

1 Long-term temporal trend of PCBs 2 and their controlling sources in China

3 *Shizhen Zhao*^{1,5}, *Knut Breivik*^{2,3}, *Guorui Liu*⁴, *Minghui Zheng*⁴, *Kevin C. Jones*¹, *Andrew J.*
4 *Sweetman*^{1*}

5

6 ¹ Lancaster Environment Centre, Lancaster University, Lancaster, LA14YQ, UK

7 ² Norwegian Institute for Air Research, Box 100, NO-2027 Kjeller, Norway

8 ³ Department of Chemistry, University of Oslo, Box 1033, NO-0315 Oslo, Norway

9 ⁴ State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Centre for
10 Eco-Environmental Sciences, Chinese Academy of Sciences, P.O. Box 2871, Beijing 100085,
11 China

12 ⁵ State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry,
13 Chinese Academy of Sciences, Guangzhou 510640, China

14

15

16

17 *Corresponding author:

18 Andrew J. Sweetman

19 Tel: +44 (0) 1524 594715

20 Fax: +44 (0) 1524 594715

21 Email: a.sweetman@lancaster.ac.uk.

22 **Abstract**

23 Polychlorinated biphenyls (PCBs) are industrial organic contaminants identified as persistent,
24 bioaccumulative, toxic (PBT) and subject to long-range transport (LRT) with global scale
25 significance. This study focuses on a reconstruction and prediction for China of long-term emission
26 trends of intentionally and unintentionally produced (UP) \sum_7 PCBs (UP-PCBs, from the
27 manufacture of steel, cement and sinter iron) and their re-emissions from secondary sources (e.g.,
28 soils and vegetation), using a dynamic fate model (BETR-Global). Contemporary emission
29 estimates combined with predictions from the multimedia fate model suggest that primary sources
30 still dominate, although unintentional sources are predicted to become a main contributor from 2035
31 for PCB-28. Imported e-waste is predicted to play an increasing role until 2020-2030 on a national
32 scale due to the decline of IP emissions. Hypothetical emission scenarios suggest that China could
33 become a potential source to neighbouring regions with a net output of $\sim 0.4 \text{ t year}^{-1}$ in the case of 7
34 PCBs around 2050. However, future emission scenarios and hence model results will be dictated by
35 the efficiency of control measures.

36

37 **Keywords:**

38 Polychlorinated biphenyls; primary emissions; secondary emissions; multimedia fate model;
39 controlling sources

40

41

42

43

44

45 TOC

The image shows a Table of Contents (TOC) for a document. It consists of a grid of page numbers from 1 to 249, arranged in rows and columns. The grid is partially obscured by a chemical structure diagram and several highlighted yellow boxes. The chemical structure diagram shows two benzene rings, each with a chlorine atom (Cl) at the para position, connected by a central bond. The highlighted yellow boxes are labeled with page numbers: 66, 69, 90, 91, 92, 93, 115, and 116. The grid is color-coded: the top row (pages 1-24) is light blue, the second row (pages 25-48) is light red, the third row (pages 49-72) is light blue, the fourth row (pages 73-96) is light red, the fifth row (pages 97-120) is light blue, the sixth row (pages 121-144) is light red, the seventh row (pages 145-168) is light blue, the eighth row (pages 169-192) is light red, the ninth row (pages 193-216) is light blue, the tenth row (pages 217-240) is light red, the eleventh row (pages 241-264) is light blue, and the twelfth row (pages 265-288) is light red. The page numbers in the grid are: Row 1: 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24; Row 2: 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48; Row 3: 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72; Row 4: 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96; Row 5: 97, 98, 99, 100, 101, 102, 103, 104, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115, 116, 117, 118, 119, 120; Row 6: 121, 122, 123, 124, 125, 126, 127, 128, 129, 130, 131, 132, 133, 134, 135, 136, 137, 138, 139, 140, 141, 142, 143, 144; Row 7: 145, 146, 147, 148, 149, 150, 151, 152, 153, 154, 155, 156, 157, 158, 159, 160, 161, 162, 163, 164, 165, 166, 167, 168; Row 8: 169, 170, 171, 172, 173, 174, 175, 176, 177, 178, 179, 180, 181, 182, 183, 184, 185, 186, 187, 188, 189, 190, 191, 192; Row 9: 193, 194, 195, 196, 197, 198, 199, 200, 201, 202, 203, 204, 205, 206, 207, 208, 209, 210, 211, 212, 213, 214, 215, 216; Row 10: 217, 218, 219, 220, 221, 222, 223, 224, 225, 226, 227, 228, 229, 230, 231, 232, 233, 234, 235, 236, 237, 238, 239, 240; Row 11: 241, 242, 243, 244, 245, 246, 247, 248, 249, 250, 251, 252, 253, 254, 255, 256, 257, 258, 259, 260, 261, 262, 263, 264; Row 12: 265, 266, 267, 268, 269, 270, 271, 272, 273, 274, 275, 276, 277, 278, 279, 280, 281, 282, 283, 284, 285, 286, 287, 288, 289, 290.

46

47

48 **1 Introduction**

49 Polychlorinated biphenyls (PCBs) are industrial organic contaminants identified as persistent,
50 bioaccumulative, toxic and subject to long-range transport (LRT) with global scale significance.
51 They are among the twelve persistent organic pollutants (POPs) initially regulated by the
52 Stockholm Convention¹ in order to protect environmental and human health from these hazardous
53 compounds. The cumulative global production of PCB was approximately 1.3 million tonnes with
54 only ca 10 thousand tonnes produced in China since 1965.² These chemicals were mainly emitted as
55 a direct result of intentional historical production, use and disposal of products or accidental
56 release.³ Though they have been banned for several decades, they are still of great concern because
57 of the legacy of past usage, their persistence in the environment, bioaccumulation in biota and
58 potential toxicity.^{4,5}

59 PCBs can be emitted from both primary and secondary sources. Primary sources account for the
60 main direct releases of PCBs to the environment from their major use categories while secondary
61 sources represent the re-emission from environmental reservoirs including soils, sediments and
62 other contaminated compartments. Secondary sources can be viewed as “capacitors” that were
63 charged with pollutants deposited from the atmosphere when emissions were higher and may now
64 be net sources to the atmosphere.⁵ In industrialized countries, primary emissions of PCBs to the
65 environment peaked in the early 1970s and largely occurred through leakage and losses from the
66 PCB-containing products and systems. More recently, secondary sources have been demonstrated to
67 represent a significant fraction among the total source inventory, especially in some remote areas.⁵
68 Under such conditions, the reduction in primary emissions may not be directly apparent in declining
69 atmospheric concentrations due to on-going releases from secondary sources. Therefore, an
70 understanding of both primary and secondary emissions is a prerequisite to successful control
71 measures.

72 The production volume of PCBs in China accounts for approximately 1% of the global production.⁶
73 However, China has received PCBs from long-range atmospheric transport (LRAT) and trans-
74 boundary movement of e-waste products containing PCBs.⁷ Therefore, the release of PCBs into the
75 environment could be a combination of both primary and secondary emissions. Several studies
76 suggested that contaminated soil could be a secondary source, particularly contributing to low
77 molecular weight PCBs.^{8, 9} Seasonal patterns of air-soil exchange have been observed when net
78 volatilization occurred in summer.⁹⁻¹¹ Therefore, the relative significance of primary and secondary
79 emission is still under debate.

