

1 **ENVIRONMENTAL DISTRIBUTIONS OF BENZO[a]PYRENE (BaP) IN CHINA:**
2 **CURRENT AND FUTURE EMISSION REDUCTION SCENARIOS EXPLORED**
3 **USING A SPATIALLY EXPLICIT MULTI-MEDIA FATE MODEL**

4 **Ying Zhu¹, Shu Tao², Oliver R. Price³, Huizhong Shen², Kevin C. Jones¹, and Andrew J.**
5 **Sweetman^{1*}**

6 ¹**Lancaster Environment Centre, Lancaster University, Lancaster LA1 4YQ, United**
7 **Kingdom**

8 ²**Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences,**
9 **Peking University, Beijing 100871, China**

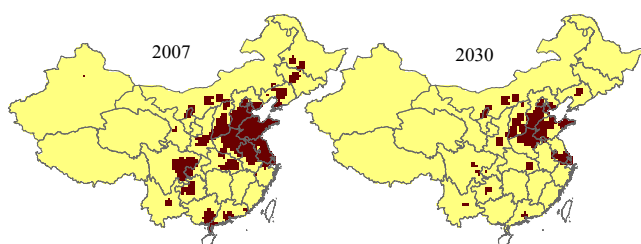
10 ³**Safety and Environmental Assurance Centre, Unilever, Sharnbrook MK44 1LQ,**
11 **United Kingdom**

12 * Corresponding author: Email: a.sweetman@lancaster.ac.uk; Tel: +44 1524 594715 and +44
13 1524 595994; Mob: 07549535969

14 **Abstract**

15 SESAMe v3.0, a spatially resolved multi-media fate model with $50 \times 50 \text{ km}^2$ resolution, has
16 been developed for China to predict environmental concentrations of Benzo(a)pyrene (BaP)
17 using an atmospheric emission inventory for 2007. Model predictions are compared with
18 environmental monitoring data obtained from an extensive review of the literature. The model
19 performs well in predicting multi-media concentrations and distributions. Predicted
20 concentrations are compared with guideline values; highest values with some
21 exceedances occur mainly in the North China Plain, Mid Inner Mongolia, and parts of three
22 northeast provinces, Xi'an, Shanghai and south of Jiangsu province, East Sichuan Basin,
23 middle of Guizhou and Guangzhou. Two potential future scenarios have been assessed using
24 SESAMe v3.0 for 2030 as BaP emission is reduced by 1) technological improvement for coal
25 consumption in energy production and industry sectors in Scenario 1 (Sc1); and 2)
26 technological improvement and control of indoor biomass burning for cooking and indoor
27 space heating, and prohibition of open burning of biomass in 2030 in Scenario 2 (Sc2). Sc2 is
28 more efficient in reducing the areas with exceedance of guideline values. Use of SESAMe
29 v3.0 provides insights on future research needs and can inform decision-making on options
30 for source reduction.

31 **TOC art**



32

33 **Introduction**

34 Polycyclic aromatic hydrocarbons (PAHs) inputs into the environment are largely derived
35 from inefficient combustion processes and several are known mutagens/carcinogens.¹⁻³ They
36 are of environmental and public health concern, so many countries have developed
37 environmental emission and quality criteria. They are also the subject of international
38 agreements and concern due to their long-range atmospheric transport potential.⁴⁻⁸ While
39 emissions and atmospheric concentrations have been declining in some developed countries,⁹⁻
40 ¹³ global emissions inventories show shifts in primary emissions to rapidly developing
41 countries,¹⁰ where inefficient fossil fuel combustion for power generation, metal production
42 and other industrial processes, along with transport, waste incineration and biomass burning
43 are often major sources.¹⁰ China is now a key part of the global inventory – constituting an
44 estimated 20% of the global emissions in 2007.¹⁰ It is therefore particularly important to
45 understand/confirm the key sources in China, where they are distributed and how efforts at
46 control may affect contemporary and future environmental concentrations.

47 Benzo(a)pyrene (BaP) is a carcinogenic high molecular weight PAH which is emitted to
48 atmosphere on fine particulates (PM10 and 2.5) and often associated with black carbon (BC).
49 The estimated emission of BaP across China in 2007 was ~1100 tonnes.¹⁰ However, spatial
50 resolution is important for China, a country of >1.3 billion people, because the population,
51 industrial development, PAH sources and environmental features vary widely. BaP is one of
52 the most studied compounds in China, with many papers reporting on PAHs in the
53 environment. This makes it an ideal choice as a test chemical when exploring the links
54 between sources and environmental levels with a multi-media environmental fate model.
55 Other studies have previously focussed on assessing human exposure to airborne PAHs via
56 inhalation¹⁴⁻¹⁶ and the distribution and ecological risk of waterborne PAHs in 7 major river
57 basins by collecting measured data from literature.¹⁷ China has also recently released a
58 ‘National Soil Pollution Gazette’ reporting on a countrywide soil pollution survey.¹⁸ This
59 survey may result in new legislation that addresses soil protection for China. However,
60 despite there now being: i. specific source inventory information; ii. a prevalence of air, water
61 and soil data; iii. Chinese national standards for BaP in air, water and soils;¹⁹⁻²¹ iv.
62 commitments made to reduce and regulate²² emissions of particulate matter and other air

63 pollutants by the State Council of China (SCC) five-year ‘Action Plan for Air Pollution
64 Control’,²³ there has been little attempt to integrate this information with multi-media
65 modelling tools or to assess the potential impacts of sources and their controls on ambient
66 levels and – ultimately – for risk management. That is therefore the purpose of this paper.

