Electronic Supplementary Information

1. Sample Preparation and Isotope Standards

Hydrochloric (HCl) and Nitric acid (HNO_3) (Sigma Aldrich Chemie GmbH, Munich Germany) as well as hydrogen fluoride (HF, 48%, Merck, Darmstadt, Germany) were all analytical grade. The MilliQ water (Millipore, Bedford, MA, USA) used was purified by reverse osmosis followed by ion-exchange. The AG MP-1M ion exchange resin (macro porous, 100–200 μm dry mesh size, 75–150 μm wet bead size, Bio-Rad Laboratories AB, Solna, Sweden) was cleaned before use by soaking in 0.7 M HNO_3, followed by rinsing with MilliQ water and loaded into 2 mL columns as a slurry. The following chemicals were used as δ-zero standards: NIST SRM 3108 Cd solution Lot 130116, NIST SRM 976 Cu standard solution, NIST SRM 981 common Pb and IRMM 3702 zinc solution from the Institute of Reference Materials and Measurements, Geel, Belgium. Working solutions of the mentioned standard solutions were prepared daily and diluted to 200 μg L^{-1} in 0.14 M HNO_3. Internal standards were prepared from 1000 mg L^{-1} stock standard, all from Ultra Scientific, and added to all measurement solutions at half of the respective analyte concentration.

All handling of sample and digests were performed in clean laboratory areas (Class 10 000) by personnel wearing clean room gear and following all general precautions to reduce contaminations as described by Rodushkin et al., (2010). All laboratory plastic ware in contact with samples and sample digests was soaked in 0.7 M HNO_3 (24 h at room temperature) and rinsed with MilliQ water prior to use. Teflon beakers (250 mL) were used for evaporation on designated ceramic-top hotplates. For sediments and process samples, 0.025 to 0.2 g of dried, homogenized material was weighed (precision of 0.2% of mass) into 50 mL polypropylene tubes. Then, 5 mL 14 M HNO_3, 4 mL HCl and 0.5 mL HF were added, and samples were placed in a heat block for 4 hours. To ensure digestion of all material, an additional 1 mL 14M HNO_3 was used to wash down adhering sample material after agitation using a vortex mixer. Samples were then centrifuged for 4 minutes at 4000 rpm, and sample digests were decanted into 250 mL high-density Teflon beakers and evaporated on a ceramic-top hotplate. Samples were re-dissolved in concentrated HCl, but before the separation of selected elements, small aliquots of 50 μL were taken for pre-analysis. An inductively coupled plasma double-focusing sector-field mass spectrometer (ICP-SFMS) was used to screen elemental composition and underlying data for recovery calculations. The matrix separation was adopted with only minor modifications from Rodushkin et al., (2016), where sample digests are loaded onto the AG MP-1M column in 4 mL of 9.6 M HCl matrix. Copper, Zn, and Cd were quantitatively eluted as the HCl molarity of the eluent decreased. The resin does not retain Pb, which is flushed through the column and qualitatively recovered during the matrix wash steps. To ensure that no introduction of artificial fractionation occurred, all measurements of samples with recoveries <90% were excluded.

2. Analytical Methods

A single-collector, inductively coupled plasma double-focusing sector-field mass spectrometer (ICP-SFMS, ELEMENT XR, Thermo Fisher Scientific, Bremen, Germany) was used to measure the element concentrations in sample digests and column fractions with uncertainty <15%. The instrument was equipped with a demountable quartz torch, a nickel sampler cone, a high sensitivity X-skimmer cone, a PFA (perfluoroalkoxy alkane) spray chamber, and an SD2 auto-sampler (ESI, Perkin-Elmer, Santa Clara, USA) equipped with a six-port valve and 1.5 mL sample loop filled and rinsed utilizing vacuum suction. For in-depth details on instrumental parameters and measurement conditions see Rodushkin et al. (2005). Isotopic measurements were performed after confirming >90% analyte recovery using ICP-SFMS screening analysis after matrix separation. The MC ICP-MS instrument used for isotope analysis was a Neptune PLUS Thermo Fisher Scientific, Bremen, Germany. The isotopic composition is expressed using the d notation according to Equation (1):