80 Primary PCB emissions into the atmosphere can be from intentionally produced (IP-PCB) and
81 unintentionally produced PCBs (UP-PCBs) formed during industrial thermal processes.^{12, 13}
82 Emissions trends of IP-PCBs have been predicted by Breivik and his co-workers on a global scale
83 and show a constantly decreasing trend since the middle of the 1970s when production was phased
84 out.^{2, 3, 14} This emission inventory was recently updated to cover the e-waste contributed IP-PCBs.¹⁵
85 On the other hand, since the ban on manufacture and use of commercial products containing PCBs,
86 UP-PCBs are likely to have become more important.¹⁶ Hogarh *et al.* (2012) reported that ambient
87 air concentrations in China have increased by one order of magnitude over the period 2004 to
88 2008.¹⁷ This is mainly linked to widespread industrial thermal process (e.g., thermal processes of
89 producing steel, cement and iron ore).^{16, 17} As the economy in China grows, there is an increasing
90 demand for construction materials such as steel and cement. China has contributed around 45% of
91 global steel production and become the world's largest consumer of iron ore since 1993.¹⁸
92 Consequently, the temporal trends and historical/future contribution of UP-PCBs needs to be
93 explored further. To understand which factors are controlling PCB burdens in environmental
94 compartments in China, it is important to quantify the relative significance of primary emissions
95 (controllable) versus secondary emissions (uncontrollable). An overestimate of the primary
96 emissions may lead to costly and inefficient control measures, whereas an underestimation of the

97 secondary emissions will result in an over-optimistic assessment of recovery rates following
98 primary emission reductions.¹⁹ A further important question would be what are the most important
99 primary sources, ‘intentional’ or ‘unintentional’ and do these overlap? These questions are of key
100 interest for policy makers since it will affect their perception of the need to reduce or eliminate
101 primary emissions and the effectiveness of emission reduction strategies.

102 The main aims of this study were 1) to simulate the individual contribution of primary sources
103 (from imported e-waste and IP/UP-PCBs emission) and secondary sources; 2) to evaluate modelling
104 results in air and soil with limited observations in China; 3) to provide suggestions to policy makers
105 on rational control measures for PCBs. These objectives were achieved by using the BETR-Global
106 fugacity-based model,²⁰ a dynamic level IV fate and transport model, which has been evaluated and
107 applied successfully for a range of organic contaminants, including PCBs.²⁰⁻²³

108 **2 Methods**

109 **2.1 Emission data and selected PCBs**

110 In this study, the emission, fate and transport, covering both intentionally and unintentionally
111 produced PCBs, were modelled under several scenarios for seven indicator \sum_7 PCBs (PCB-28, 52,
112 101, 118, 138, 153, and 180). These congeners were selected due to their representative
113 physicochemical properties and contribution in technical mixtures of PCBs.²⁴ The distribution of e-
114 waste emission was accounted based on the e-waste location in China.¹⁵ Other assembled emission
115 data were distributed into a 1°× 1° latitude/longitude grid system using a global population density
116 as a surrogate.²⁵ The physicochemical properties of selected congeners are presented in Table S1.²⁶

117 ²⁷

118 **2.1.1 IP-PCBs emission in China**

119 The recently revised global emission inventory by Breivik and co-workers was utilized in this
120 study,¹⁵ using a dynamic mass balance/flow analysis to calculate 22 IP-PCBs from 1930 to 2100.^{2,3}

121 ¹⁴ This emission inventory was recently developed to additionally account for the transport of e-

122 waste.^{3, 15} Scenarios of baseline-IP and worst-case IP with or without considering imported e-waste
123 as detailed in elsewhere.¹⁵ They are used to explore the relative contribution of PCBs from imported
124 e-waste to China.

125 **2.1.2 UP-PCBs emission in China**

126 Three major UP-PCB types were identified as representing dominant contributions to UP sources,
127 which capture more than 90% of known UP-sources so far.^{12, 13} These were cement kilns, electric
128 arc furnaces (EAF) used in steel making and the sintering process, also used in steel production.¹⁶
129 There is a potential underestimation of UP-PCBs emissions, since there are other UP-PCB sources
130 (e.g., coking, secondary aluminium production, and thermal power stations) that have not been
131 considered.¹³ Consequently, two scenarios were used to explore this potential uncertainty: (1) the
132 default scenario using measured emission factors;²⁸ and (2) a 'high' scenario using the measured
133 emission factors multiplied by a factor of 10 as a conservative assumption, since emission
134 inventories may often be uncertain by at least an order of magnitude.²⁹ These emission factors were
135 assumed constant over time during each simulation.

136 Three source types (IP-PCB, UP-PCB and secondary sources) were considered for past and future
137 emission scenarios. The secondary sources were calculated using the BETR Global model as
138 described in detail in section 2.3. The recorded (<http://www.stats.gov.cn/tjsj/ndsj/>, accessed on
139 27/09/2015) and estimated production volume of cement, EAF produced steel and sinter iron ore
140 between 1930-2100 are illustrated in Figure S1. The estimated annual emission data was assigned
141 onto a 1°×1° grid map using population density as a surrogate.²⁵ These estimates just represent a
142 first approximation, which may not be appropriate for some large plants located near sources of raw
143 materials and thus, would not correlate with population density.

144 **2.2 Selected fate model and study region**

145 The BETR-Global model was used to predict the fate and distribution of PCBs with a spatial
146 resolution of 15° latitude ×15° longitude and 288 grid cells. It was selected due to its relative coarse
147 resolution. Since the population density was used as a surrogate to the UP-PCBs emission with high

148 uncertainty. The coarse resolution of BETR-Global could potentially “even out” this simplification.
149 Each grid cell consists of seven bulk compartments, which are ocean water, fresh water, planetary
150 boundary layer (PBL), free atmosphere, soil, freshwater sediments and vegetation.²⁰ The model
151 accounts for advective transport between the regions by air/ water and inter-compartment transport
152 processes such as dry and wet deposition and reversible partitioning.²¹

153 The model simulations were performed at a global scale during the period 1930~2100 using a
154 dynamic level IV structure that assumes non-steady state conditions. The study region focussed on
155 China as shown in **Figure S2**. The temperature in the upper and lower atmosphere is taken from the
156 NCEP/NCAR reanalysis of climate data
157 (<https://www.esrl.noaa.gov/psd/data/reanalysis/reanalysis.shtml>). They are 15° x 15° averages for
158 the years 1960 - 1999. Multi-year model simulations repeat the same cycle of environmental
159 conditions. Only emission to the lower air compartment was considered. The initial model
160 concentration in all compartments was assumed to be zero.

161 **2.3 Estimation of source-receptor relationships**

162 Multiple emission inventory scenarios were investigated to explore different source-receptor
163 relationships. The employed emission profiles were defined as: 1) baseline-IP: no imported e-waste
164 and 5% of the disposed e-waste subject to open burning; 2) worst-case IP scenario: considering
165 imported e-waste and the fraction of open burning is 20%. The scenarios of baseline-IP and worst-
166 case IP were defined in detail elsewhere.¹⁵ They are used to explore the relative contribution from
167 imported e-waste to China; 3) default (IP+UP): UP-PCBs and worst-case IP-PCBs sources
168 combined, with calculated UP-PCBs using measured emission factors¹⁶; 4) worst case (IP+UP):
169 high scenario combined worst-case IP-PCBs and “high” UP-PCBs using a factor of 10 as defined in
170 section 2.1.2, to explore the uncertainty of emission factors for seven UP-PCBs.

