	26 B	28 B	16 +18 B	32 B	22 B	20 B	30 B	12+14 B	38 + 40 B
Time (min)										
0	2.9		1.2	0.00	2.9	2.4		3.8	0.69	2.2
1	4.6	6.2	1.7	0.00	3.1	3.3	5.3	3.4	0.74	1.4
2	4.7	5.9	2.2	0.00	2.7	3.4	5.8	4.1	0.88	1.3
3	4.8	5.4	2.0	0.24	3.1	3.5	4.7	3.6	0.84	1.8
4	5.6	5.6	2.0	1.1	3.1	3.6	4.5	4.1	0.74	1.3
5	4.9	5.5	2.6	0.89	3.1	3.7	4.6	3.5	0.87	1.3
6	4.8	7.3	2.1	1.1	3.3	3.8	4.7	3.9	0.81	1.4
8	5.5	5.4	2.2	1.1	3.1	3.5	5.7	4.1	0.81	1.3
10	4.8	5.7	2.2	0.92		3.6	7.8	3.3	0.87	1.4
12	4.6	5.5	2.3	1.5	3.7	3.5	4.6	3.6	0.86	1.4
14	4.7	5.2	2.3	1.2	3.5	3.3	4.8	3.4	0.78	1.3
16	5.2	5.3	2.2	1.3	3.2	3.5	4.6	4.0		1.3
20	5.5	5.9	2.2	1.2	3.2	4.0	6.9	4.3	0.81	1.3
24	5.0	8.4	2.2	1.0	3.5	3.8	4.4	3.4	0.90	1.3
28	4.6		2.3	1.3	3.3	3.9	4.3	3.4	0.93	1.4
32	5.0	5.9	2.3	1.2	3.6	3.8	4.4	3.7	0.95	1.4
60	4.7	5.6	2.3	1.4	3.6	3.8	4.5	3.8	0.87	1.5
120	4.3	4.6	2.2	1.5	3.2	3.3	4.5	3.3	0.87	1.4
180	3.8	4.5	2.1	1.6	2.8	3.1	4.3	3.1	0.82	1.4
240	3.8	4.4	2.2	1.7	2.7	3.2	4.3	3.0	0.81	1.4

Table S8. Data of the dissolution kinetics in SELF for Sn reported as concentrations in air (ng/m^3).

Sample ID	24 B	26 B	28 B	16 +18 B	32 B	22 B	20 B	30 B	12+14 B	38 + 40 B
Time (min)										
0	37.4	73.6	103.7		61.8	73.4	58.2		10.0	0.43
1	51.1	74.6	70.4			65.2	51.3	16.1	11.7	0.18
2	42.4		74.1		17.7	89.6	66.9	18.0	10.0	4.7
3	46.9	74.0	138.9		24.4	74.6	55.6	33.8	19.0	5.9
4	60.0	81.8	97.8		35.6	99.4	50.4	32.6	8.9	1.7
5	50.4	79.1	42.1		62.9	89.7	38.9	26.3	14.2	3.6
6	61.8	92.4	98.4		23.0	124.7	46.0	25.7	22.3	1.6
8	74.5	99.5	94.7		32.7	106.6	71.4	34.2	12.2	2.5
10	53.1	83.9	117.8			116.8	55.9	30.8	17.4	3.0
12	48.9	84.7	113.9		65.0	100.5	60.1	27.8	14.0	3.1
14	57.5	69.1	112.7		69.0	109.0	57.3	25.9	18.6	4.4
16	58.8	86.8	118.1		53.5	99.7	56.7	38.9		2.6
20	64.1	84.2	120.8		51.4	105.0	68.2	34.0	17.1	1.5
24	47.9	72.3	121.3		65.8	107.8	63.3	35.6	17.0	2.3
28	67.2	78.8	107.6		47.2	102.4	52.8	36.7	15.6	3.3
32	52.2	92.4	139.0		62.4	99.0	53.6	38.8	20.7	4.2
60	52.7	82.2	136.4		42.5	92.4	63.1	39.4	8.9	0.90
120	30.0	51.2	84.0		43.6	71.2	28.0	16.8	7.5	0.90
180	20.3	45.7	82.2		53.6	55.9	25.5	13.3	4.8	0.90
240	21.4	33.9	62.8		59.6	52.6	24.1	11.4	4.8	0.90

Results of the fitting of the kinetics profiles in SELF for all elements and in fog for Pb

Table S9. Results of the fitting of the first-order leaching kinetics for Cu in SELF, together with the results of the time-dependent significance test.