67 Previous multimedia models developed by other researchers have not been parameterized
68 with fine enough spatial resolution for China,²⁴⁻²⁶ nor at a national scale.²⁷⁻³⁰ As a result, we
69 have developed SESAMe v3.0, a spatially explicit multi-media fate model which has been
70 specifically parameterised for whole of mainland China with $50 \times 50 \text{ km}^2$ resolution. Initially
71 the model uses spatially resolved BaP atmospheric source inventory information for 2007¹⁰ to
72 generate model predictions to compare with environmental monitoring data obtained from the
73 literature over the period 1997 to 2011. Quantification of the transfer of BaP between media
74 is briefly discussed in relation to atmospheric transport, key storage media, ambient
75 distributions etc. Based on the good model performance, predicted high background regions
76 are identified across the country by comparing predicted ambient BaP levels with guideline
77 values made for air, freshwater and soils. We then move on to explore two possible future
78 scenarios where emission from coal and biomass burning are controlled and reduced by 2030
79 and making comparisons with the base year 2007. This is important, given the opportunity in
80 China for large scale state intervention and control of sources (e.g. power stations; waste
81 incinerators; vehicles), as well as changing patterns of individual behaviour on the
82 consumption of biofuels. SESAMe v3.0 has been used to assess the efficiency of emission
83 reduction in the two scenarios. Our purpose is to show how multimedia models such as
84 SESAMe v3.0 could be used to make informed decisions about possible source control
85 options.

86 **Materials and Methods**

87 **Model definition**

88 SESAMe v3.0 is an improved version of the previously described SESAMe model³¹ and
89 equations for transport processes are taken from Simplebox 3.24a³² and MAMI III³³ models;
90 it has a higher spatial resolution than SESAMe with 5468 independent $50 \times 50 \text{ km}^2$ grid cells
91 that cover mainland China and is therefore similar in structure to ChemCan²⁶ and
92 CHEMFRANCE³⁴. Each grid cell represents a region, which is surrounded by 8 adjacent $50 \times$
93 50 km^2 grid cells that constitute a movable continental scale; the regional scale and the
94 continental scale are connected by non-directional advective flow exchange, and so the model
95 doesn’t directly simulate how BaP is transported from one grid cell to another (details see
96 [Supporting Information \(SI\) and Figure S1](#)). This structure, therefore, considers the influence
97 of the emission and environmental processes of the surrounding region to each cell, which is

98 in contrast to ChemCan and CHEMFRANCE. Further improvements have been made for this
99 study, including: the addition of sea water compartments, referring to Simplebox 3.24a;³²
100 spatial data layers for soil density,³⁵ soil pH³⁵ and aerosol contents in air; the consideration of
101 the temperature effect on degradation rates, referring to Simplebox 3.24a³² (SI) and the
102 inclusion of agricultural soil irrigation by surface freshwater to ensure environmental
103 processes in the model more complete. For the agricultural soil irrigation, this version of the
104 model assumes that ~370 billion m³ water was consumed in the whole country for irrigation³⁶
105 and distributed uniformly to agricultural soil across the country, and that the irrigation water
106 in each grid cell originated from local freshwater sources in the same grid cell. Each grid cell
107 has compartments describing air, freshwater and sediment, sea water and sediment, natural
108 soil (defined as forest land, grassland, desert, wetland and all the other unused land),
109 agricultural soil (cropland soil), urban soil (rural residential land was included), natural
110 vegetation (on natural soil) and agricultural vegetation (on agricultural soil). There are 65
111 environmental parameters for each grid cell, in which 47 are fixed default values for all
112 regions and 18 are spatially variable (Table S1 and S2 in SI); the chemical parameters of BaP
113 are given in Table S3.

114 **Emission inventory**

115 The BaP emission dataset for the year 2007 used in this study originated from Shen et al.¹⁰
116 From their study, it was estimated that the emission of BaP in 2007 for the whole of mainland
117 China (exclusive of Taiwan and Hainan islands) was approximately 1032 tonnes. Nationally,
118 the major BaP sources in China are indoor biomass burning (43%, firewood and crop residue),
119 coke production (21%) and primary Al production (12%); the domestic coal burning is about
120 13%; motor vehicle emissions only constituted an estimated 2% and open fire agriculture
121 waste burning 1.2%.¹⁰ However, the dominant sources vary geographically as a complex
122 function of industrial activity, urbanization level, living standards, climate and policies across
123 China. For example, indoor biomass burning is an important source in large areas of the North
124 China Plain (NCP), Yangtze River Delta (YRD), Anhui, Hubei, Sichuan and the northeast
125 provinces etc.; coal consumed in coke production is a major source in Shanxi and Inner
126 Mongolia (Ordos); vehicle diesel is an important source for Shanghai and parts of Beijing.¹⁰
127 Sources were aggregated in the 50 × 50 km² grid, as detailed in SI, with the national
128 distribution shown in Figure 4a.