171 First of all, to examine individual contribution from imported e-waste and UP-PCBs, the emission
172 scenarios of baseline IP, worst-case IP and default (IP+UP) were investigated by allowing

173 contaminants from both primary and secondary emissions in environmental reservoirs. Secondly, to
174 distinguish primary and secondary sources, the default (IP+UP) scenario was repeated with re-
175 emission from the ‘blocked’ surface compartments. The ‘blocked processes’ from surface-to-air
176 included diffusion from soil, water and vegetation to air, as well as re-suspension from soils via dust
177 and from oceans via marine aerosol production.³⁰ Thirdly, to explore the role of China in its global
178 context (sink or source), the model was also run using only the emission estimated within China
179 (regional emission) while the emission to other parts of the world was disabled (extra-regional
180 emission). The Chinese emission part was extracted from the global emission inventory according
181 to eight selected grids.

182 **3 Results and Discussion**

183 **3.1 Evaluation with measurements**

184 Firstly, the modelling results were evaluated using available measurement data to build confidence
185 for further model exploration. A model such as the one presented here can only be evaluated to a
186 limited extent, especially for a region where measurement data is scarce. However, it is also useful
187 to assess the accuracy of model predictions where possible. The output from the model with the
188 default scenario (IP+UP), over a limited period, was compared with available measured PCB data in
189 air and soil. As the BETR-Global model does not provide information on urban-rural gradients,
190 model predictions were compared against observed background concentrations. Atmospheric PCBs
191 concentrations have been measured in China over the last decade in rural and urban sites.^{31, 32}
192 Surveys providing PCBs concentration data in background soils have been conducted in 2005 and
193 2013^{33, 34} and normalized by total organic carbon (TOC). For comparisons to be made with studies
194 that do not distinguish between PCB congeners 28 and 31, PCB-28 was assumed to account for
195 55%.²⁷ This is a reflection of the composition of the technical mixtures.

196 Figures S11 ~ S13 compare predicted and observed time trends in air and soil for PCB congeners.
197 This comparison suggests that the model generally captures the main trends in observations over the

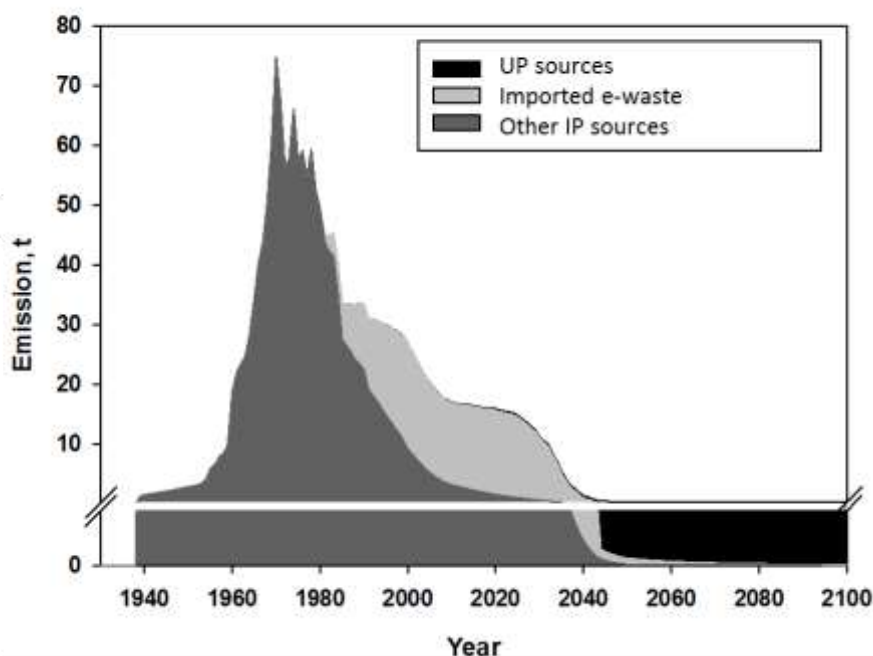
198 period 2001 to 2008. The agreement between predicted and observed air concentrations is better for
199 heavier PCBs than for the lighter congeners (PCB-28/52). Most modelled concentrations are within
200 a factor of three compared to the limited observations in background air. The model tended to
201 underestimate the atmospheric concentrations for PCB-28 and PCB-52 with the largest difference
202 occurring in 2001 by a factor of seven for PCB-52. This could be due to underestimated emission
203 from local sources.³⁵ The peak concentration, which occurred around 1970 predicted by the model,
204 is difficult to confirm with measurements. However, several preliminary findings from dated
205 sediment cores could potentially support the model estimation. The historical trend was observed to
206 increase until the mid-1970s in a dated sediment core from the Yangtze River Estuary adjacent to
207 the East Sea region and Pearl River Delta.^{36, 37} Predicted concentrations increase again from the
208 1980s, mainly associated with the imported electrical equipment containing PCBs and e-waste
209 recycling activities in nearby regions.^{36, 37}

210 Soil responds much slower to changes in emissions than air, especially for the heavier and more
211 persistent PCB congeners. Measured surface soil concentrations from 2005³³ and 2013 in forest
212 soil³⁴ were compared with model predictions and agreed well, within a factor of 4 except for tri-
213 PCB congeners, although the measured concentrations varied over a wide range. Soil data showed
214 similar results with the largest deviation observed for PCB-28 for both studies, indicating the
215 greater underestimation of soil concentrations by over a factor of 100. This may be caused by the
216 combined effect of parameter uncertainty (e.g. soil depth and organic content) and/or unaccounted
217 emissions. The measured data was limited to two sampling years: 2005 and 2013, but it showed
218 evidence of a decrease for PCB-28 and PCB-101. However, for PCB-138 and PCB-153, an increase
219 was observed from 0.28 to 0.42 ng/g OC (dw) for PCB-138 and from 0.09 to 0.31 ng/g OC (dw) for
220 PCB-153. These differences are small but could be attributed to the more recalcitrant nature of
221 heavier PCB congeners.³⁸

222 The homologue profile of PCBs (Figure S10) during the simulation period is also compared with
223 observations. The predicted change in homologue trend is generally consistent with the measured
224 profile.³⁸ Many studies have been conducted around heavily polluted areas (i.e. ‘hotspots’), and
225 much less data are available in background regions. Therefore, the high spatial variability of PCB
226 concentrations in soil with relatively low numbers of measurements at the background sites makes it
227 difficult to draw a reliable conclusion. A much larger dataset would be required to establish reliable
228 ranges for background concentrations to determine the whole picture of POPs pollution in China.

229 **3.2 Temporal trend of UP-PCBs in China**

230 The predicted time trends for past and future emissions of 7PCBs as well as their individual
231 contribution from imported e-waste and unintentionally sources are illustrated in Figure 1. Profiles
232 for other congeners are presented in Fig S3. Since the optimum scenario of unintentional-sources is
233 difficult to confirm with measurements, the default scenario (IP + UP) based on measured emission
234 factors was assumed to be the most representative of reality and used for further discussion. In
235 addition, the impact of an uncertainty factor of 10 on UP emissions from \sum_7 PCBs was also
236 explored (see Figure S3).



