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Electronic Supplementary Information

The Presence and Leachability of Antimony in different Wastes and Waste Handling Facilities in Norway

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S1. Field Campaign and extra data of waste and waste facility samples analysed for Sb

Table S1.1. Details regarding the type of the solid waste, leachate and air samples collected for Sb analysis as part of the sampling campaign.

Site ***Date of campaign***	**Compartment sampled**	**Type of solid sample (number)** ***Collection date***	**Number leachate grab samples[a]:** ***Collection date***	**Number leachate DGT samples[b]:** ***Exposure dates***	**Number air/dust samples[c]:** ***Exposure dates***	**Additional Information**
Landfill A ***25/06/13-07/10/13*** ***07/10/13-03/12/13*** ***27/03/14-23/06/14***	Solid, leachate	Bottom ash (n=5), Vehicule fluff (n=2) *25/06/13*	2 1: *02/07/2013* 1: *27/03/14*	6 3: *26/06/13-02/07/13*[g] 3: *27/03/14-03/04/14*[g]	e	Solid ground and sieved ≤2 mm Water: Leachate effluent
Landfill B ***09/07/13-15/10/13*** ***15/10/13-10/12/13*** ***01/04/14-18/06/14***	Solid, leachate	Fly ash (n=5) *10/12/13*	3 1: *09/07/2013* 1: *15/10/2013* 1: *01/04/2014*	9 3: *09/07/13-16/07/13*[g] 3: *15/10/13-22/10/13*[g] 3: 01/04/14-08/04/14[g]	e	Solid ground and sieved ≤2 mm Water: Pond
Landfill C ***28/08/13-10/12/13*** ***01/04/14-18/06/14***	Solid, leachate	Digestate (n=6) *22/10/13*	3 2: *28/08/2013* 1: *01/04/14*	6 3: *28/08/13-02/09/13*[g] 3: *01/04/14-08/04/14*[g]	e	Solid ground and sieved ≤2 mm Water: Leachate effluent
WEEE/Vehicle A ***27/06/13-10/10/13*** ***10/10/13-04/12/13*** ***26/03/14-16/06/14***	Solid, leachate,	Glass (n=6), Vehicule fluff/Plastic (n=3) *27/06/13*	3 1: 04/*07/2013* 1: *10/10/2013* 1: 26/*03/2014*	3 1: *27/06/13-04/07/13* 1: *10/10/13-17/10/13* 1: *26/03/14-02/04/14*[g]	e	Solid ground and sieved ≤4 mm Water: Manhole effluent
WEEE/Vehicle B ***05/09/13-09/04/14*** ***09/04/14-23/06/14***	Solid, leachate, air chimney	WEEE (n=2), Plastic (n=6), WEEE/Plastic (n=4), Vehicule fluff/Plastic (n=3) *05/09/13*	2 1: *05/09/2013* 1: *09/04/14*	6 3: *05/09/13-12/09/13*[g] 3: *09/04/14-15/04/14*[g]	Chimney Vent sample 1:03/10/2013 2:24/04/2014	Solid ground and sieved ≤4 mm Water: Leachate effluent
WEEE/Vehicle C ***03/09/13-03/12/13*** ***27/03/14-23/06/14***	Solid, leachate,	d	2 2: *03/09/2013*	2 1: *03/09/13-11/09/13*[g] 1: 27/03/14-02/04/14[g]	e	Water: Manhole effluent

