

Supplementary Information for

On the Occurrence of Single- and Double-Peaked Emission Profiles of Synthetic Chemicals

Li Li and Frank Wania

Department of Physical and Environmental Sciences, University of Toronto Scarborough, 1265 Military Trail, Toronto, Ontario, Canada M1C 1A4

* Corresponding Author: Li L.; E-mail: environ.li@mail.utoronto.ca. Phone: +1 (647) 601-4450.

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Text S1 Mathematical derivations of a simplified case

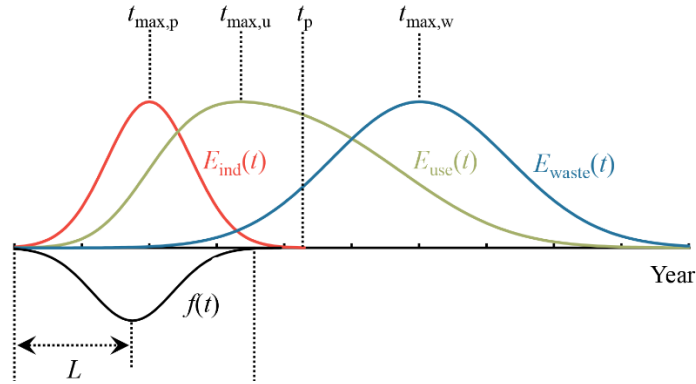


Figure: Notes used in the mathematical derivations

Assumptions

In order to facilitate the mathematical derivation (Theorems S1 – S4), without loss of generality, we make the following simplifying assumptions:

Assumption 1 The temporal trend of production, $P(t)$, is single peaked, which is positive within the domain of $(0, t_p)$ and zero otherwise, with a single maximum occurring at $t = t_{\max,p}$ (see the figure above), i.e.,

$$\frac{dP(t)}{dt} = \begin{cases} > 0, & \text{if } t \in (0, t_{\max,p}) \\ = 0, & \text{if } t = t_{\max,p} \\ < 0, & \text{if } t \in (t_{\max,p}, t_p) \end{cases}, \quad (1)$$

$$\text{and } \frac{d^2P(t_{\max,p})}{dt^2} < 0. \quad (2)$$

The cumulative production over the simulation period is defined as P_{tot} , i.e.,

$$\int_0^\infty P(\tau) d\tau = \int_0^{t_p} P(\tau) d\tau = P_{\text{tot}} \quad (3)$$

Note that such a single-peaked production profile is the general case of persistent and pseudo-persistent synthetic chemicals.¹ We do not consider the special case that production and use of a chemical reemerge long after the phase-out of its original intended use, e.g., the recent production and application of DDT for regional malaria control after its agricultural use has been restricted in 1980s.

Assumption 2 The lifespan distribution function of a chemical, $f(t)$, is defined for $t \geq 0$, and it satisfies:²

$$\int_0^{\infty} f(t)dt = 1. \quad (4)$$

Assumption 3 For their entire domains, $P(t)$, $U(t)$, $W(t)$, $f(t)$ and their combinations are continuously differentiable.

Assumption 4 The change in in-use and waste stocks due to emission is negligible. Meanwhile, 100% of the generated waste enters waste stocks ($F_{\text{waste}} = 1$). Therefore, eqs.2 and 4 in the main text are simplified to:

$$\frac{dU(t)}{dt} = P(t) \cdot (1 - EF_{\text{ind}}) - D(t) \quad (5)$$

$$\frac{dW(t)}{dt} = D(t) - k \cdot W(t) \quad (6)$$

Theorem 1

In-use stock $U(t)$ is single peaked, i.e., $U(t)$ has a single stationary point.

Proof:

Eq. 3 in the main text and eq. S5 (**Assumption 4**) give the first derivative of $U(t)$:

$$\frac{dU(t)}{dt} = (1 - EF_{\text{ind}}) \cdot \left[P(t) - \int_0^t P(\tau) \cdot f(t - \tau) d\tau \right] \quad (7)$$

Let $\frac{dU(t)}{dt} = 0$, we have:

$$EF_{\text{ind}} = 1 \text{ or} \quad (8)$$

$$P(t) - \int_0^t P(\tau) \cdot f(t - \tau) d\tau = 0. \quad (9)$$

Eq. S9 is a Volterra integral equation of convolution type. Laplace transforming eq. S9 according to the definition of the unilateral Laplace transform:

$$\mathcal{L}\{g\}(s) = \int_0^\infty g(t) e^{-st} dt, \quad (10)$$

and given the convolution theorem:

$$\mathcal{L}\{g * h\}(s) = \mathcal{L}\{g\}(s) \cdot \mathcal{L}\{h\}(s) \quad (* \text{ denotes convolution}), \quad (11)$$

we obtain

$$\mathcal{L}\{P\}(s) = \mathcal{L}\{P\}(s) \cdot \mathcal{L}\{f\}(s). \quad (12)$$

Because $\mathcal{L}\{P\}(s) = \int_0^\infty P(t) e^{-st} dt > 0$, eq. S12 holds when

$$\mathcal{L}\{f\}(s) = 1, \quad (13)$$

Namely,

$$\int_0^\infty f(t) e^{-st} dt = 1. \quad (14)$$

A comparison between eqs. S4 (**Assumption 2**) and S14 shows $s = 0$.