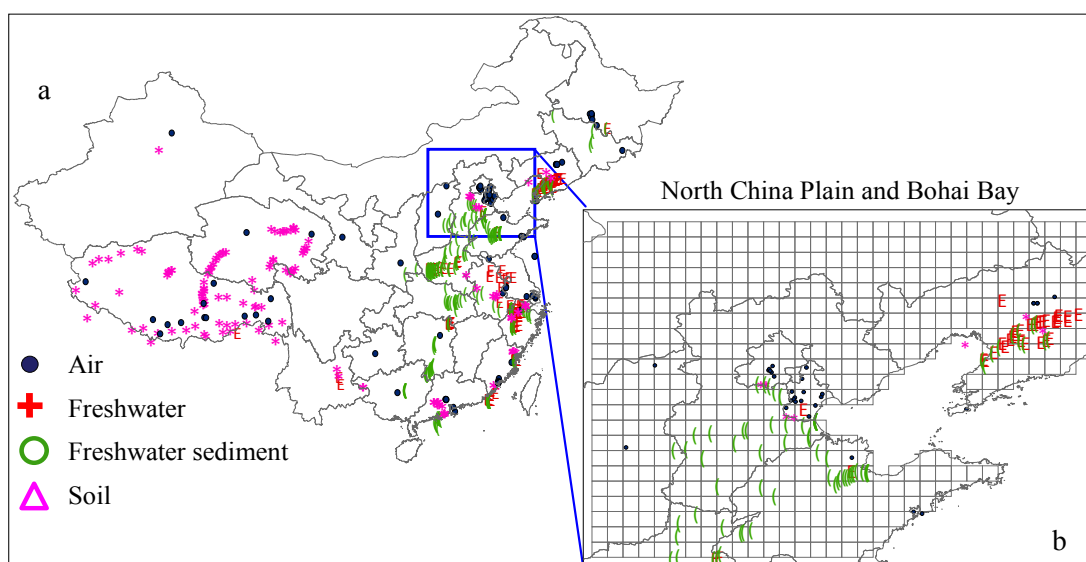
129 **Model evaluation**

130 Emissions for the base year of 2007 were run to steady state to generate predicted BaP
131 environmental concentrations, which could be compared with measured data and the
132 environmental guideline values/quality standards. The following guideline values were used:

133 ambient air quality values of 1 ng/m^3 as an annual average and 2.5 ng/m^3 as a daily average;¹⁹
134 soil values of 100 ng/g taken from the ‘Technical regulations for national soil contamination
135 assessment’³⁷ for Chinese soil pollution survey for soil pollution identification, to compare
136 with the results in the ‘National Soil Pollution Gazette’; freshwater guideline values of 2.8
137 ng/L from Chinese standards²⁰ and 15 ng/L from Canadian standards.³⁸

138 To evaluate the model, hundreds of peer-reviewed papers on BaP occurrence in China were
139 found. BaP concentration data were compiled without any filter for environmental media
140 from ca. 130 peer-reviewed literature sources where sampling site location information was
141 given. Focus was on the sampling years 1997-2011 and proximity to the 2007 base emission
142 year (Table S4). Figure 1 shows the location of the measurement data used for air, freshwater,
143 freshwater sediment and soil. When several sampling sites fall into the same grid cell (Figure
144 1b), the mean measured value was taken to compare with the predicted concentration in that
145 grid cell.

146 The dimensions of the selected continental and regional scales within the current model
147 structure are believed to be suitable to capture the majority of the emissions considering the
148 transport distance of BaP is probably a few hundreds of kilometres. However, to ensure that
149 the selected spatial scale is appropriate for BaP, it was compared with two previous versions
150 of the model which have different spatial scales and grid cell dimensions (more details see SI).

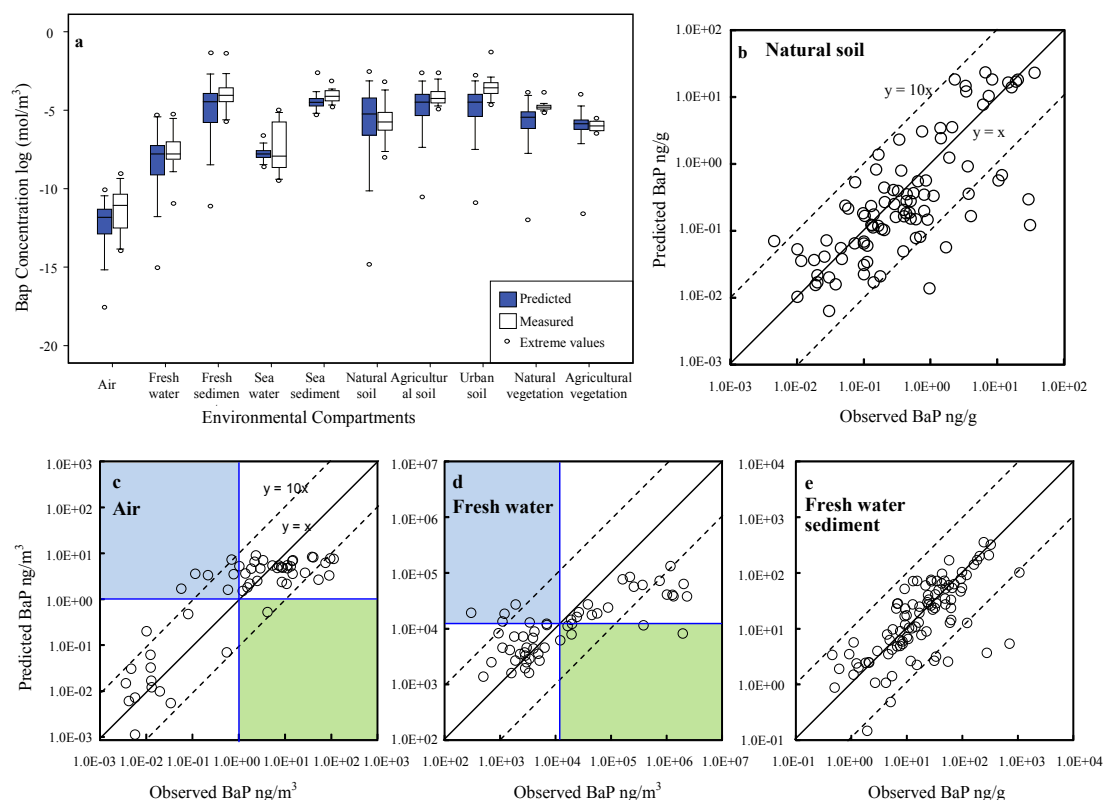


151
152 Figure 1. a, sampling sites of observed data taken from the literature across China; b,
153 illustrating the $50 \times 50 \text{ km}^2$ grid covering North China Plain and Bohai Bay in the model and
154 the overlay of sampling sites and model grid

155 The uncertainty of SESAME v3.0 was explored by Monte Carlo simulation (details see SI).
156 The BaP concentration in 10 media was calculated 10,000 times. Normal and lognormal

157 distributions were assessed according to the probability distribution of different input
 158 parameters. Values for the emission vector and the 18 environmental variable vectors were
 159 randomly produced based on their probability distributions. A variability-based sensitivity
 160 coefficient (SCV, see SI) was applied to the sensitivity analysis, considering the coefficient of
 161 variation of the 18 spatially variable parameters²⁸ for identifying influential parameters. The
 162 parameters with an SCV index > 0.1 were identified as most influential to the model output.