Site *Date of campaign*	Compartment sampled	Type of solid sample (number) *Collection date*	Number leachate grab samples[a]: *Collection date*	Number leachate DGT samples[b]: *Exposure dates*	Number air/dust samples[c]: *Exposure dates*	Additional Information
WEEE/Vehicle D ***13/09/13-10/12/13*** ***25/03/14-18/06/14***	Solid, leachate,	WEEE (n=10) *13/09/13*	2 1: 13*/09/2013* 1: 2*5/03/14*	1 (2 lost) 1: 13/09/13-20/09/13[g]	e	Solid ground and sieved ≤2 mm Water: Manhole effluent
WEEE/Vehicle E ***27/06/13-10/10/13*** ***10/10/13-04/12/13*** ***26/03/14-16/06/14***	Solid	Vehicule fluff (n=2), Vehicule fluff/Plastic (n=2) *27/06/13*	e	e	e	Solid ground and sieved ≤4 mm
Incineration/Sorting A ***20/06/13-03/10/13*** ***03/10/13-05/12/13*** ***25/03/14-16/06/14***	Solid, air	Combustible (n=5) *20/06/13*	e	e	Hivol PM10 sample 1 (+1 Field blank: 25/06/14) 1: *12/06/14-13/06/14*	Solid ground and sieved ≤4 mm AAS: Inside the facility
Incineration/Sorting B ***05/07/13-03/10/13*** ***03/10/13-05/12/13*** ***25/03/14-17/06/14***	Solid, air chimney	d	e	e	Chimney Vent Sample 1. 25/06/2013 2. 20/09/2013	

a) sampled by submerging 200 mL water bottles in the leachate stream; b) sampled by submerging DGT units for one week; c) Air sample either a PM10 sample using a hivol or a chimney filter sample, d) no solid waste collected as similar waste sampled in other facilities, e) no sample for Sb analysis due to logistical reasons.

Table S1.2. Extra Sb Analysis data for unique waste fractions, including the concentration of Sb in solid waste materials (mg/kg), and the concentrations of dissolved Sb(TOT) and Sb(III) when exposing the waste fraction to a 1/10 ratio of water as part of the Leach Test.

Group	Type	Solid $C_{waste,Sb}$				$C_{leachable,Sb(Tot)}$				$C_{leachable,Sb(III)}$			
		median mg/kg	max mg/kg	min mg/kg	n	median µg/kg	max µg/kg	min µg/kg	n	median µg/kg	max µg/kg	min µg/kg	n
Glass	Recycled glass	33			1	20	22	20	3	0.05	0.53	0.05	3
	Composite glass	29	33	21	3	20	33	19	9	0.2	2.1	0.0	9
Vehicle	Coarse Fluff	360	4565	61	6	155	180	97	6	6.6	7.3	5.3	6
	Fine Fluff	145	256	34	2	110	130	78	6	0.6	1.1	0.4	6
WEEE	BFR plastic	4600			1	404	427	380	2	10.6			2
	Cable Plastic	640	823	150	6	385	842	210	9	16.1	58.1	0.0	9
	Remaining plastic	1476	1715	1238	8	1550	2000	600	6	6.3	8.6	3.4	6
Plastic	Packaging plastic	1476	1715	1238	8	1550	2000	600	6	6.3	8.6	3.4	6
Combustibles	Coarse	11			1	35	50	33	3	5.8	7.9	4.3	3
	Fines	3			1	66	79	40	3	0.3	0.3	0.3	3
Bottom Ash	Coarse	84			1	640	710	530	3	0.6	0.6	0.6	3
	Fine	96			1	680	770	410	3	1.1	1.1	1.0	3
Flyash		676	1016	39	5	12	44	5	18	0.4	0.7	0.2	18
Digestate		3	4	3	2	7	9	7	2	0.8	0.8	0.8	2

S2. Extra DGT Sampling Details and Results

Calculating the DGT labile Sb concentration

Based on the mass of elements accumulated in the resin and the Fick's first law of diffusion, the time averaged C_{DGT} concentration can be calculated using equation S4:

$$C_{DGT} = \frac{(m(t)/f)\Delta g}{DAt} \qquad \text{(S4)}$$

Where the *m(t)* is the time integrated uptake in the resin (µg), *f* is an elution factor for each element (typically 0.8), and the Δg is the thickness of the diffusive gel (mm) plus the thickness of the filter membrane. The *D* is the ion-specific diffusion coefficient ($cm^2\ s^{-1}$) in the gel, *A* is the area of the gel exposed (cm^2) whereas *t* is the time of exposure (s).