$$\delta^{x/y}Me = [\frac{(^{x/y}Me)_{sample}}{(^{x/y}Me)_{standard}} - 1] \times 1000 \quad (1)$$

where ^{x}Me and ^{y}Me correspond to the two different isotopes of each studied metal, the $(^{x}Me/^{y}Me)_{sample}$ value refers to the measured ratio and $(^{x}Me/^{y}Me)_{standard}$ is the isotope ratio of the corresponding standard. The factor 1000 is used to convert the δ-values to per mil notation.

The instrument was equipped with a platinum guard electrode and nine Faraday cups (eight movable and one fixed center cup) with a coupled introduction system consisting of a cyclonic/Scott spray chamber. Instrumental operating conditions and measurement parameters are summarized in Table S1. After plasma ignition, the instrument was allowed a minimum of one hour to stabilize while aspirating 0.14 M HNO_3 blank solution prior to performing the daily optimization of operational parameters (gas flow, torch position, and lens settings) and mass calibration. All measurement solutions were diluted to equal concentrations, providing concentration matching between samples and bracketing δ-zero standards for each individual run. With a standard introduction system, the analyte-element concentrations in samples were adjusted to 2 mg L^{-1} in 0.14 M HNO_3. Outlier elimination was activated using the 2σ-criterion in the resident Neptune software, and commercially available spreadsheet software was used to calculate signal intensities. The isobaric interference for Cd, Cu, Pb, and Zn was corrected mathematically using monitored isobar signals together with tabulated isotope abundances and computed instrumental mass bias for each individual Neptune session. The measured δ-values for isotope ratios were calculated against the bracketing corresponding standard.

Instrumental mass bias was corrected by using the revised exponential correction model by Baxter et al. using the internal standard, and δ-values were calculated against the bracketing δ-zero solution. The mean value of the two consequent measurements of the sample ratio was calculated against ratios for standards in each block (block: standard 1—sample 1—sample 2—sample 3—standard 2). Assuming a linear change in mass bias, ratios for samples 1 and 3 were calculated relative to those for standards 1 and 3, respectively, while sample 2 was calculated against the mean ratio for both standards. Results from the two measurements were used to calculate mean δ-values for each sample.

3. Quality Assurance

The analytical results for reference standards [GBW 07,312 (GSD-12) (sediment) (National Research Centre of Geoanalysis, Beijing, China), Nod-A1 and Nod-P1 (Fe-Mn nodules) (Branch of Geochemistry, US Geological Survey, 923 National Center, Reston, VA, USA), NASS-6, CASS-5 (seawater) and SLRS-4 (stream water) (National Research Council of Canada, Ottawa, Ontario) are reported in Table S1. All reference standards were processed and analyzed in parallel with the samples.

The reference materials utilized in this study are not certified for isotopic composition, and, to the authors' knowledge, there is no other published isotope data for the measured CRMs (certified reference material). Therefore, the presented isotope ratio data's accuracy was evaluated by comparing it with published isotope data for the Nod-A1 and Nod-P1 CRMs (Table S2).

4. Material Flow, Rönnskär

A simplified process flow sheet demonstrates the production lines for major metals. where in the respective production line, the F1-dust and the K1-dust are created. It should be noted that there is a significant internal circulation of material that is not shown (Figure S1).

Table S1. Inductively coupled plasma double-focusing sector-field mass spectrometer (ICP-SFMS) results for reference materials. Where not stated otherwise, concentrations are in mg kg^{-1} for solids and in μg L^{-1} for waters.