Sample ID	C_e	A	k	$t_{1/2}$	X^2 (fit)	Adj. r^2 (fit)	p (ξ)
24 B	9.1	4.0	0.69	1.01	1.12	0.43	0.416
26 B	15.3	3.5	0.29	2.42	0.63	0.60	**0.005**
28 B	12.8	5.7	0.40	1.74	2.25	0.48	**0.001**
16 & 18 B	3.4	3.0	0.22	3.12	0.23	0.79	**<0.001**
32 B	5.3	1.7	0.08	8.87	1.01	0.19	0.095
22 B	13.1	3.6	0.43	1.60	0.81	0.51	0.122
20 B	9.4	1.9	0.08	8.92	0.32	0.54	**0.003**
30 B	9.1	3.0	0.22	3.14	0.11	0.88	**<0.001**
12 & 14 B	2.7	2.1	0.21	3.28	0.10	0.80	**<0.001**
38 & 40 B	2.0	1.0	2.03	0.34	0.05	0.51	0.299

Values reported in bold red refers to $P < 0.05$. Concentrations are reported in nanograms per cubic metre in air, times in minutes, and kinetics constants in per minutes.

Table S10. Results of the fitting of the non-monotonic kinetics for Fe in SELF, together with the results of the time-dependent significance test.

Sample ID	C_e	t_c	C_c-C_e	C_0-C_e	k_g	k_d	$t_{g1/2}$	$t_{d1/2}$	χ^2 (fit)	Adj. r^2 (fit)	p (ξ)
24 B	19.9	16	41.2	24.5	0.11	0.016	6.1	44.3	72.08	0.32	0.063
26 B	29.2	32	39.8	28.5	0.052	0.012	13.4	56.6	64.99	0.27	**0.022**
28 B[A]											**0.006**
16 & 18 B[B]	8.7			10.2	0.27		2.6		3.84	0.60	0.061
32 B[B]	31.7			6.4	0.15		4.8		36.82	1.60	0.196
22 B	29.7	6	23.5	12.7	0.31	0.011	2.2	61.8	34.39	0.16	**0.020**
20 B	15.5	8	10.5	5.1	0.22	0.027	3.2	25.9	11.10	-0.08	**0.022**
30 B	10.4	60	17.0	7.1	0.034	0.016	20.3	43.0	4.30	0.75	0.246
12 & 14 B	1.5	60	4.3	4.0	0.018	0.012	37.8	59.3	0.63	0.62	**0.003**
38 & 40 B	0.52	4	2.8	3.3	0.54	0.091	1.3	7.6	0.46	0.40	**0.004**

Values reported in bold red refers to $P < 0.05$. Concentrations are reported in nanograms per cubic metre in air, times in minutes, and kinetics constants per minutes.

[A]Fitting failed.

[B]First-order kinetics.

Table S11. Results of the fitting of the non-monotonic kinetics for Mn in SELF, together with the results of the time-dependent significance test.