The specific diffusion coefficients were corrected for average temperature logged during DGT exposure. The diffusion coefficient (*D*) in water depends on viscosity (η) and temperature (*T*) as expressed in the Stoke-Einstein relationship:

$$\frac{D_t \eta_t}{T_t} = \frac{D_0 \eta_0}{T_0} \qquad \text{(S5)}$$

The viscosity is corrected for temperature using equation 3, as reported in Garmo et al.[1]:

$$log\frac{\eta_0}{\eta_t} = \frac{1.37023(t-25) + 0.000836(t-25)^2}{109+t} \qquad \text{(S6)}$$

A combination of equations 1 and 2 (eq. 4) provides the temperature correction relative to 25°C.

$$D = \left(0.0001947 * T^2 + 0{,}01716 * T + 0{,}4492\right) * D_{25°\mathrm{C}} \qquad \text{(S7)}$$

Where *T* is the recorder temperature. The different *D* constants for metal cations (normalised at 25°C) are obtained from DGT research (www.dgtresearch.com), whereas $D_{25°C}$ constants for Sb were obtained from Luo et al.[2]

Sampling Dates and DGT concentrations from individual sampling campaigns

Table S2.1. Comparing of fluctuations in DGT sampling from the different field campaigns

Landfill	sampling campaigns	Date of campaign	DGT $Cl_{eachate,Sb}$ (µg/L)	n
Landfill A	1st.	25/06/13-07/10/13	<LOD	3
	3rd	27/03/14-23/06/14	0.05 ± 0.00	3
Landfill B	1st	09/07/13-15/10/13	3.65 ± 0.23	3
	2nd	15/10/13-10/12/13	3.74 ± 0.66	3
	3rd	01/04/14-18/06/14	22.40 ± 1.91	3
Landfill C	1st	28/08/13-10/12/13	<LOD	3
	2nd	01/04/14-18/06/14	0.53	3
WEEE/Vehicle A	1st	27/06/13-10/10/13	0.66	1
	2nd	10/10/13-04/12/13	0.33 (Not used due to high pH[c])	1
	3rd [a]	26/03/14-16/06/14	0.14	1
WEEE/Vehicle B	1st	05/09/13-09/04/14	<LOD	3
	2nd	09/04/14-23/06/14	0.74 ± 0.13	3
WEEE/Vehicle C	1st	03/09/13-03/12/13	0.28	1
	2nd	25/03/14-18/06/14	0.32	1
WEEE Vehicle D	1st [b]	13/09/13-10/12/13	0.53	1

a) Activity partially stopped at this facility some months before sampling
b) The value 0.53 is the maximum, not average, as the other two replicates were below the limit of detection.
c) the DGT sampling data was not used for this time point, as the measured pH was over 8 (see Table S2.2)

Measured parameters in the Leachate Water

Table S2.2. Important parameters analysed in the leachate water. One grab sample was collected at each sampling campaign.

Landfill	sampling campaigns	Date of campaign	pH	Fe-W[a]	DOC	F	Cl^{-}	SO_4^{2-}
						mg/l		
Landfill A	1st	26.06.2013	6.8	1.1	39	0.1	280	100
	3rd	27.03.2014	5.6	0.1	6	0.1	47	9.4
Landfill B	1st	09.07.2013	6.9	13	-	-	-	-
	2nd	15.10.2013	6.2	2.8	4.6	0.1	13300	2330
	3rd	01.04.2014	7.1	1	4.8	0.8	13300	1050
Landfill C	1st	28.08.2013	7.0	35	350	1.3	310	73
	2nd	01.04.2014	7.0	34	380	0.6	260	7.1
WEEE/Vehicle A	1st	27.06.2013	7.2	4	-	-	-	-
	2nd b)	*10.10.2013*	*8.4 b)*	*16*	*14*	*0.4*	*130*	*330*
	3rd	26.03.2014	7.3	7.2	38	0.5	95	14
WEEE/Vehicle B	1st	05.09.2013	7.2	1.6	12	0.1	79	13
	2nd	09.04.2014	7.2	2.4	14	0.1	130	54
WEEE/Vehicle C	1st	03.09.2013	7.2	22.5	16	1	91	30
	2nd	27.03.2014	7.5	22.5	-	-	-	-
WEEE Vehicle D	1st b	13.09.2013	7.23	3.2	8.3	0.6	220	13
	2nd	25.03.2014	7.7	1.7	12	0.4	400	140