	GBW 07312 n = 4 Mean (SD) Certified	Nod-A1 n = 4 Mean (SD) Certified	Nod-P1 n = 4 Mean (SD) Certified		NASS-6 n = 2 Mean (SD) Published*	CASS-5 n = 2 Mean (SD) Published*	SLRS-4 n = 4 Mean (SD) Published *
Ag	1.07(0.07)1.15	0.16(0.02)NA	0.20(0.02)NA	**Ag**	<0.005	<0.005	0.0006(0.0001)0.0005-0.039
Al, %	4.5(0.3)4.9	2.10(0.09)2.05	2.45(0.10)2.54	**Al**	0.33(0.06)	0.87(0.07)	54.8(2.5)53-54
As	109(6)115	302(19)NA	84.9(6.1)NA	**As**	1.38(0.06)1.43	1.33(0.04)1.24	0.68(0.01)0.68-0.7
Au	0.004(0.001)0.006	<0.005(NA)NA	<0.005(NA)NA	**Au**	0.002(0.001)	0.008(0.002)	<0.0005(NA)0.0009-0.0028
B	25(2)24	106(9)NA	98(7)NA	**B**	4890(280)	5600(400)	6.3(0.2)5.95
Ba	188(14)206	1550(50)1670	3100(270)3350	**Ba**	6.30(0.12)6.86	6.47(0.09)7.4	12.3(0.2)12.2-12.6
Be	7.7(0.4)8.2	5.5(0.3)NA	2.2(0.2)NA	**Be**	<0.001(NA)NA	<0.001(NA)NA	0.012(0.001)
Bi	10.2(0.7)10.9	9.9(0.5)NA	5.6(0.4)NA	**Bi**	<0.001(NA)NA	<0.001(NA)NA	0.0018(0.0002)0.0021-0.011
Ca, %	0.74(0.07)0.84	11.1(0.6)11.0	2.18(0.15)2.22	**Ca, mg L^{-1}**	450(6)	480(10)	5.9(0.1)5.9
Cd	4.1(0.2)4.0	7.37(0.39)NA	21.7(0.9)22.6	**Cd**	0.008(0.002)0.006-0.033	0.026(0.001)0.022	0.013(0.001)0.012-0.014
Ce	54(6)61	705(40)NA	296(21)NA	**Ce**	0.005(0.001)0.004-0.006	0.005(0.001)0.004	0.365(0.008)0.36
Co	8.2(0.6)8.8	3110(190)3110	2200(140)2240	**Co**	0.013(0.003)0.015	0.100(0.009)0.095	0.034(0.002)0.033-0.048