Sample ID	C_e	t_c	C_c-C_e	C_0-C_e	k_g	k_d	$t_{g1/2}$	$t_{d1/2}$	χ^2 (fit)	Adj. r^2 (fit)	p (ξ)
24 B	2.0	8	6.6	5.1	0.22	0.031	3.2	22.2	1.49	0.58	**0.002**
26 B	2.9	10	7.4	4.3	0.15	0.016	4.8	43.2	1.08	0.79	**0.011**
28 B	2.3	28	6.8	7.0	0.16	0.021	4.5	32.9	5.67	0.04	0.370
16 & 18 B	0.88	5	2.5	2.2	0.37	0.015	1.9	47.4	0.18	0.76	**0.002**
32 B	1.3	12	3.2	3.0	0.13	0.028	5.5	25.0	0.63	0.28	0.054
22 B[A]	1.9	6	5.9	6.0	-	0.039	<1	17.7	5.55	-0.82	0.110
20 B	2.0	2	4.9	1.4	1.51	0.011	0.46	62.1	0.55	0.78	**0.007**
30 B[A]	1.0	10	3.4	2.0	-	0.16	<1	4.2	1.89	-1.56	0.124
12 & 14 B[A]	0.48	5	1.5	0.75	-	0.041	<1	16.7	0.19	0.07	0.188
38 & 40 B	0.16	3	0.71	0.49	0.79	0.073	0.88	9.5	0.01	0.74	**<0.001**

Values reported in bold red refers to $P < 0.05$. Concentrations are reported in nanograms per cubic metre in air, times in minutes, and kinetics constants in per minute.

[A]Fast leaching with $t_{1/2} < 1$ min.

Table S12. Results of the fitting of the non-monotonic kinetics for Pb in SELF, together with the results of the time-dependent significance test.

Sample ID	C_e	t_c	C_c-C_e	C_0-C_e	k_g	k_d	$t_{g1/2}$	$t_{d1/2}$	χ^2 (fit)	Adj. r^2 (fit)	p (ξ)
24 B	1.9	28	5.2	6.2	0.13	0.012	5.5	55.7	0.46	0.88	**<0.001**
26 B	4.1	12	5.7	6.0	0.17	0.015	4.2	47.5	2.01	0.60	**<0.001**
28 B	2.2	28	3.6	3.2	0.08	0.014	8.9	48.1	0.87	0.47	**0.002**
16 & 18 B	0.72	32	1.6	2.5	0.13	0.021	5.2	32.7	0.04	0.92	**<0.001**
32 B	3.3	12	3.6	6.0	0.16	0.020	4.4	34.4	2.36	0.38	**<0.001**
22 B	4.3	28	6.8	7.0	0.073	0.015	9.4	44.9	1.62	0.74	**<0.001**
20 B	2.1	20	3.8	6.3	0.046	0.011	15.1	60.6	0.21	0.88	**<0.001**
30 B	2.3	60	4.3	4.3	0.13	0.031	5.4	22.2	0.29	0.88	**<0.001**
12 & 14 B	0.50	5	1.2	1.0	0.34	0.014	2.0	48.8	0.13	0.04	**0.003**
38 & 40 B	0.23	16	0.69	0.77	0.32	0.030	2.2	23.4	0.01	0.74	**<0.001**

Values reported in bold red refers to $P < 0.05$.

Table S13. Results of the fitting of the non-monotonic kinetics for Pb in fog, together with the results of the time-dependent significance test.

Sample ID	C_e	t_c	C_c-C_e	C_0-C_e	k_g	k_d	$t_{g1/2}$	$t_{d1/2}$	χ^2 (fit)	Adj. r^2 (fit)
24 A	7.5	12	0.10	5.5	0.77	292	0.9	0.0024	0.35	0.83
26 A	4.8	2	2.2	4.2	9.8	0.055	0.1	12.6	0.01	0.99
28 A	3.4	0.01	1.1	2.4	146	0.073	0.0	9.5	0.01	0.93
16 & 18 A	5.4	20	0.23	2.9	0.62	0.021	1.1	32.9	0.02	0.96
32 A[A]	4.4			2.9	0.081		8.5		0.10	0.43
22 A	6.1	2	2.2	4.7	3.2	0.054	0.2	12.7	0.01	0.99
20 A[A]	5.1			1.3	0.019		36.4		0.06	1.43
30 A[A]	2.4			-0.87	0.064		10.8		0.00	4.43
12 & 14 A[A]	5.3			2.3	0.065		10.7		0.10	2.43
38 & 40 A[A]	2.4			1.0	0.70		1.0		0.01	3.43

Values reported in bold red refers to $P < 0.05$.

[A]First-order kinetics.

Table S14. Results of the fitting of the non-monotonic kinetics for Sn in SELF, together with the results of the time-dependent significance test.