a) Fe-W is the total concentration of Fe in leachate water

b) the DGT sampling data was not used for this time point, as over a pH 8 the binging of Sb to Fe oxide in the DGT can diminish.

Table S2.3. Comparison of grab sample leachate Sb and DGT sampling Sb, along with leachate pH and DOC content

Location	$C_{leachate}$ Sb (µg/L)	n	$C_{leachate\text{-}DGT}$ Sb (µg/L)	n	pH	DOC (mg/L)
Landfills (lit.)						
Landfill A	0.5 ± 0.3	2	0.05 ± 0.00	3[a]	6.2	23
Landfill B	26.0±16.1	3	9.93 ± 9.41	9	6.7	5
Landfill C	2.8 ± 1.8	3	0.19 ± 0.09	3[a]	7	365
WEEE/Vehicle A	13.6 ± 8.2	2	0.40 ± 0.37	2	7.3	38
WEEE/Vehicle B	3.6 ± 3.2	2	0.74 ± 0.13	3[a]	7.2	13
WEEE/Vehicle C	50.0 ± 0.0	2	0.30 ± 0.02	2	7.4	16
WEEE/Vehicle D	3.3 ± 0.2	2	0.53	1	7.5	10

[a]: n total=6 but 3 were <LOD

Illustration of a DGT sampler

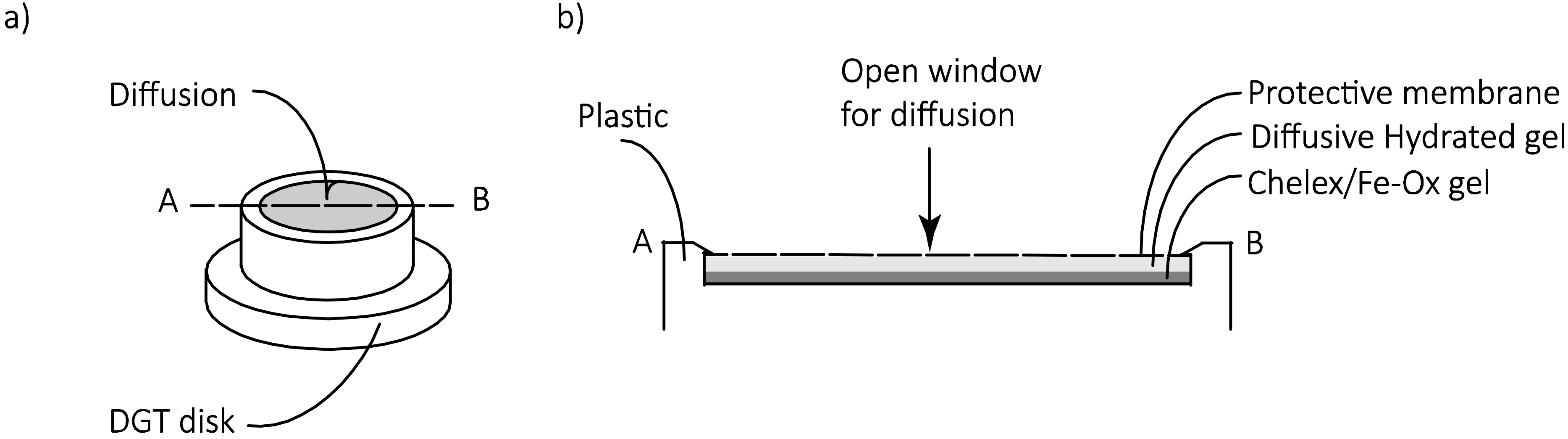

Figure S2.1. The illustration shows geometry of the DGT disk. The grey area indicates the open window where trace elements can diffuse through (panel a). A cross section from A to B is indicated and the composition of gel-layers is shown in panel b. After deployment, the devices are dismantled and the trace elements accumulated in the Chelex or Fe-Gel is extracted using a strong acid in lab.