Cr	29(3)35	18.9(1.6)NA	12.8(1.0)NA	**Cr**	0.129(0.003)0.118	0.116(0.006)0.106	0.328(0.003)0.33-0.37
Cs	7.7(0.4)7.9	0.60(0.04)NA	1.77(0.11)NA	**Cs**	0.222(0.006)0.266	0.224(0.013)0.266	0.007(0.001)0.007-0.009
Cu	1170(60)1250	1160(80)1110	11400(600)11500	**Cu**	0.228(0.010)0.248	0.388(0.025)0.38	1.87(0.02)1.81-1.93
Fe,%	4.19(0.29)4.12	11.4(0.7)10.9	5.93(0.38)5.81	**Fe**	0.47(0.03)0.495	1.34(0.06)1.44	104(3)103-108
Ga	13.2(1.3)14.1	6.15(0.34)NA	27.4(2.0)NA	**Ga**	<0.005(NA)NA	<0.005(NA)NA	0.009(0.002)0.012
Hf	7.5(0.7)8.3	5.98(0.47)	4.42(0.27)NA	**Hf**	0.0009(0.0002)0.0009	0.0008(0.0002)NA	0.0028(0.0002)0.003
Hg	0.05(0.01)0.06	<0.2(NA)NA	<0.2(NA)NA	**Hg**	<0.005(NA)NA	<0.005(NA)NA	0.002(0.001)0.032-0.21
K,%	2.35(0.18)2.41	0.51(0.04)0.50	1.02(0.05)0.996	**K, mg L^{-1}**	396(23)NA	397(11)NA	0.726(0.005)0.71
La	30.5(2.4)32.7	109(5)120	103(4)104	**La**	0.010(0.001)0.010-0.013	0.009(0.001)0.008	0.285(0.010)0.287
Li	40.2(3.1)39.0	73.3(6.0)	136(9)NA	**Li**	181(9)174-217	190(6)176-185	0.465(0.020)0.44-0.62
Mg,%	0.34(0.03)0.36	2.80(0.14)2.87	19.6(0.9)19.9	**Mg, mg L^{-1}**	1170(200)NA	1120(200)NA	1.66(0.11)1.7
Mn,%	0.135(0.09)0.140	18.0(1.0)18.5	29.3(1.2)29.1	**Mn**	0.504(0.026)0.53	2.40(0.03)2.62	2.99(0.25)3.0
Mo	8.1(0.5)8.4	415(22)448	691(41)760	**Mo**	8.96(0.06)9.7	9.58(0.10)9.82	0.219(0.012)0.21