This means that $\frac{dU(t)}{dt} = 0$ holds if, and only if, $s = 0$.

Meanwhile,

at the point $t = t_{\text{max},p}$,

$$\begin{aligned}
\left. \frac{dU(t)}{dt} \right|_{t=t_{\max,p}} &= (1 - EF_{\text{ind}}) \cdot \left[P(t_{\max,p}) - \int_0^{t_{\max,p}} P(\tau) \cdot f(t_{\max,p} - \tau) d\tau \right] \\
&> (1 - EF_{\text{ind}}) \cdot \left[P(t_{\max,p}) - \int_0^{t_{\max,p}} P(t_{\max,p}) \cdot f(t_{\max,p} - \tau) d\tau \right] \\
&= (1 - EF_{\text{ind}}) \cdot \left[P(t_{\max,p}) - P(t_{\max,p}) \cdot \int_0^{t_{\max,p}} f(t_{\max,p} - \tau) d\tau \right] \\
&= (1 - EF_{\text{ind}}) \cdot \left\{ P(t_{\max,p}) \cdot \left[1 - \int_0^{t_{\max,p}} f(t_{\max,p} - \tau) d\tau \right] \right\} \geq 0,
\end{aligned} \tag{15}$$

and at the point $t = t_p$,

$$\begin{aligned}
\left. \frac{dU(t)}{dt} \right|_{t=t_p} &= (1 - EF_{\text{ind}}) \cdot \left[P(t_p) - \int_0^{t_p} P(\tau) \cdot f(t_p - \tau) d\tau \right] \\
&= (1 - EF_{\text{ind}}) \cdot \left[0 - \int_0^{t_p} P(\tau) \cdot f(t_p - \tau) d\tau \right] \leq 0.
\end{aligned} \tag{16}$$

Therefore, the stationary point where $\frac{dU(t)}{dt} = 0$ (denoted as $t = t_{\max,u}$, see the figure above) is a maximal turning point (i.e., a peak rather than a valley) and locate within the interval $(t_{\max,p}, t_p)$.

Theorem 1 is proved.

Theorem 2

Waste stock $W(t)$ is single peaked, i.e., $W(t)$ has a single stationary point.

Proof:

According to the general solution to the “convolution equation”³ that represents the general form of linkage between input and output using coupled linear differential equations, the analytical solution to eq. S6 (Assumption 4) is

$$W(t) = e^{-k \cdot t} \cdot \int_0^t D(\tau) \cdot e^{k \cdot \tau} d\tau. \quad (17)$$

Therefore, the first derivative of $W(t)$ is

$$\frac{dW(t)}{dt} = D(t) - k \cdot W(t) = D(t) - k e^{-k \cdot t} \cdot \int_0^t D(\tau) \cdot e^{k \cdot \tau} d\tau. \quad (18)$$

Note that if the pseudo-first-order degradation rate constant k approaches 0, i.e., the degradation half-life is infinite, eq. S18 is simplified to

$$\frac{dW(t)}{dt} = D(t) \geq 0. \quad (19)$$

This means that, as time elapse, $W(t)$ will increase until ultimately level off with an asymptote towards

$$\lim_{t \rightarrow +\infty} W(t) = \int_0^\infty D(\tau) d\tau = \int_0^\infty \left[\int_0^\tau P(u) \cdot (1 - EF_{\text{ind}}) \cdot f(\tau - u) du \right] d\tau = P_{\text{tot}} \cdot (1 - EF_{\text{ind}}) \quad (20)$$

This means that all produced chemicals will eventually accumulate in waste stock if our observation lasts sufficiently long.

When the pseudo-first-order degradation rate constant k is non-zero, i.e., the degradation half-life is finite, let $\frac{dW(t)}{dt} = 0$, we have:

$$D(t) - k e^{-k \cdot t} \cdot \int_0^t D(\tau) \cdot e^{k \cdot \tau} d\tau = 0. \quad (21)$$

Namely,

$$D(t) \cdot e^{k \cdot t} - k \cdot \int_0^t D(\tau) \cdot e^{k \cdot \tau} d\tau = 0. \quad (22)$$

Defining $g(t) = D(t) \cdot e^{k \cdot t}$, eq. S22 can be rewritten as

$$g(t) - k \cdot \int_0^t g(\tau) d\tau = 0. \quad (23)$$

Laplace transforming eq. S23 results in

$$\mathcal{L}\{g\}(s) - \frac{k}{s} \cdot \mathcal{L}\{g\}(s) = 0, \quad (24)$$

Therefore, eq. S24 holds if, and only if, $s = k$. This means there is only a single case that satisfies

$$\frac{dW(t)}{dt} = 0.$$

In addition, we assume that $\frac{dW(t)}{dt} = 0$ holds at $t = t_{\max, w}$ (see the figure above). Given that $D(\tau) > 0$,

we have

$$W(t_{\max, w}) = e^{-k \cdot t_{\max, w}} \cdot \int_0^{t_{\max, w}} D(\tau) \cdot e^{k \cdot \tau} d\tau > 0 = W(0) \quad (25)$$

Therefore, the stationary point $t = t_{\max, w}$ is a maximal turning point (i.e., a peak rather than a valley).