163 Model evaluation and discussion of results



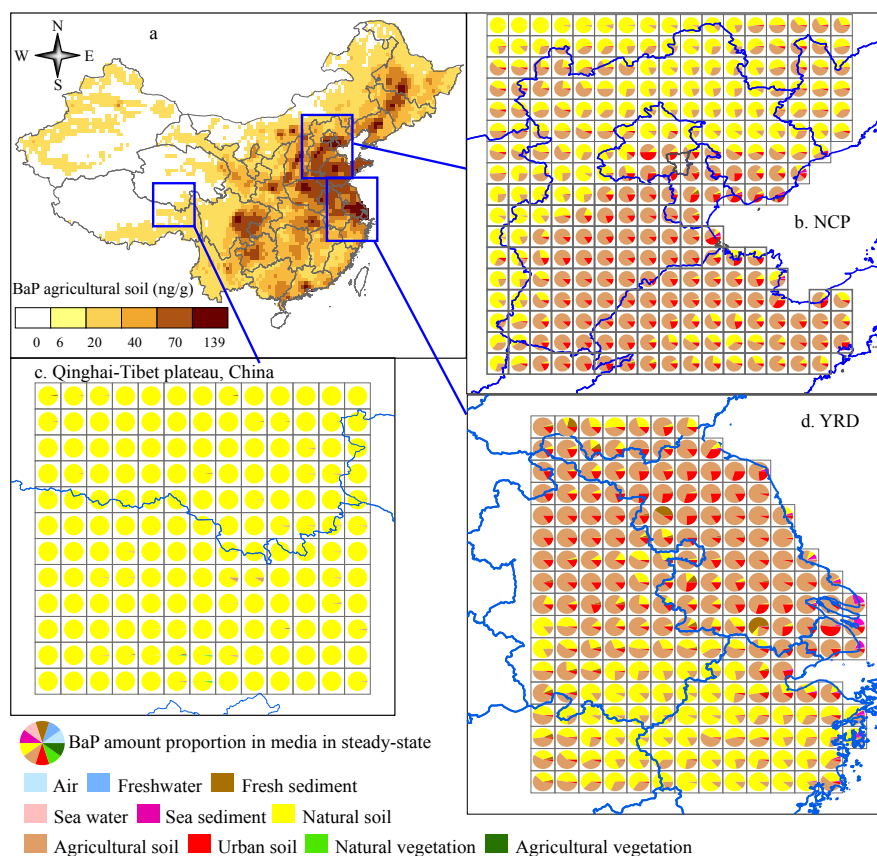
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165 Figure 2. a. Comparison of predicted (all predicted data) and measured BaP concentrations in
 166 each compartment across China; b-e. point-to-point comparison of BaP concentrations in
 167 natural soil, air, freshwater and sediment (only predicted data with corresponding measured
 168 data was used); the blue lines in c-d are BaP guideline values for air (1 ng/m³) and freshwater
 169 (1.5e+4 ng/m³)

170 This version of SESAMe predicts regional or countrywide ‘background’ concentrations. Such
 171 models do not identify hotspots, but predict the regional/grid cell averaged background levels.
 172 Figure 2a presents a comparison of predicted and measured BaP concentrations for each
 173 modelled environmental compartment. Generally, the agreement across all media is good,
 174 with the model capturing the range and actual concentrations very effectively. Most points
 175 fall within the 1:10 line, with many clustered around the 1:1 line (Figure 2b-e and Figure S4),
 176 which indicates that model performance is better for freshwater, sediment and soil than that

177 shown by Simplebox 3.0, EVn-BETR and IMPACT2002 in the study by Armitage et al.³⁹ The
178 selected dimensions of the regional and continental scales are important for models of this
179 type. The selected grid dimensions in SESAME v3.0 provide improved agreement with the
180 measurement data compared to two previous versions of the model with different grid
181 dimensions for regional and continental scales (SI). [Figure 2a](#) suggests that slightly systematic
182 under-prediction appears for several media but not for background concentrations. Often the
183 measured data collected from the literature highlights industrial parks or cities,^{40, 41}
184 agricultural soil irrigated by wastewater,⁴²⁻⁴⁴ locations where dense coal burning for heating
185 takes place,⁴⁵ urban areas with intensive transportation⁴⁶⁻⁴⁸ and so on, e.g. those points falling
186 below the lower 1:10 line. This may cause the underestimation shown in [Figure 2a](#), but the
187 model actually reflects the average situation in each grid cell. As precipitation is assumed to
188 be continuous in the model, it is most likely to overestimate deposition rates for BaP,⁴⁹
189 leading to the underestimation of air concentrations in some regions. However, this effect
190 appears to be limited at a national scale. The distribution of BaP concentrations in different
191 media is log-normal and the interquartile range can be found in [SI Figure S3](#) obtained from
192 the Monte Carlo simulation.