Na,%	0.88(0.07)0.86	0.88(0.05)0.74	16.8(0.8)16.3	**Na, mg L^{-1}**	10300(600)	10200(700)	2.18(0.09)2.3
Nb	14.6(0.9)15.4	42.4(3.1)NA	20.6(1.4)NA	**Nb**	0.021(0.002)	0.026(0.002)	0.005(0.001)0.0041
Ni	13.0(0.8)12.8	6390(470)6360	13500(550)13400	**Ni**	0.318(0.017)0.301	0.356(0.011)0.33	0.716(0.013)0.67-0.82
P	232(18)235	6010(420)6110	2040(140)2010	**P**	21.7(0.8)	23.6(0.9)	6.9(0.2)6.1-9.3
Pb	290(19)285	855(62)846	494(32)560	**Pb**	0.007(0.001)0.006	0.018(0.001)0.011	0.086(0.002)0.084-0.086
Pd	0.009(0.002)NA	<0.2(NA)NA	<0.2(NA)NA	**Pd**	<0.001(NA)NA	<0.001(NA)NA	0.0015(0.0004)0.0016-0.021
Pt	<0.001	0.48(0.04)NA	0.116(0.011)NA	**Pt**	0.002(0.001)	0.003(0.001)	0.0014(0.0001)0.0013
Rb	252(23)270	10.2(0.4)NA	22.9(1.2)NA	**Rb**	112(4)94-103	114(5)104	1.59(0.05)1.4-1.6
Re	0.004(0.001)NA	<0.002(NA)NA	<0.002(NA)NA	**Re**	0.007(0.001)NA	0.009(0.001)0.0091	0.007(0.001)0.007
S	920(70)940	3280(260)NA	970(50)NA	**S**	1100(35)	1210(100)	2.80(0.12)
Sb	26.1(1.8)24.3	33.5(2.0)NA	49.9(2.7)NA	**Sb**	0.29(0.01)	0.63(0.02)	0.238(0.009)0.23-0.27
Sc	4.7(0.4)5.1	12.5(0.6)13	9.5(0.4)7.6	**Sc**	0.006(0.001)	0.005(0.001)	0.011(0.001)0.011-0.052
Se	0.21(0.04)0.25	3.1(0.8)NA	3.4(1.0)NA	**Se**	0.029(0.022)	0.027(0.015)	0.125(0.009)0.14-0.27
Sn	50(3)54	3.3(0.3)NA	2.1(0.2)NA	**Sn**	0.091(0.002)	0.045(0.004)	0.008(0.001)0.008-0.011
Sr	22.7(1.5)24.4	1670(90)1750	690(40)680	**Sr**	7270(480)6770-6890	7220(370)5780-6830	27.7(1.3)26.3-28.2
Ta	2.6(0.3)3.2	0.82(0.05)NA	0.39(0.04)NA	**Ta**	<0.001(NA)NA	<0.001(NA)NA	<0.0005(NA)0.0003
Te	0.25(0.03)0.29	29.7(2.0)NA	47.1(3.5)NA	**Te**	0.040(0.009)	0.045(0.009)	0.005(0.001)0.004
Th	20.1(1.4)21.4	25.6(1.4)NA	16.9(0.9)NA	**Th**	<0.001(NA)NA	<0.001(NA)NA	0.018(0.002)0.014-0.022
Ti,%	0.142(0.009)0.151	0.31(0.02)0.3177	0.28(0.02)0.2998	**Ti**	0.118(0.053)	0.171(0.031)	1.34(0.04)1.31-1.56
Tl	1.66(0.10)1.76	116(7)NA	205(14)NA	**Tl**	0.007(0.001)	0.007(0.001)	0.008(0.001)0.0076
U	7.1(0.6)7.8	6.88(0.37)NA	3.86(0.27)NA	**U**	2.85(0.05)3	2.92(0.07)3.18	0.051(0.003)0.05
W	37.0(3.4)37.4	85.8(5.1)NA	55.9(3.6)NA	**W**	0.011(0.001)	0.014(0.001)	0.005(0.001)0.005-0.013
V	44.1(2.9)46.6	690(50)770	530(40)570	**V**	1.38(0.03)1.46	1.39(0.04)1.32	0.330(0.012)0.32-0.35
Y	25.9(2.2)29.3	122(9)NA	88.1(6.0)NA	**Y**	0.015(0.001)0.017	0.019(0.001)0.022	0.143(0.008)0.146
Zn	511(42)498	770(60)590	1850(140)1600	**Zn**	0.225(0.040)0.257	0.793(0.034)0.719	0.984(0.019)0.93-1.24
Zr	197(16)234	290(20)NA	265(19)NA	**Zr**	0.009(0.002)0.023	0.008(0.002)NA	0.095(0.008)0.093-0.12