237

238 Figure 1. Predicted trends of total PCBs emission in China from 1930 to 2100 under the default
 239 scenario (IP+UP). The black area indicated the emission from UP sources; light grey area indicated
 240 the emission from imported e-waste and dark grey area presented emission from other IP sources.

241

242 The cumulative emission of intentionally produced \sum_7 PCBs from 1930 to 2040 was extracted from
 243 Ref 16 and estimated at 2300 tonnes in China (illustrated in Figure 1) with future emissions
 244 estimated to be about 2 tonnes from 2040 to 2100. Emissions of \sum_7 UP-PCBs were predicted to be
 245 9.5 tonnes between 1949 and 2040. However, their future emissions (2040-2100) were estimated
 246 around 23 tonnes under the default scenario with measured emission factors. Therefore, \sum_7 UP-PCB
 247 emissions only account for a minor portion of the total PCB emission, approximately 0.4% during
 248 the period of 1930-2040. However, they are predicted to play an increasingly important role in the
 249 near future (2040-2100) accounting for up to 91% of the \sum_7 PCB (UP+IP) emissions.

250 The predicted atmospheric concentrations were almost identical for the three emission scenarios,
 251 over the period 1930 to 2010 for \sum_7 PCBs (see Figure S4). This further supports the assumption that
 252 UP-PCBs did not contribute significantly over that period. After 2010, however, predicted air

253 concentrations started to diverge for each congener, attributed to different congener abundances
254 among the UP-PCB sources. In addition, the identification of markers could be informative for
255 future monitoring activities. Previously, PCB-118 was demonstrated to be a good marker congener
256 to describe and evaluate the emission trends from the industrial thermal process, since it falls in
257 both classes of dioxin like PCBs (dl-PCBs) and indicator PCBs.¹⁶ On the other hand, PCB-28 was
258 also demonstrated to have a significant correlation with seven congeners.¹² In this study, both
259 relationships were explored for PCB-28 and PCB-118, and a correlation coefficient (R^2) of 0.98 and
260 0.90 was observed ($p < 0.001$), respectively. Therefore, PCB-28 was suggested to be a useful
261 indicator congener for atmospheric PCBs concentrations from three considered emission sources.

262 For UP sources, PCB-28 was the dominant congener of the \sum_7 PCBs emission, accounting for
263 approximately 78% during 1930-2100. It also contributes about 28% of the \sum_7 PCBs (IP+UP)
264 emissions over the period dominated by IP-PCBs (1940-2010). The historical predominance of IP-
265 PCB-28 was anticipated as tri-PCBs were dominant in commercial mixtures used in China.²⁴
266 Predicted atmospheric concentrations of PCB-28 show the largest difference under three scenarios
267 as defined in Section 2.3, which is up to six orders of magnitude (Figure S4). This difference is
268 minimal for PCB-153 in Figure S4, which suggests that UP sources are more important for lighter
269 PCBs (PCB-28/52) than heavier ones (PCB-138/153), contributing less than 50% to concentrations
270 in air. In addition, atmospheric concentrations of different congeners will be dominated by
271 unintentional sources at different times. For example, as presented in Figure S4, PCB-28 is
272 predicted to be dominated by UP-PCB sources from 2035, due to high abundance in emission
273 sources, while PCB-52 will be dominated by UP sources after 2040 with a relatively gradual shift.

274 **3.3 Contribution from imported e-waste**

275 The trans-boundary movements of e-waste from developed countries to developing countries has
276 made it a potentially substantial inventory and emission source of PCBs.^{7, 15} Therefore, the
277 contribution of imported e-waste was explored to identify its influence (national or regional in
278 China). The cumulative emissions from imported e-waste are predicted to contribute around 30% to

279 the total emissions for seven congeners during 1930-2100. PCB-180 received the highest
280 percentage (45%) from imported e-waste. In terms of the cumulative atmospheric concentration in
281 different study grids (see Fig S3), the contribution of e-waste was largest for Grid 116 (which
282 included most e-waste recycling sites in South China), making up more than 30% of all congeners.

283 The influence of e-waste varied in different sampling years as illustrated in Figure S5. The import
284 of e-waste into China started around 1980. It is obvious that the Grid 116 received the highest
285 burden in atmosphere contributed by the imported e-waste, since the main e-waste recycling sites
286 (e.g., Guiyu and Qingyuan) with informal recycling activities are located here.³⁹ Evident regional
287 differences are predicted in terms of influence from imported-waste, e.g., Grid cell 66 (mainly
288 covering Xinjiang) received the least e-waste associated PCBs, as it is remote from the e-waste
289 recycling sites. Imported e-waste is predicted to play an increasing role until 2020-2030 on a
290 national scale in relative terms, when Grid cell 116 received more than 90% of input contributed by
291 imported e-waste. This is not because PCBs produced by imported e-waste will increase in the
292 period, but rather because other IP-sources are on a continuous decline since then ban on production.
293 After 2030, the relative contribution from imported e-waste is predicted to diminish (Figure S5)
294 representing less than 5% to the total modelled air concentration by 2100. However, the future
295 emissions of e-waste may be different to the emission scenario used herein, largely depending on
296 Chinese and international control strategies. For instance, Chinese government had issued a variety
297 of laws and legislations to establish a formal e-waste recycling system.⁴⁰ If the e-waste treatment
298 gradually transits from open-burning by backyard workshop to integrated recycling process by
299 qualified companies, the PCBs emission may decline faster than anticipated.

300 **3.4 Contribution from secondary sources**

301 Being able to distinguish between primary and secondary sources is important for understanding
302 our ability to control sources and to aid policy makers to develop the most effective control
303 measures. The advection into (and out of) China from the wider Asian region also needs to be

304 quantified to place China's activities into a regional context. Therefore, the primary and secondary
305 sources from China (region) and out of China (extra-region) were estimated for PCB-28 and PCB-
306 153 (see Figure S7-a, b). In addition, the individual contribution of secondary sources from soil,
307 water and vegetation to air, was explored (Figure S7-c, d), where regional primary/secondary
308 emission represents emissions from the domestic sources (China) while extra-regional/primary
309 emission represents the emissions from outside China, as result of LRAT.

310 When separating secondary sources into regional and extra-regional, the profiles for PCB-28 and
311 PCB-153 were similar until 2030 (see Figure S7). The extra-regional primary and secondary
312 sources dominate the emission during the initial period from 1930 to 1960 for both PCB-28 and
313 PCB-153. During that period, China did not have any domestic production or usage of PCBs.
314 Therefore, LRAT would have been responsible for supplying PCB to the Chinese environment.
315 However, when China started to produce PCBs in 1964, primary sources became increasingly
316 important and had provided a steady contribution of approximately 70%, which is predicted to
317 continue until around 2030. Afterwards, both congeners are predicted to behave differently. Future
318 levels for PCB-28 are predicted to be mainly dominated by regional primary emission whilst PCB-
319 153 is mostly controlled by extra-regional secondary sources. This could be due to PCB-28 mainly
320 being supplied by ongoing and increasingly important UP sources as discussed in Section 3.2. In
321 contrast primary sources of PCB-153 should gradually decline within China with secondary extra-
322 regional emission becoming slowly more important.