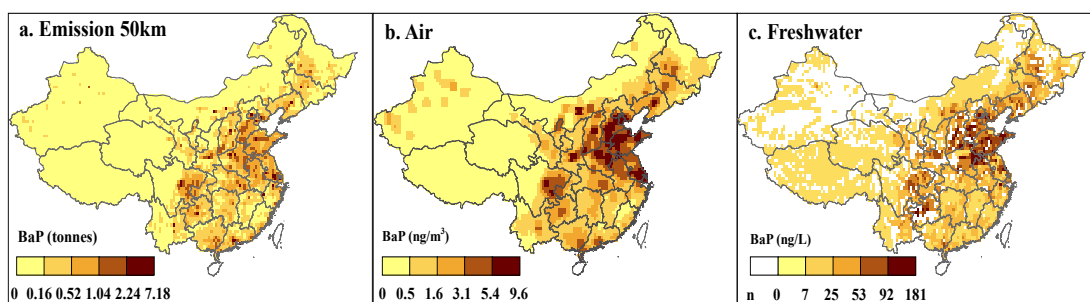
193 **BaP transport and partitioning between media:** The model predicts that after being
194 released to the atmosphere, BaP is mainly transported to soils, water and vegetation by wet
195 and dry deposition of particle-bound BaP; from water to sediment by sedimentation and
196 absorption; from vegetation to soils by litter production; and from soils to water by runoff
197 (see [Figure S5-S6](#)). The reverse processes - volatilization to air, desorption and re-suspension
198 from sediments and transpiration are relatively unimportant. Higher precipitation rates
199 increase the particle-bound BaP flux scavenged from air to soil or water. Areas with high soil
200 organic matter (e.g. northeast in Heilongjiang Province) will have greater storage and
201 retention of BaP. BaP reaches above ground vegetation primarily via particle-bound
202 deposition, rather than from soils – in agreement with measured and other modelling
203 studies.^{43, 50, 51}



204

205 Figure 3. a. predicted BaP background levels in agricultural soil; proportional mass of BaP in
 206 different environmental media under steady-state in b. North China Plain (NCP), c. Qinghai-
 207 Tibet plateau, d. Yangtze River Delta (YRD)

208 Soil is the primary sink for BaP in China; at steady state 99% of BaP will be found in soil.
 209 The model defines different soil land use categories and so nationally the loadings are ~56%
 210 in agricultural soils, 35% in natural soils and 8% in urban soils. Obviously there are major
 211 regional differences, for example, with most in natural soils in the Qinghai-Tibet plateau,
 212 most in agricultural soils in NCP and Jiangsu and urban soils in Shanghai (Figure 3). For
 213 some coastal or inland catchment regions, most BaP is found in sediments (Figure 3d).
 214 Storage in sediments is only significant for the mass balance in the coastal or freshwater
 215 catchment regions. The SCV index in Table S5 shows that the BaP concentration in a region
 216 can be affected by the emission and some environmental parameters in surrounding regions.
 217 Some grid cells are more influenced by emission and transport processes in nearby areas than
 218 those within its own cell.



219

220 Figure 4. a. BaP emission;¹⁰ predicted BaP background levels in air (b) and freshwater (c); n
 221 in c - no freshwater

222 **The spatial distribution of BaP:** The model predicts a range of regional background
 223 concentrations (5th - 95th percentiles, plus median) in the different media - as follows: air,
 224 0.002-4.6 ng/m³ (median, 0.4 ng/m³); soils, 0.002-51 ng/g for natural soil (0.9 ng/g) and
 225 0.05/0.06 - 56/58 ng/g for agricultural/urban soil (6.4 ng/g); fresh and sea water, 0.002-60 (4)
 226 ng/L and 1.5-11 (3.5) ng/L; fresh and marine sediments, 0.004-96 ng/g (6 ng/g) and 1.7-17
 227 ng/g (5 ng/g); vegetation, 0.0003-4 ng/g (0.1 ng/g) and 0.001-1.8 ng/g (0.2 ng/g) in natural
 228 and agricultural vegetation (Figure 3-4 and Figure S7-S8).

229 The predicted geographic distribution pattern is generally similar for air, soils and vegetation
 230 and the emission distribution pattern at the national scale (Figure 3-4). Air, soil and
 231 vegetation concentrations are generally predicted to be higher in areas such as NCP, Mid
 232 Inner Mongolia (Baotou, Ordos and Hohhot), part of the three northeast provinces (middle of
 233 Liaoning and Jilin provinces and south Heilongjiang), Xi'an in Shaanxi province, Shanghai
 234 and south of Jiangsu province, East Sichuan Basin, middle of Guizhou, Guangzhou in
 235 Guangdong province. However, environmental conditions can produce contrasting regional
 236 air and soils backgrounds. For example, a region in south Hebei has the same air
 237 concentration but nearly three times the concentration of BaP in soil compared to a region in
 238 west Inner Mongolia (Baotou). As the emission in the region of south Hebei is much higher
 239 and with higher precipitation rates in NCP compared to Inner Mongolia, more BaP is
 240 transferred to soils after being released to the atmosphere. Another interesting contrasting
 241 region in Liaoning in northeast China exhibits high soil levels close to that in another region
 242 in north Tianjin, but the air concentration is only half of that in the region in Tianjin. Runoff
 243 is similar in the two regions but the higher soil OC contents in Liaoning enhance BaP
 244 retention by soil. For most of these areas, the high background concentrations are caused
 245 mainly by indoor biomass burning (crop residue and firewood); in contrast, in Inner Mongolia,
 246 middle of Guizhou, Xi'an, some areas in Shanxi, Hebei and Shandong province in NCP, coal
 247 consumed by industry is also a key source.

248 Primarily BaP emissions reach aquatic systems via deposition and soil runoff. The geographic
249 distribution in freshwater and sediment viewed at the country scale is similar to that in air and
250 soil. However regional differences are apparent, caused by variation in the discharge volume
251 of rivers, runoff and soil OC etc. For example, BaP concentrations are extremely low in
252 Shanghai, south Jiangsu province (22 ng/L in freshwater, 35 ng/g in sediment) in the lower
253 reaches of the Yangtze River and the lower reaches of Yellow River (20 ng/L in freshwater,
254 31 ng/g in sediment) in NCP, but the air and soil concentrations in the two regions are
255 relatively high. A region with predicted high water concentrations (173 ng/L) in the northern
256 boundary of Anhui province has a moderate predicted BaP level in soil (68 ng/g), because the
257 soil OC contents are lower here and runoff can transfer more BaP from soil to water, but the
258 water discharge volume is also low.