Theorem 2 is proved.

Theorem 3

The sub-total of E_{ind} and E_{use} , denoted as $E_{\text{ind/use}}(t)$, is single peaked, i.e., it has a single stationary point.

Proof:

$E_{\text{ind/use}}(t)$ is calculated as

$$E_{\text{ind/use}}(t) = EF_{\text{ind}} \cdot P(t) + EF_{\text{use}} \cdot U(t). \quad (26)$$

Therefore, its first derivative is

$$\begin{aligned} \frac{dE_{\text{ind/use}}(t)}{dt} &= EF_{\text{ind}} \cdot \frac{dP(t)}{dt} + EF_{\text{use}} \cdot \frac{dU(t)}{dt} \\ &= EF_{\text{ind}} \cdot \frac{dP(t)}{dt} + EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot \left[P(t) - \int_0^t P(\tau) \cdot f(t - \tau) d\tau \right] \\ &= EF_{\text{ind}} \cdot \frac{dP(t)}{dt} + EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot P(t) - EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot \int_0^t P(\tau) \cdot f(t - \tau) d\tau. \end{aligned} \quad (27)$$

Let $\frac{dE_{\text{ind/use}}(t)}{dt} = 0$, we have

$$EF_{\text{ind}} \cdot \frac{dP(t)}{dt} + EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot P(t) - EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot \int_0^t P(\tau) \cdot f(t - \tau) d\tau = 0 \quad (28)$$

Laplace transforming eq. S28 results in

$$EF_{\text{ind}} \cdot s \cdot \mathcal{L}\{P\}(s) + EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot \mathcal{L}\{P\}(s) - EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot \mathcal{L}\{P\}(s) \cdot \mathcal{L}\{f\}(s) = 0 \quad (29)$$

Given that $\mathcal{L}\{P\}(s) > 0$, it can be cancelled from the equation. Thus, eq. S29 holds if

$$\mathcal{L}\{f\}(s) = \frac{EF_{\text{ind}}}{EF_{\text{use}} \cdot (1 - EF_{\text{ind}})} \cdot s + 1 \quad (30)$$

A comparison between eqs. S4 (**Assumption 2**) and S30 shows $s = 0$.

This means $\frac{dE_{\text{ind/use}}(t)}{dt} = 0$ holds if, and only if, $s = 0$, i.e., there is only a single case that satisfies

$$\frac{dE_{\text{ind/use}}(t)}{dt} = 0.$$

Meanwhile, given $P(t) \geq 0$ and $U(t) \geq 0$, it is obvious that the stationary point where $\frac{dE_{\text{ind/use}}(t)}{dt} = 0$ is a maximal turning point (i.e., a peak rather than a valley).

Theorem 3 is proved.

Theorem 4

The sub-total of E_{use} and E_{waste} , denoted as $E_{\text{use/waste}}(t)$, is single peaked, i.e., it has a single stationary point.

Proof:

$E_{\text{use/waste}}(t)$ is calculated as

$$E_{\text{use/waste}}(t) = EF_{\text{use}} \cdot U(t) + EF_{\text{waste}} \cdot W(t) \quad (31)$$

Therefore, its first derivative is

$$\begin{aligned} \frac{dE_{\text{use/waste}}(t)}{dt} &= EF_{\text{use}} \cdot \frac{dU(t)}{dt} + EF_{\text{waste}} \cdot \frac{dW(t)}{dt} \\ &= EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot [P(t) - D(t)] + EF_{\text{waste}} \cdot [D(t) - k \cdot W(t)] \\ &= EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot P(t) + [EF_{\text{waste}} - EF_{\text{use}} \cdot (1 - EF_{\text{ind}})] \cdot D(t) - EF_{\text{waste}} \cdot k \cdot W(t). \end{aligned} \quad (32)$$

Let $\frac{dE_{\text{use/waste}}(t)}{dt} = 0$, we have

$$EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot P(t) + [EF_{\text{waste}} - EF_{\text{use}} \cdot (1 - EF_{\text{ind}})] \cdot D(t) - EF_{\text{waste}} \cdot k \cdot W(t) = 0. \quad (33)$$

Because

$$\mathcal{L}\{D\}(s) = \mathcal{L}\{P\}(s) \cdot \mathcal{L}\{f\}(s), \text{ and} \quad (34)$$

$$\mathcal{L}\{W\}(s) = \mathcal{L}\{D\}(s) \cdot \frac{1}{s + k}, \quad (35)$$

Laplace transforming eq. S33 results in

$$\begin{aligned} &EF_{\text{use}} \cdot (1 - EF_{\text{ind}}) \cdot \mathcal{L}\{P\}(s) + [EF_{\text{waste}} - EF_{\text{use}} \cdot (1 - EF_{\text{ind}})] \cdot \mathcal{L}\{P\}(s) \cdot \mathcal{L}\{f\}(s) \\ &- EF_{\text{waste}} \cdot \frac{k}{s + k} \cdot \mathcal{L}\{P\}(s) \cdot \mathcal{L}\{f\}(s) = 0, \end{aligned} \quad (36)$$