323 Several studies have suggested that the main contribution to PCB emission should move from
324 primary to secondary sources as production and use of PCBs declines.^{11,41} In China, the same trend
325 can be seen for PCB-28 when simulations were performed only considering IP-PCBs (see Figure
326 S8-b). However, when taking UP-PCB into account, it appears that the primary sources remained
327 dominant over the whole simulation period as in Figure S8-a. As for the individual sources of UP-
328 PCBs, the main contribution to emissions moved from cement kilns to EAF production over the

329 period 2010 to 2020 (see Figure S9). EAF allows steel to be made from 100% scrap, and as a result,
330 it could greatly reduce energy consumption.⁴² So this technology is being strongly promoted.
331 However, without effective control measures, EAF may have potential to cause increased emission
332 of UP-PCBs.

333 **3.4.1 Re-emission from soil-air**

334 The exchange of POPs across the air-soil interface is one of the most important processes
335 determining their long-term environmental fate, as the soil is thought to be a major reservoir in the
336 terrestrial environment.¹¹ When individual contribution of secondary sources from soil and
337 vegetation for PCB-153 was explored (see Figure S7-c, d), vegetation was predicted to dominate
338 until 2030 with soil gradually becoming the main secondary source. This is a reflection of
339 difference in the relative size of vegetation and soils as storage compartments. Delayed re-emissions
340 normally occur from compartments that are slow to respond to changes in atmospheric
341 concentrations such as soils and the oceans.³⁰ Therefore, soil represents an initial sink for PCBs
342 until it reaches equilibrium with air, after which it becomes a net source as primary emissions
343 decline.⁸ It is important to take into account that these calculations assume a well-mixed soil depth
344 of 20 cm and increasing the depth would increase soil capacity⁴³ and vice versa.

345 Secondary emissions also occur from vegetation, although over a much shorter time-scale as
346 vegetation responds rapidly to the changes in atmospheric concentrations.³⁰ The model suggests that
347 vegetation is a dominant secondary source for the whole simulation period for PCB-28 (see Figure
348 S4-c). This may be because primary sources are controlling the emission to the atmosphere, with
349 soils acting as a reservoir during the simulated period. It was demonstrated that atmospheric
350 deposition is the main contamination pathway for vegetation, rather than uptake from the soil, based
351 on a study of paddy rice in China.⁴⁴

352 **3.4.2 Analysis of compartment response times (VZ/D)**

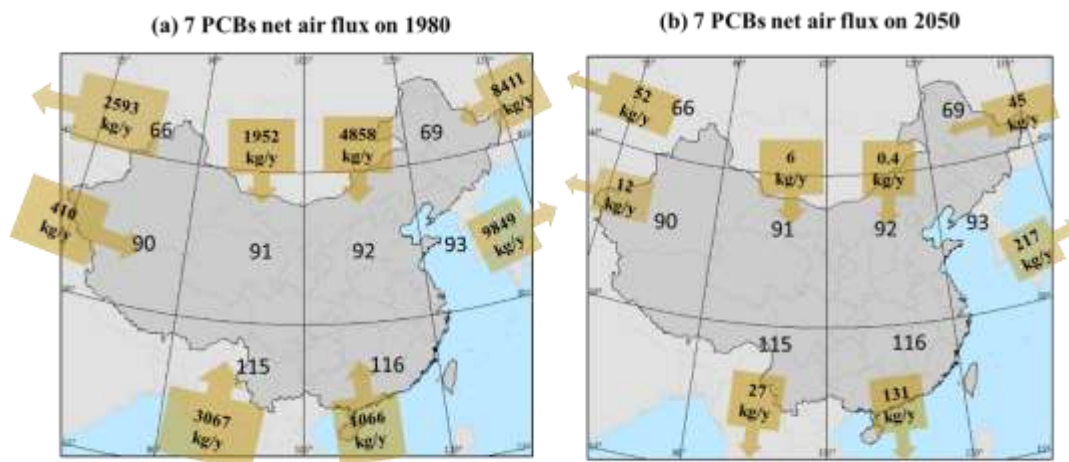
353 The roles of soil and vegetation compartments as secondary sources can be further explained by
354 model calculations. Taking air (A), soil (S) and vegetation (V) as examples. The 'storage capacity'

355 of each medium can be calculated using compartment volume (V , m^3) and fugacity capacity (Z , mol
356 $m^{-3} Pa^{-1}$). For PCB-28, the $V_s Z_s$ is $2.6 \times 10^{15} mol Pa^{-1}$, $V_v Z_v$ is $2.5 \times 10^{12} mol Pa^{-1}$, $V_a Z_a$ is $7.4 \times 10^{11} mol Pa^{-1}$.
357 Thus, the soil has approximately 3500 times the storage capacity of the air and has approximately
358 1000 times the capacity of the vegetation. The transport parameter D value for soil-to-air transfer
359 D_{sA} is $2.3 \times 10^9 mol Pa^{-1} h^{-1}$ and vegetation-to-air transfer $D_{v,A}$ is $9.6 \times 10^9 mol Pa^{-1} h^{-1}$. The
360 characteristic time (VZ/D), is the average time that a chemical 'spends' in a single compartment and
361 is the first indication of persistence.⁴⁵ This was calculated to be approximately 92 years and ten days
362 in soil and vegetation, respectively.⁴⁶ Therefore, PCB-28 in the atmosphere will rapidly exchange
363 with the vegetation as it attempts to approach equilibrium. In addition, the pathways of air-to-soil
364 and vegetation-to-soil were also calculated to compare the relative importance of these two
365 pathways. The calculations suggest that the characteristic time from air to soil is 18 days while
366 vegetation to soil is about one year. However, leaves can represent a large effective surface area
367 which is greater than the soil surface area covered by the vegetation⁴⁷ and so may represent an
368 important deposition pathway for PCBs.

369 **3.5 Atmospheric advection**

370 The importance of atmospheric advection between Chinese study regions and the extra-region was
371 investigated and the results presented in Figure 2 for two contrasting years 1980 and 2050,
372 respectively representing the 'in-use' and 'phase-out' periods. PCB production and use were
373 restricted around 1974,² and peak emissions were expected around 1980. At that time, the central
374 part of China (Grid 91 and 92) acted as a PCB storage reservoir while east of the country as
375 industrialized areas acted as sources of PCBs to outside regions. It is interesting to note that the
376 western parts of the country, which are not highly industrialized, have been acting as a net source,
377 which may be attributed to high abundance of lighter PCB congeners in China. Their volatility and
378 advection from the rest of the world or low TOC in these soils may cause this. When looking at
379 future predictions up to 2050, the central part of China is still predicted to receive PCBs from
380 industrialized regions with decreasing quantity. The direction of the net flux changes from the west

381 and south part. When examining China a whole, the model predicts that this country has moved
 382 from a sink with a net atmospheric input of $\sim 7\text{ t year}^{-1}$ for 7 indicator PCBs to acting as a potential
 383 source to neighbouring regions with a net output of $\sim 0.4\text{ t year}^{-1}$. However, model results will be
 384 dictated by the efficiency of relative control measures.



385

386 Figure 2. The net flux of 7 indicator PCBs atmospheric advection between region and extra-region
 387 on 1980 (a) and 2050 (b), using the worst-case emission scenario (IP+UP).