259 Large areas in western China (e.g. Qinghai-Tibet plateau, large areas of Xinjiang) have lower
260 predicted median BaP values. They are generally in the order of 0.04 ng/m³ in air, 0.06 ng/g
261 in natural soil, 0.32 ng/g in agricultural and urban soil, 0.06 ng/L in freshwater, 0.1 ng/g in
262 sediment, and ca. 0.006 ng/g in natural and agricultural vegetation.

263 **Comparison of regional values and suggested guideline values for different media**

264 The previous sections show that the model performs well in predicting environmental
265 concentrations. Air, soil and freshwater are important media for public health and as noted
266 earlier - have guideline values suggested in China and elsewhere. For air, 2.5 ng/m³ has been
267 proposed as a daily average standard. The model predicts that this is exceeded in ca. 13% of
268 mainland China (e.g. NCP, three northeast provinces, Shanxi, north Ningxia, mid Inner
269 Mongolia, YRD, east Sichuan, mid Guizhou, Pearl River Delta (PRD), southeast Guangxi
270 etc.). A value of 1 ng/m³ has been proposed as an annual average. This is exceeded in ca. 32%
271 areas of mainland China (e.g. additional areas such as north and west of Guangxi, south
272 Ningxia, south Gansu, Hunan and coastal areas in Fujian province). This exceedance ratio is
273 close to 30% obtained in another Chinese study using the model CanMETOP which used a 1
274 km² spatial resolution.¹⁴ For soils, 100 ng/g has been adopted as a guideline value for all soils
275 in ‘Technical regulations for national soil contamination assessment’³⁷ for national soil
276 pollution survey. This is predicted to be exceeded in ca. 0.25% of mainland China. In contrast,
277 this is exceeded by 1.4% of the sampling sites across China in ‘National Soil Pollution
278 Gazette’,¹⁸ which reports the result of national soil pollution survey, probably because it is
279 indicated in the Gazette that many samples were taken from seriously contaminated land. 100
280 ng/g is also proposed as the Chinese soil quality standard value for agricultural soil. The
281 model predicts that it is exceeded by ca. 0.8% (about 22 thousand km²) agricultural soil.
282 These regions are mainly in Shanghai, south Jiangsu, Tianjin and Liaoning (Shenyang and
283 Liaoyang). For freshwater, 15 ng/L has been proposed as the Canadian guideline value. It is

284 exceeded by ca. 13% (ca. 20 thousand km²) of freshwater in mainland China (e.g. NCP,
285 northeast China provinces, Shaanxi, Guizhou and east Sichuan). A value of 2.8 ng/L is
286 proposed as the Chinese standard value. It is exceeded in ca. 40% (ca. 69 thousand km²) of
287 freshwater (covering almost half mainland China in the east). The overestimation of median
288 or low concentrations (blue area in Figure 2c-d) and the underestimation of high
289 concentrations (green area in Figure 2c-d) may cause a small but acceptable bias of
290 exceedance rates estimation but demonstrates good performance of the model.

291 **Future scenario selection**

292 In this section we illustrate how the model can be used to investigate the efficiency of source
293 reduction/controls. Two scenarios are used to generate possible future ambient concentrations
294 for the nominal year 2030.

295 The State Council of China (SCC) has made a five-year ‘Action Plan for Air Pollution
296 Control’²³. This refers to: controlling current major industrial emissions (e.g. by improved
297 combustion and stack controls); reducing the usage of and dependence on coal, particularly in
298 the Beijing-Tianjin-Hebei region, the Yangtze River Delta and the Pearl River Delta;
299 expediting the use of clean energy; controlling the number of vehicles or encouraging electric
300 vehicles and so on. Control measures that could be most relevant to BaP emissions relate to
301 the use of coal in industry and energy production.

302 Future scenario 1 (Sc 1): This considers control of coal consumption in two sectors
303 (centralised energy production and major industrial sources). The following assumptions and
304 principles were used to develop this scenario:

- 305 1. Coal consumption itself was not changed (2007 and 2030 levels assumed to be the
306 same), as it’s unrealistic to foresee a reduction in coal usage in 2030 considering that
307 the coal consumption/production in China has risen by ca. 48% from 2007 to 2012⁵²
308 and may currently be higher with many researchers arguing that coal consumption is
309 likely to remain high and key to China’s economic growth⁵³⁻⁵⁶. However, an action
310 plan was developed in 2013 to reduce the coal usage in certain regions and the
311 percentage of coal consumption in total energy consumption in China declined ca. 6%
312 from 2007 to 2012.⁵⁷
- 313 2. The emissions from key sources were assumed to be reduced between 2007 and 2030
314 by improvements in technology and combustion conditions. (see [Table 1](#) for details);
- 315 3. The fraction of uncontrolled and controlled activities was calculated by assuming
316 improved technologies are introduced as described by Bond et al.⁵⁸ in their
317 projections about future Black Carbon emission scenarios. Controlled coal boilers

318 were assumed to increase from 70% to 98% for energy production and from 46% to
319 87% for industry (see [SI](#) for further details and [Table 1](#));

320 4. Emission factors (EFs) were assumed stable over time for uncontrolled sources, while
321 improved EFs were employed for the controlled sources. These were derived from
322 Shen et al,¹⁰ with average values from their report being given in [Table 1](#).

323 5. The technology improvement in the energy and industry sectors was presumed to be
324 performed in the regions of China where coal consumption in the two sectors were >
325 50% to all sources, or in the regions with air backgrounds > 1 ng/m³.