Given that $\mathcal{L}\{P\}(s) > 0$, it can be cancelled from the equation. Thus, eq. S36 holds if

$$\mathcal{L}\{f\}(s) = \frac{1}{1 + \frac{s}{s + k} \cdot \frac{EF_{\text{waste}}}{EF_{\text{use}} \cdot (1 - EF_{\text{ind}})}} \quad (37)$$

A comparison between eqs. S4 (**Assumption 2**) and S37 shows $s = 0$.

This means $\frac{dE_{\text{use/waste}}(t)}{dt} = 0$ holds if, and only if, $s = 0$, i.e., there is only a single case that satisfies

$$\frac{dE_{\text{use/waste}}(t)}{dt} = 0.$$

Meanwhile, given $U(t) \geq 0$ and $W(t) \geq 0$, it is obvious that the stationary point where $\frac{dE_{\text{use/waste}}(t)}{dt} = 0$ is a maximal turning point (i.e., a peak rather than a valley).

Theorem 4 is proved.

Table S1 Parameters used in the realistic calculations for actual chemicals

	Fitted $P(t)$	EF_{ind}	EF_{use}	EF_{waste}	F_{waste}	LS (year)	HL (year)*
PFOA (all media)	Peak year 1996	75%	Negligible	Negligible	–	10 (normal dist.) ⁴	6900 ⁵
Developed regions	Standard dev. 10.0						
β-HCH (atmosphere)	Peak year 1972	Negligible	Negligible	10.5%	–	Negligible	2 ⁶
Worldwide	Standard dev. 10.1						
PCB28 (atmosphere)	Peak year 1969	5.0×10^{-2}	8.26×10^{-3}	4.56×10^{-3}	69%	16 (bathtub dist.) ⁷	4.8 ⁸
Worldwide	Standard dev. 6.5						
PCB180 (atmosphere)	Peak year 1967	5.0×10^{-2}	1.73×10^{-3}	1.01×10^{-3}	71%	16 (bathtub dist.) ⁷	22 ⁸
Worldwide	Standard dev. 7.9						
HBCDD (all media)	Peak year 2013	3.2×10^{-4}	2.48×10^{-6}	1.43×10^{-4}	85%	32 (normal dist.) ⁹	1.48 ⁹
Mainland China	Standard dev. 3.3						

Notes:

* Since our illustrative calculations aim to approximate the emission profile in magnitude, we do not consider the temporal and spatial variations in LS and HL but use the values as they are reported in the literature without adjustment, while we recognize that the two parameters are country and time specific.

PFOA

- (1) Information on the annual production of PFOA (in the form of its salts) in developed regions (defined as Japan, Western Europe and the United States according to Wang et al.⁴) was collected from Wang et al.⁴. The annual production data were then fitted with a Gaussian distribution function, resulting in a mean of 1996 and a standard deviation of 10.0 years ($R^2 = 0.7876$). This fitting result means that 68% of cumulative annual production occurs within a window of respective 10.0 years (corresponding to 95% of cumulative annual production occurring within a window of respective 19.6 years) before and after the year 1996.
- (2) Industrial emissions of PFOA are associated with three main industrial processes:
- (i) 4.3% of produced PFOA is released during production stage (SI Text S3 of Li et al.¹⁰);
 - (ii) ~75% of the remainder released during fluoropolymer manufacture (SI S1.1.3 of Wang et al.⁴, assuming no intentional pollution reduction for the period); and
 - (iii) 26% of the remainder is released during the industrial use of aqueous fluoropolymer dispersion products which accounted for 15% of the manufactured fluoropolymers (i.e., , SI Text S1.1.5 of Wang et al.⁴).

Thus, EF_{ind} is calculated to be

$$4.3\% + (1 - 4.3\%) \times 75\% + (1 - 4.3\%) \times (1 - 75\%) \times 15\% \times 26\% = \sim 75\%.$$

Since most of the rest of PFOA is decomposed during industrial heat processing, merely <3% of produced PFOA remains as impurities in final consumer products. As such, emissions at use and waste disposal phases are negligible.

- (3) The reported emission estimate for evaluating our modeling performance is an average of upper and lower boundaries of the “plausible” scenario in ref.⁴.