* Jochum et al., 2005.

Table S2. Isotope composition of Cd, Cu, Pb, and Zn in Nod-A1 and Nod-P1 CRM.

		δ^{114}Cd	δ^{65}Cu	^{208}Pb/^{206}Pb	^{207}Pb/^{206}Pb	δ^{66}Zn
Nod-A1	Mean(SD)	0.116(0.061)	0.416(0.071)	2.0534(0.0011)	0.82698(0.00008)	0.726(0.058)
(n = 4)	Published *	0.086-0.160	NA	2.0542–2.05432	0.82705–0.82709	0.69 **
Nod-P1	Mean(SD)	0.132(0.045)	0.428(0.048)	2.0687(0.0014)	0.83610(0.00009)	0.571(0.034)
(n = 4)	Published *	0.12-0.21	0.35-0.46	2.0686	0.8358	0.55

* Jochum et al., 2005; ** recalculated from JMC-Lyon.

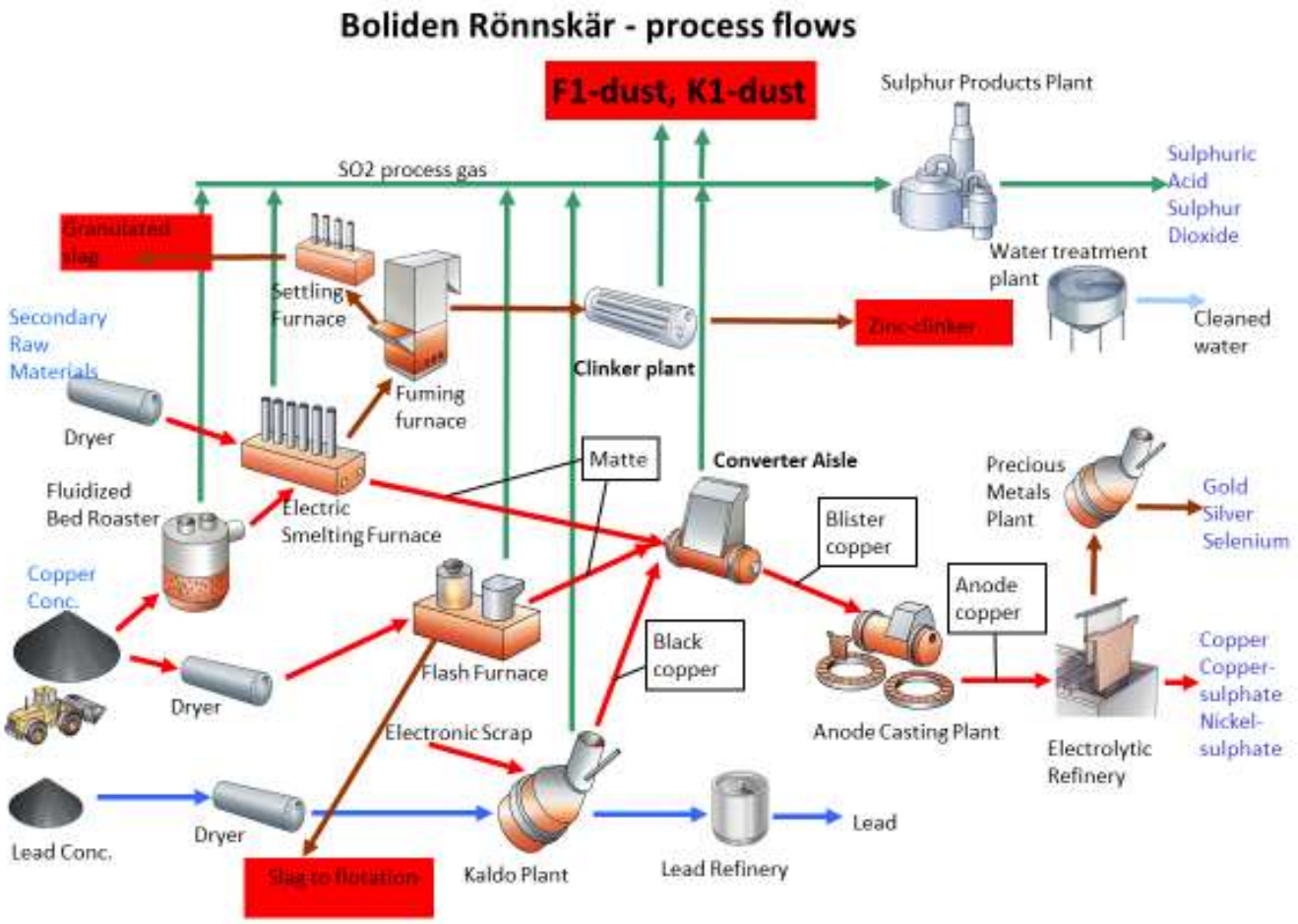
Table S3. Element concentrations in sediment samples, mg kg^{-1}. Instrumental precision: 3–7% RSD. (relative standard deviation)