326 Future scenario 2 (Sc 2): This scenario considered indoor domestic burning for cooking and
327 heating homes/buildings. Crop residues and firewood are important sources in some parts of
328 China, especially in areas with higher predicted background concentrations in this study^{10, 59}
329 and – whilst this can be an important source of indoor air pollution and associated human
330 health concerns^{60, 61} – here we focus on the potential contributions to ambient air. The
331 following assumptions were made:

332 1. A fraction of traditional and improved combustion stoves was also calculated by
333 assuming improved technologies are introduced as described by Bond et al.⁵⁸ The
334 percentage of improved stoves was assumed to increase from 34% to 84% for indoor
335 crop residue burning and from 37% to 85% for indoor firewood burning (see [Table 1](#)).

336 2. EFs were assumed stable over time for the individual technology as shown in [Table 1](#)
337 and the average values reported in Shen et al.'s research¹⁰ was adopted;

338 3. BaP EFs were assumed to be reduced by 40% when improved stoves are used for
339 indoor crop residue burning and indoor firewood burning ([Table 1](#)). At the same time,
340 half the indoor crop residue and indoor firewood burning activities were assumed to
341 be reduced due to urbanization or other alternatives (e.g. induction cookers)
342 introduced to rural residents for cooking and heating. It has been projected that half of
343 the rural areas in 2007 could become urbanized before 2030, which is based on the
344 urbanization rate of China in the past 10 years (1% each year conservatively) as
345 reported by World Bank⁶², and an assumption that this rate is maintained until 2030
346 in regions described in 4 below. Indoor biomass burning is banned in urban areas in
347 China;

348 4. The technology improvement and biomass burning reduction by urbanization or the
349 other alternatives were performed in the regions of China where the indoor biomass
350 (crop residue and firewood) burning was > 50% to all sources or in the regions with
351 air backgrounds > 1 ng/m³.

352 5. Open fire burning of agricultural waste is prohibited or regulated in many countries or
353 regions.⁶³⁻⁶⁵ China also has released a draft version of the 'Law of People's Republic

354 China on the Prevention and Control of Air Pollution' in which open burning of
 355 biomass in densely inhabited districts, areas near airports or the main traffic ways
 356 should be forbidden.⁶⁶ All open fire agricultural waste burning was assumed to be
 357 successfully banned by 2030.
 358

359 Table 1. Percent usage of different technologies in each sector (EFs mg/tonnes*¹⁰)

Coal (Sc1)	Sectors	Energy production		Industry	
	Technology	No control	Control	No control	Control
	2007	30% (1.2)	70% (1.4×10^{-2})	53% (40)	47% (0.46)
	2030	1.8% (1.2)	98.2% (1.4×10^{-2})	12.5% (40)	87.5% (0.46)
Biomass (Sc2)	Activities	Indoor crop residue burning		Indoor firewood burning	
	Technology	Traditional stove	Improved stove	Traditional woodstove	Improved woodstove
	2007	66% (1.8)	34% (0.69)	63% (1.5)	37% (0.56)
	2030	16% (1.8)	84% (0.69)	15% (1.5)	85% (0.56)

360 *Notes: the unit of EFs, mg/tonnes, means the amount of BaP (mg) released by burning one
 361 tonnes of coal or biomass

362 Outcomes predicted by the future scenarios

363 By adopting improved technology and EFs in Sc1, BaP emissions would decline by 90% and
 364 75% by 2030 in the energy production and industry sectors, respectively, from coal
 365 consumption in selected regions in Sc1. The total annual emission would decrease to ca. 900
 366 tonnes/yr in mainland China. By adopting improved stoves and EFs and the assumed
 367 urbanization scenario in Sc2, BaP emission would decline by 70% from indoor crop residue
 368 and firewood burning by 2030 in selected regions in Sc2. The total annual emission would
 369 decrease to ca. 610 tonnes/yr in mainland China. Since the two scenarios don't conflict, if
 370 implementing Sc1+Sc2, the total annual emission would decrease to ca. 470 tonnes/yr in
 371 mainland China.

372 Figure 5-6 show how more efficient Sc2 is than Sc1 in reducing the regional levels of BaP in
 373 air and freshwater. The areas where regional background concentrations would exceed 2.5
 374 ng/m³ and 1 ng/m³ in air are reduced by ca. 19% and 6% respectively in Sc1 and ca. 51% and
 375 40% in Sc2. Technology improvement for coal consumption is efficient in reducing
 376 backgrounds to < 2.5 ng/m³ only in areas such as Shanxi and boundary of north Ningxia and
 377 Inner Mongolia where there are coal mines (Figure 6). Implementation of the biomass
 378 burning and consumption control in Sc2 would reduce the regional concentration to < 2.5
 379 ng/m³ in the northeast provinces, NCP, Jiangsu, Anhui, Sichuan and Guangxi. Indeed, it
 380 would reduce BaP levels in large areas in these regions below the 1ng/m³ annual guideline
 381 value.

382 For freshwater (Figure S9), the areas where regional backgrounds would exceed 2.8 ng/L are
383 reduced by ca. 20% in Sc2 but show almost no reduction in Sc1, and those that would exceed
384 15 ng/L are reduced by ca. 20% and 50% respectively in Sc1 and Sc2. Technology
385 improvement on coal consumption in Sc1 would have little effect in reducing freshwater
386 backgrounds to < 2.8 ng/L but would work in small regions in north Ningxia, Gansu and
387 Yunnan in reducing the background concentration to <15 ng/L. Implementation of Sc2 would
388 be efficient in reducing the background level to < 15 ng/L in northeast China, east Sichuan,
389 south Henan and Hubei, and it is efficient in southeast China in reducing backgrounds to <
390 2.8 ng/L.

391 The current commitments in government regulations and action plans for air pollution
392 prevention and control focus mainly on reducing coal usage or improving technology on coal
393 consumption. These scenarios suggest that if greater efforts were put into biomass burning
394 control and technology improvement at the same time, as shown in Figure 5-6 (Sc1+2), the
395 areas with air background exceedance of 2.5 ng/m³ and 1 ng/m³ could be reduced by 82% and
396 54% respectively; and for freshwater backgrounds exceedance of 2.8 ng/L and 15 ng/L, it
397 could be reduced by 34% and 65%. It is also necessary to reduce emissions to both the
398 regional grid cell and to its surrounding area for effective pollution control considering the
399 emission and input from surrounding areas will also influence regional contamination (Figure
400 S10).