β-HCH

- (1) The annual production of β-HCH worldwide was calculated based on the global annual consumption of technical HCH¹¹ and the typical content of β-HCH in the technical mixture (9%),⁶ assuming that a pesticide is consumed at the year it is produced. The annual production data were then fitted with a Gaussian distribution function, resulting in a mean of 1972 and a standard deviation of 10.1 years ($R^2 = 0.8627$). This fitting result means that 68% of cumulative annual production occurs within a window of respective 10.1 years (corresponding to 95% of cumulative annual production occurring within a window of respective 19.8 years) before and after the year 1972.
- (2) There are three modes of HCH application: (i) spraying, (ii) soil incorporation (tilling), and (iii) seed treatment before planting, each of which is associated with a location-specific emission factor. The overall fraction of each mode in the global total HCH application was calculated based on (i) a fractional distribution of β-HCH among the three application modes in different climate zones (Table

3 of Li et al.⁶), and (ii) zonal HCH usage (Figure 1 of Li et al.⁶).

The overall (atmospheric) emission factor of β -HCH for each mode was calculated based on (i) the emission factor for each mode in different climate zones (Figure 2 of Li et al.⁶) and (ii) zonal HCH usage (Figure 1 of Li et al.⁶).

(3) The reported emission estimate for evaluating our modeling performance was taken from ref.⁶.

PCBs

(1) Information on the annual production of PCB28 and PCB180 worldwide was collected from Breivik et al.⁷. For each congener, the annual production data were fitted with a Gaussian distribution function, resulting in means of 1969 (PCB28) and 1967 (PCB180), and standard deviations of 6.5 (PCB28; $R^2 = 0.9531$) and 7.9 years (PCB180; $R^2 = 0.7179$). The fitting results mean that 68% of cumulative annual production of PCB28 occurs within a window of respective 6.5 years (corresponding to 95% of cumulative annual production occurring within a window of respective 12.7 years) before and after the year 1969, and that of PCB180 occurs within a window of respective 7.9 years (corresponding to 95% of cumulative annual production occurring within a window of respective 15.5 years) before and after the year 1967.

(2) For each congener,

- EF_{ind} was taken from ref.¹², which assumes that 5% of PCBs are emitted “as a hypothetical surrogate for omitted point source releases” from industrial activities.
- EF_{use} was an average of emission factors for (a) leakage and volatilization (Sections 2.9.1 and 2.9.2 of Breivik et al.⁸) and (b) accidental releases (Section 2.8 of Breivik et al.⁸), weighted by the global overall “use factors” (i.e., the percentage of PCBs used in individual applications considered) of individual applications. Based on information provided in Breivik et al.⁸, the overall use factors on the global scale are calculated to be 18% (open usage), 11% (nominally closed systems), 22% (small capacitors) and 49% (closed systems) for PCB28, and 22%, 12%, 21% and 46% for PCB180.
- EF_{waste} was an average of emission factors for (a) landfill (Section 2.9.3 of Breivik et al.⁸), (b) open burning (Section 2.9.4 of Breivik et al.⁸), and (c) incineration (Section 2.9.5 of Breivik et al.⁸), weighted by the global overall use factors of individual applications. Note that, in Breivik et al.⁸, the emission factor from landfill is on a basis of waste stock (i.e., a continuous process) whereas that from open burning and incineration is on a basis of the amount of products discarded per annum (i.e., an instantaneous process). Since our simple model links emission factors with waste stock, for each PCB congener, we convert the literature-reported instantaneous emission factors of open burning and incineration (Tables 3 and 4 of Breivik et al.⁸) into continuous ones by simply dividing the instantaneous emission factor by the HL of the congener in waste stock.

(3) For each congener, F_{waste} was assumed to be 100% minus the fraction subject to safe destruction (Section 2.9.5 of Breivik et al.⁸). The fraction of waste subject to safe destruction was calculated based

on the “disposal factors” of individual PCB applications (Table 3 of Breivik et al.⁸) and the global overall use factors.

- (4) The reported emission estimate for evaluating our modeling performance was taken from the “baseline” scenario in ref.¹³.

HBCDD

- (1) Information on the annual production of HBCDD in mainland China was obtained from Li et al.⁹. the annual production data were fitted with a Gaussian distribution function, resulting in a mean of 2013 and a standard deviation of 3.3 years ($R^2 = 0.9856$). The fitting result means that 68% of cumulative annual production of HBCDD occurs within a window of respective 3.3 years (corresponding to 95% of cumulative annual production occurring within a window of respective 6.5 years) before and after the year 2013.

- (2) EF_{ind} and EF_{use} were averages of emission factors for (i) expanded polystyrenes, (ii) extruded polystyrenes, and (iii) textiles, weighted by the distribution ratio of HBCDDs among the three applications (72%, 27% and 1%, respectively). In addition, EF_{ind} was a combination of emission factors for (a) production of technical HBCDDs (Table 1 of Li et al.⁹) and (b) processing of products in the three applications (Table 1 of Li et al.⁹).

EF_{waste} was an average of emission factors for (a) controlled landfill, and (b) backfill or illegal open dumping, weighted by the fractions of construct and demolition waste disposed by the two options. As the fractions of controlled landfill (increases from 15% to 69% over the simulation period) and backfill or illegal open dumping (decreases from 85% to 1%) are time dependent, for simplification, here we assumed the fractions to be fixed at 55% and 30%, respectively.