Sample ID	C_e	t_c	C_c-C_e	C_0-C_e	k_g	k_d	$t_{g1/2}$	$t_{d1/2}$	χ^2 (fit)	Adj. r^2 (fit)	p (ξ)
24 B	3.8	4	1.8	2.7	0.56	0.023	1.2	30.8	0.19	0.54	**0.032**
26 B	4.4	24	4.0	2.9	0.069	0.10	10.1	7.2	2.26	-1.56	**0.038**
28 B[A]	2.2			1.0	0.81		0.86		0.02	0.43	0.085
16 & 18 B[A]	1.4			1.6	0.20		3.4		0.06	0.60	**<0.001**
32 B	2.7	12	1.0	0.83	0.15	0.010	4.6	71.8	0.09	-0.08	**0.013**
22 B[A]	3.2	20	0.80	1.6	0.38	0.017	1.8	40.9	0.08	0.32	**0.001**
20 B	4.3	10	3.5	11.1	0.61	1.06	1.1	0.66	2.86	-2.14	**0.008**
30 B[A]	3.0	20	1.3	0.53	0.089	0.14	7.8	5.1	0.21	-0.59	0.299
12 & 14 B	0.81	32	0.14	0.26	0.040	0.017	17.3	41.8	0.00	0.38	0.077
38&40 B[B]											0.376

Values reported in bold red refers to $P < 0.05$. Concentrations are reported in nanograms per cubic metre in air, times in minutes, and kinetics constants in per minute.

[A]First-order kinetics

[B]Fitting failed.

Table S15. Results of the fitting of the non-monotonic kinetics for Zn in SELF, together with the results of the time-dependent significance test.

Sample ID	C_e	t_c	C_c-C_e	C_0-C_e	k_g	k_d	$t_{g1/2}$	$t_{d1/2}$	χ^2 (fit)	Adj. r^2 (fit)	p (ξ)
24 B	21	8	53	38	0.22	0.020	3.2	34.5	130.50	0.35	0.077
26 B	34	8	66	25	-	0.022	<1	31.3	253.35	0.06	**0.030**
28 B	63	32	76	35	0.068	0.011	10.1	64.7	742.40	-0.07	**0.032**
16 & 18 B[A]											
32 B[B]	55			30	0.13		5.4		289.51	0.43	0.151
22 B[C]	52	6	73	60	-	0.020	<1	34.2	574.62	-0.43	**0.001**
20 B	24	8	48	25	0.15	0.015	4.6	46.9	142.15	0.24	0.069
30 B	12	60	25	18	0.12	0.027	6.0	25.3	24.77	0.70	**0.002**
12&14 B[C]	4.7	6	18	12	-	0.025	<1	27.6	43.55	-0.59	0.067
38&40 B[C]	0.90	3	5.0	6.0	-	0.038	<1	18.2	5.26	-1.10	**0.019**

Values reported in bold red refers to $P < 0.05$. Concentrations are reported in nanograms per cubic metre in air, times in minutes, and kinetics constants in per minute.

[A]Data not available.

[B]First-order kinetics.

[C]Fast leaching with $t_{1/2} < 1$ min.

Details of samples collected

Table S16. List of samples with details of collection date and mass of $PM_{2.5}$ collected for each sample.

Sample ID (supplementary)	Sample ID (main text)	Collection date	Mass of $PM_{2.5}$ on filter (mg)
12 B	S12	1 Feb 2019	0.92
14 B	S14	5 Feb 2019	1.27
16 B	S16	8 Feb 2019	1.79
18 B	S18	12 Feb 2019	1.06
20 B	S20	14 Feb 2019	2.36
22 B	S22	18 Feb 2019	-
24 B	S24	20 Feb 2019	3.82
26 B	S26	22 Feb 2019	3.57
28 B	S28	26 Feb 2019	3.19
30 B	S30	28 Feb 2019	2.28
32 B	S32	4 Mar 2019	2.51
38 B	S38	12 Mar 2019	0.39
40 B	S40	14 Mar 2019	1.44