Core	Depth (cm)	Ag	Al	As	Au	B	Ba	Be	Bi	Ca	Cd	Ce	Co	Cr
A	0–5	9.1	12828	1186	0.24	27	209	1.1	13.3	6258	52	61	37	96
A	5–10	9.5	13298	1698	0.30	24	338	1.1	18.8	5764	38	62	21	71
A	10–15	15.7	13631	2853	0.33	18	385	1.1	27.8	5740	314	61	37	76
A	15–20	9.6	11806	2607	0.25	13	336	0.7	44.9	4944	7	61	16	48
B	0–5	0.1	10682	272	0.01	9	96	0.7	0.3	6587	0.4	60	9	32
B	5–10	0.2	15078	142	0.01	15	128	1.1	0.7	7094	1.0	76	11	43
B	10–15	0.5	15233	112	0.02	18	106	1.0	2.2	7298	1.7	72	16	43
B	15–20	0.1	12381	91	0.02	18	80	0.9	5.2	6136	0.9	73	13	35
C	0–5	0.9	7620	242	0.04	7	110	0.4	2.2	4449	1.0	46	9	29
C	5–10	3.3	9148	1472	0.13	9	60	0.6	22.5	4642	4.2	54	14	35
C	10–15	0.0	9751	56	0.01	6	58	0.5	2.5	4432	0.5	46	10	36
C	15–20	0.0	6806	12	0.01	3	36	0.3	0.1	3705	0.1	40	4	23
D	0–5	0.6	14264	216	0.04	11	155	0.7	1.4	7481	0.7	54	13	52
D	5–10	0.6	16106	247	0.03	15	144	0.9	1.5	7598	0.7	59	17	65
D	10–15	3.4	16935	813	0.09	16	121	1.0	19.7	7494	2.4	68	29	65
D	15–20	0.7	16528	423	0.08	14	105	0.9	39.4	7440	2.3	71	17	49
E	0–5	0.3	9038	110	0.03	7	96	0.5	0.9	5193	0.5	48	12	28
E	5–10	0.9	9138	201	0.08	7	174	0.5	2.5	4991	0.6	48	10	28
E	10–15	0.4	11914	335	0.06	9	66	0.7	27.8	5640	1.6	60	12	33
E	15–20	0.0	5822	7	0.00	2	31	0.3	0.1	3795	0.1	33	3	17

Core	Depth (cm)	Cs	Cu	Fe	Ga	Hf	Hg	La	Li	Mg	Mn	Mo	Na	Nb
A	0–5	1.4	1847	68336	4.7	0.05	5.6	33	14	6560	2076	40.1	8210	4.1
A	5–10	1.4	2083	50269	4.9	0.13	9.1	35	15	6542	719	14.7	3846	3.6
A	10–15	1.5	2177	52766	4.9	0.15	13.5	33	15	6720	720	20.7	5010	3.8
A	15–20	1.3	1238	42026	4.5	0.20	17.7	31	14	5692	564	7.6	2823	3.6
B	0–5	1.0	35	45410	3.8	0.05	0.5	32	11	4922	1542	4.6	4614	4.6
B	5–10	1.4	57	40958	5.4	0.20	0.6	43	16	6487	1030	3.1	4945	5.6
B	10–15	1.3	63	51731	5.4	0.20	0.8	40	15	6490	1151	3.3	5188	4.8

B	15–20	1.3	45	51540	4.6	0.16	0.5	41	15	6004	1392	1.9	3757	3.9
C	0–5	0.8	181	24303	2.7	0.07	1.7	23	9	3595	928	4.8	2539	3.2
C	5–10	1.0	328	21407	3.2	0.24	13.3	27	11	4123	279	2.3	2469	3.8
C	10–15	1.2	22	22038	3.5	0.27	0.5	23	13	4639	280	1.1	1830	4.2
C	15–20	0.8	13	13698	2.7	0.28	0.3	19	9	3315	209	1.2	1210	3.8
D	0–5	1.0	130	53670	4.9	0.08	1.0	27	10	5061	5114	10.2	5864	4.4
D	5–10	1.1	126	62479	5.3	0.17	0.9	33	12	5716	1263	3.4	5776	5.3
D	10–15	1.4	348	60308	6.0	0.16	7.8	38	15	6709	1128	6.3	6034	4.4
D	15–20	1.4	150	46970	5.9	0.23	2.4	38	16	6530	891	3.1	4069	4.9
E	0–5	0.8	73	29445	3.0	0.06	0.6	23	9	3992	3268	6.5	3317	3.6
E	5–10	0.8	139	19893	3.2	0.19	1.2	23	9	3763	391	1.8	2297	4.0
E	10–15	1.1	96	30357	4.5	0.21	1.9	32	13	5161	591	1.8	2546	4.3
E	15–20	0.5	8	11917	2.1	0.30	0.3	16	7	2389	199	0.5	1002	3.7

Figure S1. A simplified process flow sheet demonstrates where the F1-dust and the K1-dust are obtained in the respective production line.



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