388

389 3.6 Uncertainty

390 The emission inventory and environmental concentrations estimated in this study contain high
 391 levels of uncertainty caused by a wide range of factors. One of the most important uncertainties is
 392 the comprehensive identification of e-waste sources. Although the domestic generation of e-waste
 393 and its import from overseas have generally been captured in the current inventory, several types of
 394 electronic equipment were not considered (e.g., large household appliances and telecommunication
 395 equipment), which are still increasing. These may be considered in future work, although PCB
 396 production has been banned.⁷ Another concern is the difficulty in tracking illicit import of e-waste
 397 without effective regulation in China. A complementary approach to tracking the sources, flows and
 398 destination of e-waste could provide further insights into the emission of e-waste pollutants.⁷

399 For the emission of UP-PCBs, only three major industrial processes were considered in this study.
400 Other industrial sources could also contribute to the emission of UP-PCBs, such as secondary zinc
401 smelting and thermal wire reclamation.¹⁶ However, the individual congener profile of many
402 industrial processes is lacking, and using emission factors from other countries has been shown to
403 be misleading.¹² For example, when comparing the emission factors used in this study¹⁶ with those
404 reported from other countries, large differences were observed. Emission factors for cement
405 production were up to 1000 times lower here than those used in the Japanese Toolkit.¹³ This could
406 be due to the use of industrial thermal process, such as waste incinerators fed on alternative waste
407 material, is not very common in China. Even within this study, there were wide variations of
408 observed emission factors in the same type of plants in China with up to 100 times difference in the
409 most extreme case.¹⁶ Therefore, using emission factors from other countries should only be
410 recommended when domestic measurements are not available. Even then, caution should be taken.
411 These differences also highlight the need for a more systematic survey of emission sources on a
412 national scale to provide an unbiased and comprehensive reference for the emission inventory. A
413 better characterization of emission factors is essential to help to produce a more accurate estimation
414 of the time trends in the future.

415 The actual sources of PCBs via industrial processes also needs to be further scrutinized. Since PCBs
416 are not only formed by *de novo* synthesis or precursors, they may also be present in the raw
417 materials.⁴⁸ For example, PCB concentrations in iron ores were reported to be around 1-1.6 mg t⁻¹ in
418 a European sinter plant.⁴⁹ They are likely to be destroyed mostly in the combustion zone but may be
419 driven off due to their volatility. Therefore, it is very important but also, a great challenge to
420 differentiate the portion existing in the raw material and from new formation, in order to avoid
421 double accounting for emission estimation and minimize input of contaminants going into industrial
422 thermal processes or end of pipe measures.

423 The Chinese cement industry uses coal almost exclusively as fuel.⁵⁰ There is very little use of
424 alternative fuels (defined as waste materials with heat value more than 4000 kcal kg⁻¹ for cement
425 clinker burning) or the compression of waste materials (defined as the incineration of wastes for
426 disposal purposes) in cement production. However, Chinese laws and policies now tend to
427 encourage industry to use alternative fuels and waste materials.⁵⁰ This may result in more recycled
428 waste material being used for cement production.

429 In the steel and iron industries, the raw materials are mainly from internally produced steel scrap
430 with some imported from abroad. The process of scrap preheating used in EAF may result in higher
431 emissions of PCBs from contaminated scrap with paints and lubricants containing PCBs, which
432 could be minimized post-combustion using additional oxygen burners.⁴⁹ However, the related
433 information is very limited in China. For recycled scrap, it is forbidden to have hazardous material
434 with more than 50 mg kg⁻¹ PCBs which is regulated by the Chinese government (GB13015-91). So
435 the impact caused by the presence of PCBs in raw materials for steel industry is assumed to be
436 negligible.

437 In this study, population density was used to distribute PCBs emission to each grid cell. For the UP-
438 PCBs, high uncertainty may exist due to the recent movement of industrial sources from urban to
439 rural or semi-rural areas. For example, most PCB-containing equipment is stored at special sites
440 after they have become waste. However, due to poor management and storage conditions, PCBs
441 from some of these special storage locations have leaked into the environment of surrounding areas,
442 especially to the soil.^{32, 38}

443 **3.7 Implications for control measures**

444 The environmental response to regulatory measures for the control of persistent chemicals can be
445 very slow and substance-specific.⁵¹ Further, regional differences are also anticipated, particularly
446 for a large country with varied geographical variations and levels of economic development like
447 China. For this reason, an effective strategy should be developed and implemented as early as

448 possible. Results from this study suggest that the effectiveness of emission control measures may
449 vary significantly for individual substances and specific regions. For example, primary sources are
450 still predominant for PCB-28, which means controllable sources could be effectively mitigated via
451 implementing policy and regulations, especially for controlling the UP-PCBs from industrial
452 processes. The predictions suggest that UP-PCBs had little impact on the past emission profile, but
453 may potentially provide a greater contribution from around 2050, if current industrial thermal
454 processes continue without further control strategies. Although the emission abatement techniques
455 have been developed, further work is needed to control POPs from industrial activities, and on-site
456 monitoring.²⁸ Nevertheless, this may not work well for PCB-153 and PCB-180, since imported e-
457 waste is a more important contributor at this stage, particularly in the southern part of China.

458 **Support Information**

459 Detailed information on chemical properties, PCB production history in China, prediction approach
460 of UP-PCBs emission and additional model results. This material is available free of charge via the
461 Internet at <http://pubs.acs.org/>.

462 **Acknowledgement**

463 KB was supported by the Research Council of Norway (213577). Thanks to Dr Matthew MacLeod
464 and Dr Henry Wöhrnschimmel for their guidance on running BETR-Global model. Thanks to Dr
465 Stefan Pauliuk and Prof Zeyi Jiang for offering data estimated production of EAF and iron sinter.

466 **References**

- 467 (1) UNEP *The Stockholm Convention on Persistent Organic Pollutants*; United Nations
468 Environmental Programme: 2001.
469 (2) Breivik, K.; Sweetman, A.; Pacyna, J. M.; Jones, K. C., Towards a global historical emission
470 inventory for selected PCB congeners - a mass balance approach 1. Global production and
471 consumption. *Science of the Total Environment* **2002**, *290*, (1-3), 181-198.