- (3) F_{waste} was assumed to be 100% minus the fraction of waste subject to sorting, recovery and recycling. Since this fraction is time dependent (keeps increasing from <1% to 30% since 2011), here we assume it to be fixed at 15% for simplification.
- (4) The reported emission estimate for evaluating our modeling performance was taken from the “best estimate” (i.e., median values) in ref.⁹.

Figure S1 In-use and waste stocks (as percentage in total production over the simulation period) of hypothetical chemicals as a function of time scales of residence in stocks (product lifespan and degradation half-life in waste stock)

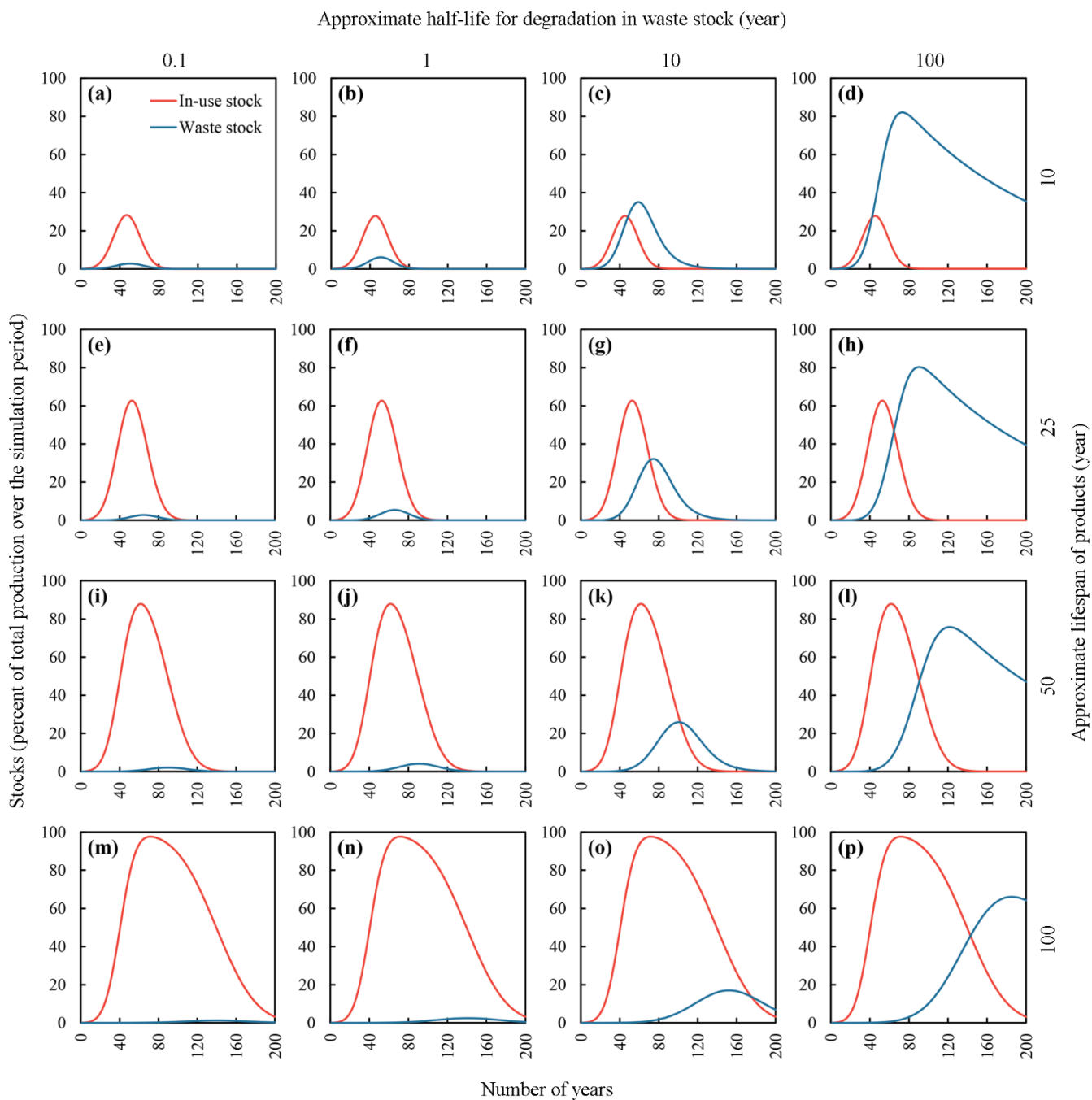
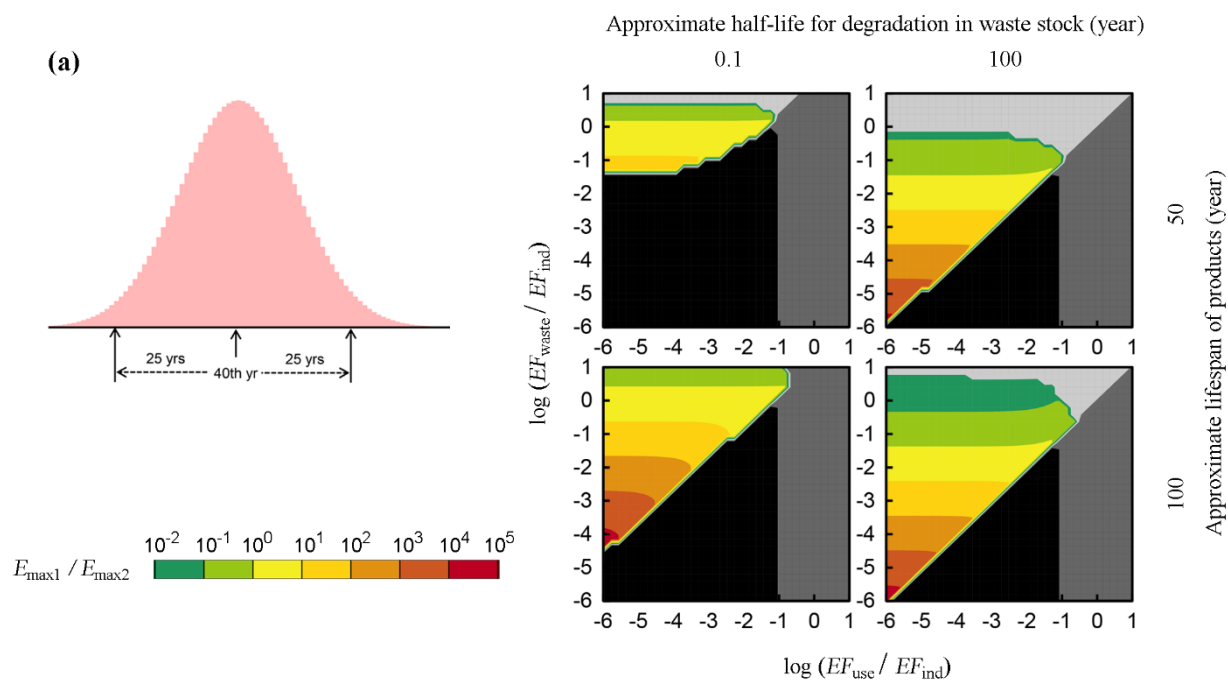
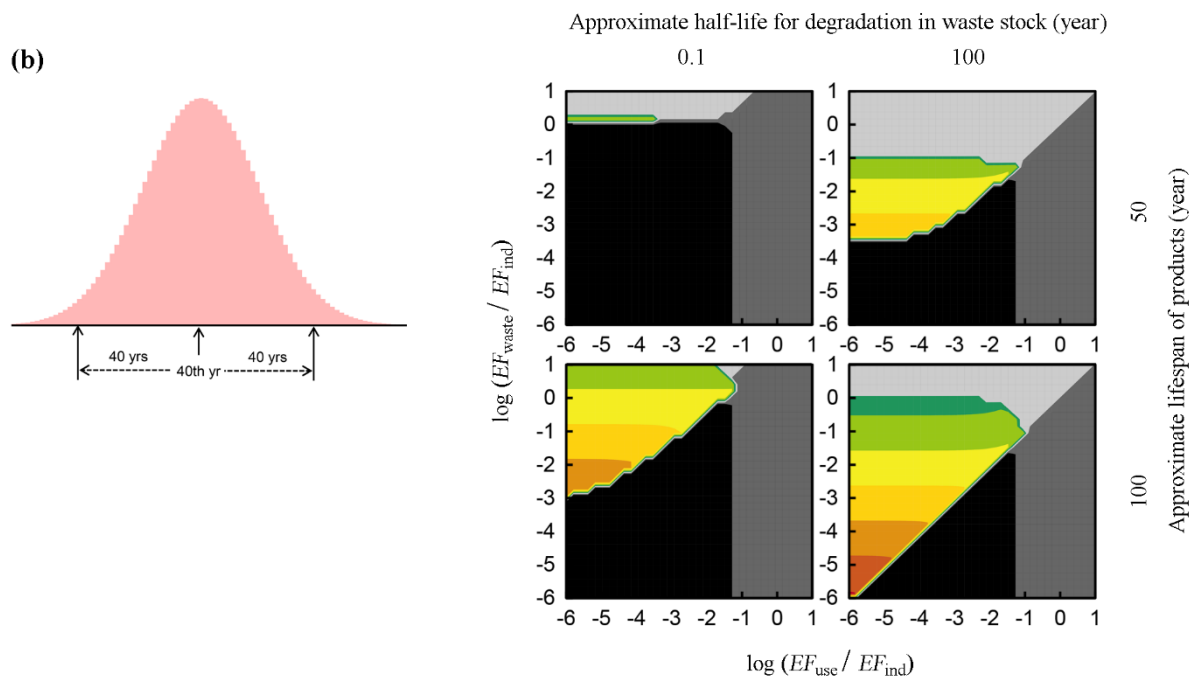


Figure S2 Shape of emission profile (doubled peaked indicated by colors vs. single-peaked indicated by greys and black) under different assumptions of the temporal trend of production