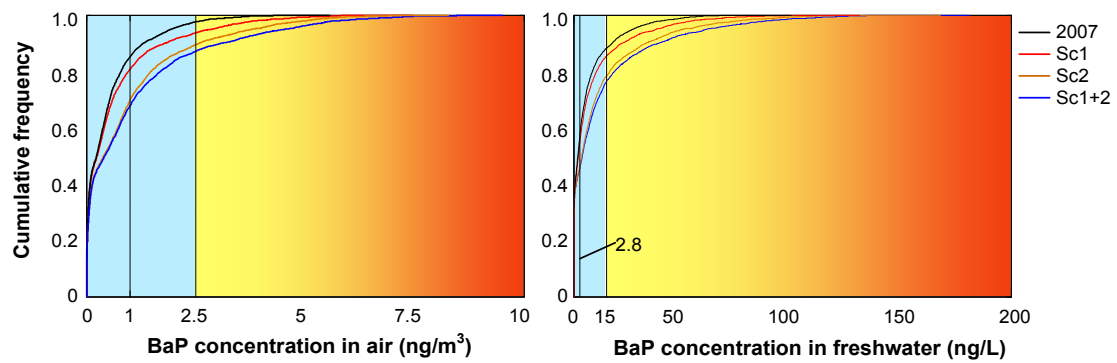
401 This section provides an illustration of the model application in the future scenario
402 discussions above. SESAMe v3.0 can perform well on assessing the efficiency of potential
403 implements made by governments or scientific research on preventing or controlling pollution
404 nationally or regionally. We stress that the scenarios are hypothetical and make no claim
405 about their likely development in the future.

406 **Potential future applications for SESAMe v3.0 as a pollution management tool**

407 In reality, future Chinese emission scenarios will be determined by a complex combination of
408 driving forces such as socio-economic development, large scale state intervention, technology
409 development, climate change and so on. They are evolving dynamically with high uncertainty,
410 so no realistic future scenarios can be predicted confidently.⁶⁷ However, research into possible
411 future scenarios can provide useful information to support decision/policy makers to prepare
412 for the challenge of reducing environmental pollution in the future.

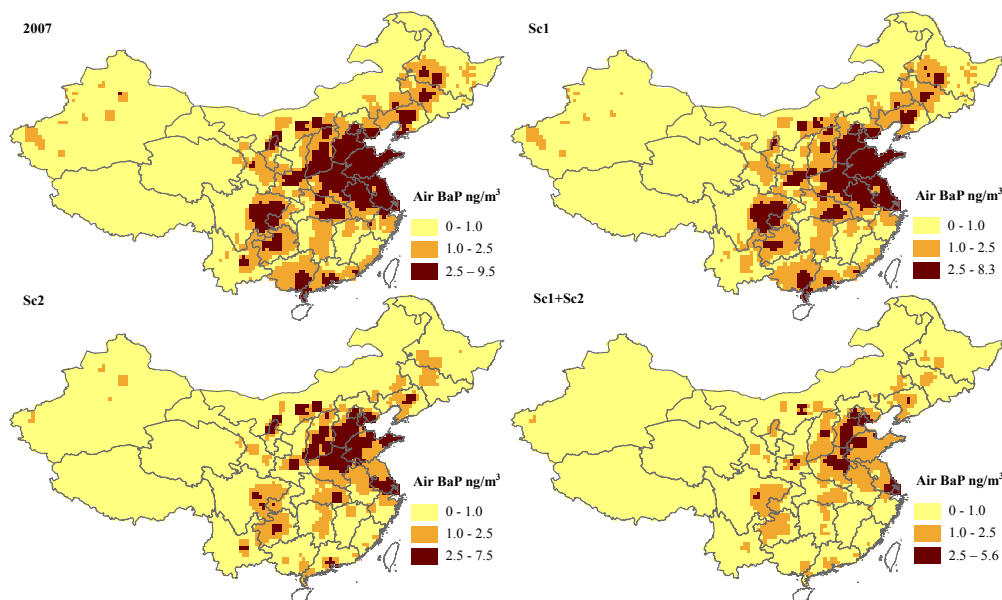
413 It has been demonstrated in this study that utilizing models such as SESAMe v3.0 for both
414 current and future scenario evaluation could provide valuable information for decision-
415 making on emission reduction strategies and future pollution management. Assessment of

416 current multi-media BaP concentrations across China assists in the identification of which
 417 regions would be most affected by the implementation of the two considered scenarios. Major
 418 emission sources were also considered when selecting regions for study. It can, therefore, be
 419 considered an economical way to evaluate the potential of commitments for pollution control
 420 in selected regions rather than in the whole country. Multi-media models have been selected
 421 for risk evaluation or pollution management and decision-making by a wide range of
 422 researchers and government institutions⁶⁸⁻⁷¹. SESAME v3.0, as a multi-media model, has the
 423 advantage of supporting science and decision-making for the evaluation of future pollution
 424 reduction and management.



425

426 Figure 5. Cumulative frequency of BaP concentration in air (left) and in freshwater (right)
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428

429

430

Figure 6. BaP air concentration in 2007, Sc1, Sc2 and Sc1+Sc2

431

Acknowledgement

432 We thank the Safety and Environmental Assurance Centre, Unilever for funding the research.
433 The production of emission inventory was funded by National Natural Science Foundation of
434 China (41390240). Thanks also to Antonio Franco (Unilever) for constructive comments on
435 the manuscript.

436 **Supporting information**

437 Additional information on description of methods, input model parameters, the literature for
438 collecting measured data and the output figures can be found in supporting information
439 document. This information is available free of charge via the Internet at <http://pubs.acs.org>.

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