- 472 (3) Breivik, K.; Sweetman, A.; Pacyna, J. M.; Jones, K. C., Towards a global historical emission
473 inventory for selected PCB congeners - a mass balance approach 2. Emissions. *Science of the Total*
474 *Environment* **2002**, *290*, (1-3), 199-224.
- 475 (4) Jones, K. C.; de Voogt, P., Persistent organic pollutants (POPs): state of the science.
476 *Environmental Pollution* **1999**, *100*, (1-3), 209-221.
- 477 (5) Nizzetto, L.; Macleod, M.; Borgå, K.; Cabrerizo, A.; Dachs, J.; Guardo, A. D.; Ghirardello, D.;
478 Hansen, K. M.; Jarvis, A.; Lindroth, A.; Ludwig, B.; Monteith, D.; Perlinger, J. A.; Scheringer, M.;
479 Schwendenmann, L.; Semple, K. T.; Wick, L. Y.; Zhang, G.; Jones, K. C., Past, Present, and Future
480 Controls on Levels of Persistent Organic Pollutants in the Global Environment. *Environmental*
481 *Science & Technology* **2010**, *44*, (17), 6526-6531.
- 482 (6) Liu, L. Y.; Ma, W. L.; Jia, H. L.; Zhang, Z. F.; Song, W. W.; Li, Y. F., Research on persistent
483 organic pollutants in China on a national scale: 10 years after the enforcement of the Stockholm
484 Convention. *Environ. Pollut.* **2016**, *217*, 70-81.
- 485 (7) Breivik, K.; Armitage, J. M.; Wania, F.; Jones, K. C., Tracking the global generation and
486 exports of e-waste. Do existing estimates add up? *Environ. Sci. Technol.* **2014**, *48*, (15), 8735-43.
- 487 (8) Li, Y. F.; Harner, T.; Liu, L.; Zhang, Z.; Ren, N. Q.; Jia, H.; Ma, J.; Sverko, E., Polychlorinated
488 biphenyls in global air and surface soil: distributions, air-soil exchange, and fractionation effect.
489 *Environ. Sci. Technol.* **2010**, *44*, (8), 2784-90.
- 490 (9) Wang, X.-p.; Sheng, J.-j.; Gong, P.; Xue, Y.-g.; Yao, T.-d.; Jones, K. C., Persistent organic
491 pollutants in the Tibetan surface soil: Spatial distribution, air-soil exchange and implications for
492 global cycling. *Environmental Pollution* **2012**, *170*, 145-151.
- 493 (10) Wang, Y.; Cheng, Z.; Li, J.; Luo, C.; Xu, Y.; Li, Q.; Liu, X.; Zhang, G., Polychlorinated
494 naphthalenes (PCNs) in the surface soils of the Pearl River Delta, South China: Distribution,
495 sources, and air-soil exchange. *Environmental Pollution* **2012**, *170*, 1-7.
- 496 (11) Cousins, I. T.; Beck, A. J.; Jones, K. C., A review of the processes involved in the exchange of
497 semi-volatile organic compounds (SVOC) across the air-soil interface. *Science of the Total*
498 *Environment* **1999**, *228*, (1), 5-24.
- 499 (12) Cui, S.; Fu, Q.; Ma, W.-L.; Song, W.-W.; Liu, L.-Y.; Li, Y.-F., A preliminary compilation and
500 evaluation of a comprehensive emission inventory for polychlorinated biphenyls in China. *Science*
501 *of The Total Environment* **2015**, *533*, (0), 247-255.
- 502 (13) Cui, S.; Qi, H.; Liu, L. Y.; Song, W. W.; Ma, W. L.; Jia, H. L.; Ding, Y. S.; Li, Y. F., Emission
503 of unintentionally produced polychlorinated biphenyls (UP-PCBs) in China: Has this become the
504 major source of PCBs in Chinese air? *Atmospheric Environment* **2013**, *67*, 73-79.
- 505 (14) Breivik, K.; Sweetman, A.; Pacyna, J. M.; Jones, K. C., Towards a global historical emission
506 inventory for selected PCB congeners — A mass balance approach: 3. An update. *Science of The*
507 *Total Environment* **2007**, *377*, (2-3), 296-307.
- 508 (15) Breivik, K.; Armitage, J. M.; Wania, F.; Sweetman, A. J.; Jones, K. C., Tracking the Global
509 Distribution of Persistent Organic Pollutants Accounting for E-Waste Exports to Developing
510 Regions. *Environ. Sci. Technol.* **2016**, *50*, (2), 798-805.
- 511 (16) Liu, G. R.; Zheng, M. H.; Cai, M. W.; Nie, Z. Q.; Zhang, B.; Liu, W. B.; Du, B.; Dong, S. J.;
512 Hu, J. C.; Xiao, K., Atmospheric emission of polychlorinated biphenyls from multiple industrial
513 thermal processes. *Chemosphere* **2013**, *90*, (9), 2453-2460.
- 514 (17) Hogarh, J. N.; Seike, N.; Kobara, Y.; Habib, A.; Nam, J.-J.; Lee, J.-S.; Li, Q.; Liu, X.; Li, J.;
515 Zhang, G.; Masunaga, S., Passive air monitoring of PCBs and PCNs across East Asia: A
516 comprehensive congener evaluation for source characterization. *Chemosphere* **2012**, *86*, (7), 718-
517 726.
- 518 (18) Feng, L., China's steel industry: Its rapid expansion and influence on the international steel
519 industry. *Resources Policy* **1994**, *20*, (4), 219-234.
- 520 (19) Breivik, K.; Alcock, R.; Li, Y. F.; Bailey, R. E.; Fiedler, H.; Pacyna, J. M., Primary sources of
521 selected POPs: regional and global scale emission inventories. *Environmental Pollution* **2004**, *128*,
522 (1-2), 3-16.