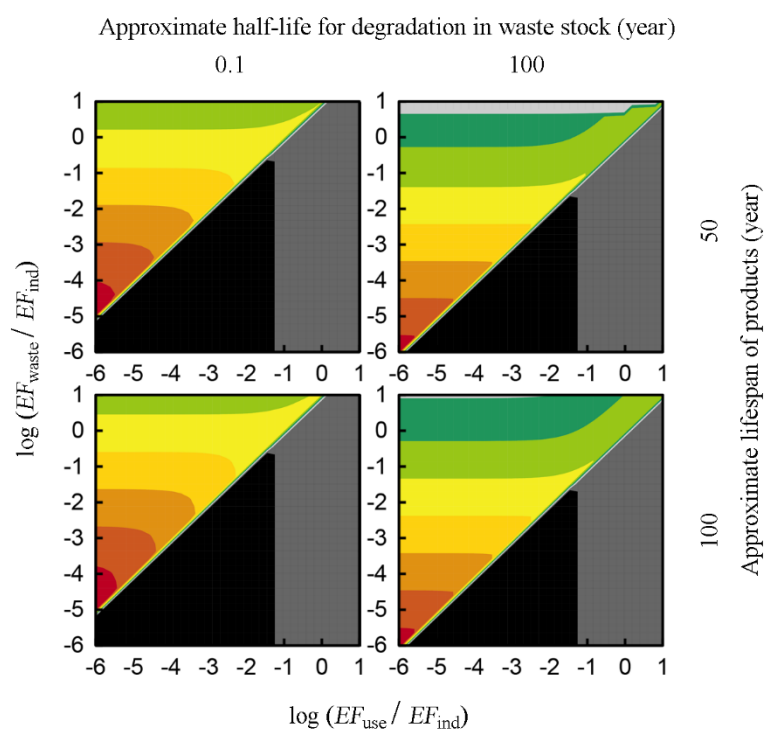
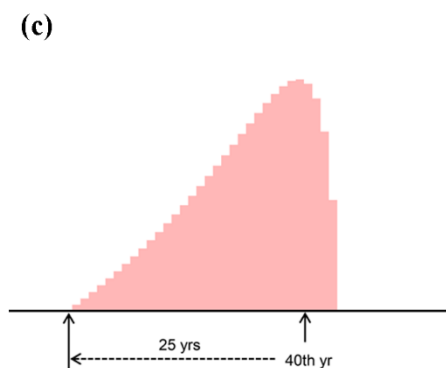
(a) Gaussian distribution with 95% of cumulative production occurring within 25 years before and after the peak year (i.e., the results presented in Figure 2 in the main text);



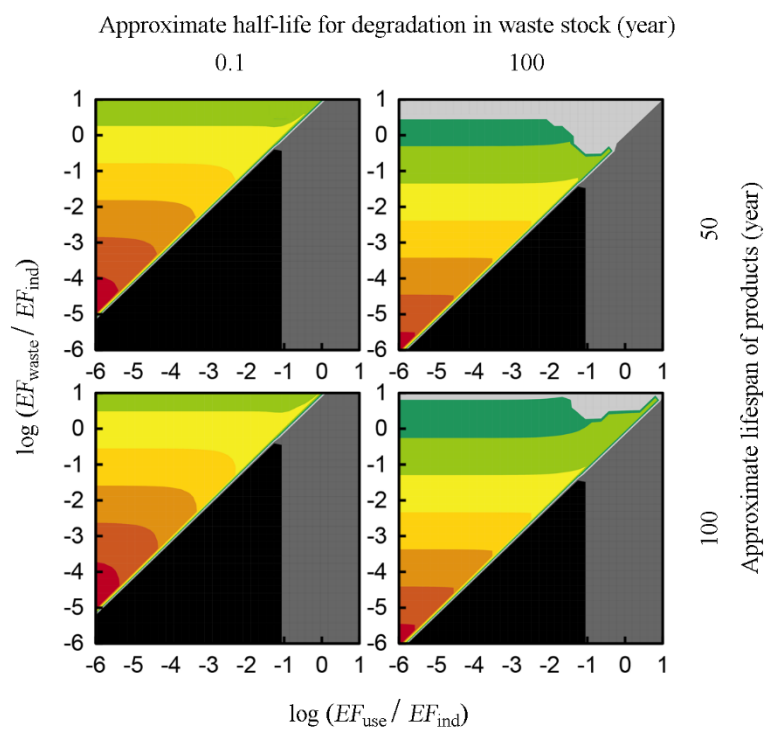
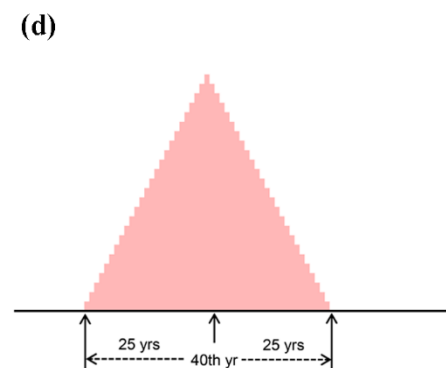
(b) Gaussian distribution with 95% of cumulative production occurring within 40 years before and after the peak year;



(c) Left-skewed distribution



(d) Triangular distribution



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