S3. Extra Air Sampling Method Details and Results.

Sampling of ambient particulate matter was done using a high-volume (HighVol) air sampler (Digitel, Switzerland), equipped with a 10 µm cutoff and 150 mm Ø glass fiber filter (GF filter, Sigma Aldrich, USA) to collect the PM10 particles (i.e. particles with a 10 µm aerodynamic diameter or smaller). GFF filters were cleaned by solvent rinse (cyclopentane) and baking overnight, and then isolated in aluminum foil prior to use. The initial air sampling speed was set to approximately 500 L/min. The total volume sampled as well as ambient temperature and pressure were recorded continuously.

From the difference in the weight of the GFF before after sampling with the HiVol air sampler, M_{sample}, minus the weight of the blank GFF, M_{filter}, the weight of PM10 ($\mu g_{particles}/m^3_{air}$) could be determined.

$$PM10 = (M_{sample} - M_{filter})/V_{air} \quad \text{(S1)}$$

Where V_{air} is the volume of ambient air sampled (corrected to standard temperature and pressure). It should be noted that when the GFF is heavily loaded, the particle size cut-off works less efficiently, and particles with an aerodynamic diameter larger than 10µm will begin to accumulate on the GFF.

GFFs were extracted for Sb content as described in the main text to derive particle concentrations, C_{dust} ($\mu g/g_{dust}$):

$$C_{dust} = \text{"mass Sb on GFF"} / (M_{sample} - M_{filter}) \quad \text{(S2)}$$

From this, it is also possible to infer the Sb in air, C_{air} (pg/m^3), because Sb in the air is expected to be associated 100% with particles, using the following relationship for non-volatile compounds

$$C_{air} = C_{dust} * PM10 \text{ (for non-volatile compounds)} \quad \text{(S3)}$$

Table S3.1. Air and dust concentrations of Sb at the vent outlet of a WEEE fragmenting facility, the chimney of a municipal waste incineration plant, as well as dust near an indoor shredder for combustibles

	Sampling Date	PM10 or TSP µg m^{-3}	C_{air} Sb ng m^{-3}	C_{dust},Sb mg kg^{-1}	Source waste	C_{dust}/C_{waste}
WEEE/Vehicle Fragmenting Vent (TSP, n=2)	Oct-2013		2200	6816		
	Apr-2014		<100	<310		
	Average	323[a)]	1150 ± 1485	3563 ± 4601	WEEE	3.3 ± 4.4
Combustable Shredding (PM10, n= 2)	June-2013		25	5		
	June-2014		217	44		
	Average	4929	86 ± 87	17 ± 18	Combustables	1.9 ± 1.9
Combustable Loading Dock (PM10, n= 1)	June-2014	206	9.0	2	Combustables	0.2 ± 0.0
Incineration Chimney (TSP, n=2)	June-2013		<30	<34		
	Sept-2013		100	114		
	Average	880[b)]	65 ± 49	74 ± 56	Fly ash	0.1 ± 0.1

PM10 = particulates with an aerodynamic dynameter of 10 µm or less measured using a high volume air sampler; TSP = total suspended particulates measured by isokinetic suction through a filter at a vent/chimney outlet ; Comb. = combustibles a) the chimney PM10 was obtained with a Hivol in sampler inside the facility, near the sampling vent; b) average of yearly measurements reported by the facility

References to the Supporting Information

1. Ø. A. Garmo, O. Royset, E. Steinnes and T. P. Flaten, *Anal. Chem.*, 2003, **75**, 3573-3580.
2. J. Luo, H. Zhang, J. Santner and W. Davison, *Anal. Chem.*, 2010, **82**, 8903-8909.