- 523 (20) Macleod, M.; Riley, W. J.; Mckone, T. E., Assessing the influence of climate variability on
524 atmospheric concentrations of polychlorinated biphenyls using a global-scale mass balance model
525 (BETR-global). *Environmental Science & Technology* **2005**, *39*, (17), 6749-6756.
- 526 (21) Lamon, L.; Von Waldow, H.; Macleod, M.; Scheringer, M.; Marcomini, A.; Hungerbuhler, K.,
527 Modeling the global levels and distribution of polychlorinated biphenyls in air under a climate
528 change scenario. *Environ. Sci. Technol.* **2009**, *43*, (15), 5818-24.
- 529 (22) MacLeod, M.; von Waldow, H.; Tay, P.; Armitage, J. M.; Wöhrnschimmel, H.; Riley, W. J.;
530 McKone, T. E.; Hungerbuhler, K., BETR Global—A geographically-explicit global-scale
531 multimedia contaminant fate model. *Environmental Pollution* **2011**, *159*, (5), 1442-1445.
- 532 (23) MacLeod, M.; Woodfine, D. G.; Mackay, D.; McKone, T.; Bennett, D.; Maddalena, R., BETR
533 North America: a regionally segmented multimedia contaminant fate model for North America.
534 *Environ. Sci. Pollut. Res.* **2001**, *8*, (3), 156-163.
- 535 (24) Fu, J.; Mai, B.; Sheng, G.; Zhang, G.; Wang, X.; Peng, P. a.; Xiao, X.; Ran, R.; Cheng, F.;
536 Peng, X.; Wang, Z.; Wa Tang, U., Persistent organic pollutants in environment of the Pearl River
537 Delta, China: an overview. *Chemosphere* **2003**, *52*, (9), 1411-1422.
- 538 (25) Li, Y. F.; McMillan, A.; Scholtz, M. T., Global HCH usage with 1 degrees x1 degrees
539 longitude/latitude resolution. *Environmental Science & Technology* **1996**, *30*, (12), 3525-3533.
- 540 (26) Schenker, U.; MacLeod, M.; Scheringer, M.; Hungerbuhler, K., Improving data quality for
541 environmental fate models: a least-squares adjustment procedure for harmonizing physicochemical
542 properties of organic compounds. *Environ. Sci. Technol.* **2005**, *39*, (21), 8434-41.
- 543 (27) Breivik, K.; Czub, G.; McLachlan, M. S.; Wania, F., Towards an understanding of the link
544 between environmental emissions and human body burdens of PCBs using CoZMoMAN. *Environ.*
545 *Int.* **2010**, *36*, (1), 85-91.
- 546 (28) Liu, G.; Zheng, M.; Jiang, X.; Jin, R.; Zhao, Y.; Zhan, J., Insights into the emission reductions
547 of multiple unintentional persistent organic pollutants from industrial activities. *Chemosphere* **2015**,
548 *144*, 420-424.
- 549 (29) Annema, J.; Beurskens, J.; Bodar, C. *Evaluation of PCB fluxes in the environment. Report*
550 *no.601014011*; National Institute for Public Health and the Environment (RIVM) 1995.
- 551 (30) Wöhrnschimmel, H.; MacLeod, M.; Hungerbuhler, K., Global multimedia source-receptor
552 relationships for persistent organic pollutants during use and after phase-out. *Atmos Pollut Res* **2012**,
553 *3*, (4), 392-398.
- 554 (31) Jaward, T. M.; Zhang, G.; Nam, J. J.; Sweetman, A. J.; Obbard, J. P.; Kobara, Y.; Jones, K. C.,
555 Passive air sampling of polychlorinated biphenyls, organochlorine compounds, and polybrominated
556 diphenyl ethers across Asia. *Environmental Science & Technology* **2005**, *39*, (22), 8638-8645.
- 557 (32) Xing, X.; Lu, Y. L.; Dawson, R. W.; Shi, Y. J.; Zhang, H.; Wang, T. Y.; Liu, W. B.; Ren, H. C.,
558 A spatial temporal assessment of pollution from PCBs in China. *Chemosphere* **2005**, *60*, (6), 731-
559 739.
- 560 (33) Ren, N.; Que, M.; Li, Y. F.; Liu, Y.; Wan, X.; Xu, D.; Sverko, E.; Ma, J., Polychlorinated
561 biphenyls in Chinese surface soils. *Environ. Sci. Technol.* **2007**, *41*, (11), 3871-6.
- 562 (34) Zheng, Q.; Nizzetto, L.; Mulder, M. D.; Sanka, O.; Lammel, G.; Li, J.; Bing, H.; Liu, X.; Jiang,
563 Y.; Luo, C.; Zhang, G., Does an analysis of polychlorinated biphenyl (PCB) distribution in
564 mountain soils across China reveal a latitudinal fractionation paradox? *Environmental Pollution*
565 **2014**, *195*, 115-122.
- 566 (35) Xing, Y.; Lu, Y. L.; Dawson, R. W.; Shi, Y. J.; Zhang, H.; Wang, T. Y.; Liu, W. B.; Ren, H. C.,
567 A spatial temporal assessment of pollution from PCBs in China. *Chemosphere* **2005**, *60*, (6), 731-
568 739.
- 569 (36) Yang, H.; Zhuo, S.; Xue, B.; Zhang, C.; Liu, W., Distribution, historical trends and inventories
570 of polychlorinated biphenyls in sediments from Yangtze River Estuary and adjacent East China Sea.
571 *Environmental Pollution* **2012**, *169*, 20-26.
- 572 (37) Mai, B. X.; Zeng, E. Y.; Luo, X. J.; Yang, Q. S.; Zhang, G.; Li, X. D.; Sheng, G. Y.; Fu, J. M.,
573 Abundances, depositional fluxes, and homologue patterns of polychlorinated biphenyls in dated

- 574 sediment cores from the Pearl River Delta, China. *Environmental Science & Technology* **2005**, *39*,
575 (1), 49-56.
- 576 (38) Zhang, Z.; Liu, L.; Li, Y.-F.; Wang, D.; Jia, H.; Harner, T.; Sverko, E.; Wan, X.; Xu, D.; Ren,
577 N.; Ma, J.; Pozo, K., Analysis of Polychlorinated Biphenyls in Concurrently Sampled Chinese Air
578 and Surface Soil. *Environmental Science & Technology* **2008**, *42*, (17), 6514-6518.
- 579 (39) Chen, S. J.; Tian, M.; Zheng, J.; Zhu, Z. C.; Luo, Y.; Luo, X. J.; Mai, B. X., Elevated levels of
580 polychlorinated biphenyls in plants, air, and soils at an E-waste site in Southern China and
581 enantioselective biotransformation of chiral PCBs in plants. *Environ. Sci. Technol.* **2014**, *48*, (7),
582 3847-55.
- 583 (40) Zhang, S.; Ding, Y.; Liu, B.; Pan, D. a.; Chang, C.-c.; Volinsky, A. A., Challenges in
584 legislation, recycling system and technical system of waste electrical and electronic equipment in
585 China. *Waste Manage* **2015**, *45*, 361-373.
- 586 (41) Harner, T.; Mackay, D.; Jones, K. C., Model of the long-term Exchange of PCBs between soil
587 and the atmosphere in the southern U.K. *Environ. Sci. Technol.* **1995**, *29*, (5), 1200-9.
- 588 (42) Pauliuk, S.; Milford, R. L.; Muller, D. B.; Allwood, J. M., The steel scrap age. *Environ. Sci.*
589 *Technol.* **2013**, *47*, (7), 3448-54.
- 590 (43) Sweetman, A. J.; Valle, M. D.; Prevedouros, K.; Jones, K. C., The role of soil organic carbon
591 in the global cycling of persistent organic pollutants (POPs): interpreting and modelling field data.
592 *Chemosphere* **2005**, *60*, (7), 959-972.
- 593 (44) Bi, X.; Chu, S.; Xu, X., Transport of PCB in contaminated paddy fields. *Acta Scientiae*
594 *Cirumstantiae* **2001**, *21*, (4), 454-458.
- 595 (45) Mackay, D., *Multimedia Environmental Models: The Fugacity Approach, Second Edition*.
596 Taylor & Francis: 2001.
- 597 (46) Sweetman, A. J.; Cousins, I. T.; Seth, R.; Jones, K. C.; Mackay, D., A dynamic level IV
598 multimedia environmental model: Application to the fate of polychlorinated biphenyls in the United
599 Kingdom over a 60-year period. *Environmental Toxicology and Chemistry* **2002**, *21*, (5), 930-940.
- 600 (47) Moeckel, C.; Nizzetto, L.; Strandberg, B.; Lindroth, A.; Jones, K. C., Air-boreal forest transfer
601 and processing of polychlorinated biphenyls. *Environ. Sci. Technol.* **2009**, *43*, (14), 5282-9.
- 602 (48) Roudier, S.; Sancho, L. D.; Remus, R.; Aguado-Monsonet, M. *Best Available Techniques (BAT)*
603 *Reference Document for Iron and Steel Production: Industrial Emissions Directive 2010/75/EU:*
604 *Integrated Pollution Prevention and Control*; Institute for Prospective and Technological Studies,
605 Joint Research Centre: 2013.
- 606 (49) Fisher, R.; GARCIA CARCEDO, E.; ALAIZ ALVAREZ, E.; Pietruck, R., Influence of sinter
607 mix materials on the environmental impact of high productivity iron ore sintering. *EUR* **2005**,
608 (21429), 1-101.
- 609 (50) Hasanbeigi, A.; Price, L.; Lu, H.; Lan, W., Analysis of energy-efficiency opportunities for the
610 cement industry in Shandong Province, China: A case study of 16 cement plants. *Energy* **2010**, *35*,
611 (8), 3461-3473.
- 612 (51) Lammel, G.; Stemmler, I., Fractionation and current time trends of PCB congeners:
613 evolvement of distributions 1950–2010 studied using a global atmosphere-ocean general circulation
614 model. *Atmos. Chem. Phys.* **2012**, *12*, (15), 7199-7213.

615

616