

Final report ECO53 assessment

A chemical categorization approach for Long-Range Transport Assessment

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Executive summary

The potential to undergo long-range environmental transport (LRTP) to remote regions is one out of four hazard criteria to be met for an organic chemical to be listed under the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP), and the Stockholm Convention on POPs (SC). However, the existing screening criteria / modelling approaches for estimating LRTP may lead to both false positives and false negative decisions with obvious implications for chemical management strategies. The initial project objective was to develop and evaluate a mechanistic and categorical approach to assessing LRTP that categorizes substances according to the major transfer processes from the atmosphere to surface media. The goal was not only to understand and describe the domain of applicability of simple models proposed for regulatory use, but also to identify future opportunities to expand their domain of applicability.

Major effort was invested towards analyzing to what extent current models and LRTP metrics reflect reality and meet anticipated regulatory needs. While not foreseen at the outset of the project, this aspect became the focus during the second half of the project as the research team was invited by the OECD to actively participate in a new Expert Group (EG) to update the OECD P_{ov} and LRTP Screening Tool (“The Tool”). This motivated the project to develop and analyze a set of three alternative and coherent LRTP metrics for potential implementation in an updated version of The Tool, referred to as the Emissions Fractions Approach (EFA). The new LRTP metrics were shown to have the potential to reduce the risk of false positive and false negative decisions in LRTP assessments (**Appendix B**) by accounting for the total dispersion of chemicals (air and water), reversible atmospheric deposition, and by allowing for a distinction between transfer versus accumulation in remote surface media. At the time of writing, it has not been decided by the OECD EG whether the EFA will be adopted and implemented in the new version of The Tool.

As The Tool ignores environmental variability, we have applied two higher-tier models which account for temporal and spatial variability to confront The Tool, and to assess the need for higher tier modelling approaches for more realistic LRTP assessment. The latter was exemplified by demonstrating how a decreasing spatial resolution could increasingly lead to an overestimation of LRTP into remote regions, and how the choice of spatial resolution affected model predictions of atmospheric deposition to the Baltic Sea (**Appendix A**).

The project team has made contributions in various regulatory contexts, notably the ECHA PBT Expert Group, CLRTAP, SC and, most extensively, the OECD. A new software allowing for calculations of the new LRTP metrics was made publicly available (The Level 3 EFA Model), with the hope that the availability of such a model will increase acceptance of the EFA and therefore augment the scientific and regulatory impact of the ECO53 project. Selected project results have been presented at SETAC Europe conferences and in two peer-reviewed scientific publications to date.

In addition to the two published papers in the appendices, this final project report summarizes different aspects of the work conducted toward the initial project objectives prior to the shift in focus to the development of the EFA metrics. More detailed information on the various research activities and project results are documented in meeting materials and minutes from the project review meetings, as well as in material presented at various meetings hosted by ECHA, CLRTAP and OECD. Throughout this report, we reference this material which is available on Sharepoint for further details on specific topics.

The report also includes a detailed bibliography of scientific papers and technical reports. The main topics that are covered in the bibliography include a general introduction to various long-range transport (LRT) modelling approaches, reviews of existing models, metrics and model intercomparisons, as well as monitoring data and monitoring programs selected based on their utility for LRTP assessments.

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Appendix A:

Breivik K., Eckhardt S., McLachlan M.S., Wania, F. 2021. Introducing a nested multimedia fate and transport model for organic contaminants (NEM). *Environmental Science: Processes & Impacts*, **23**, 1146-1157.

Appendix B:

Breivik, K., McLachlan, M.S., Wania, F. 2022. The Emissions Fractions Approach to Assessing the Long-Range Transport Potential of Organic Chemicals. *Environmental Science and Technology*, **56**, 11983-11990.

1. Modelling LRTP – key concepts and model features

A key motivation of the ECO53 project was to build trust in the ability of mechanistically based approaches to contribute to the assessment of long-range transport potential (LRTP) of chemicals, rather than relying on simple screening criteria such as atmospheric half-life and vapor pressure. These simple criteria do not differentiate substances according to the dominant mechanisms which dictate their LRTP and their potential for transfer to, and possible accumulation in, surface media of remote regions. Due to the variety and complexity of the processes involved, more complex models are required to assess LRTP. We therefore begin by introducing some of the key characteristics of different models and modelling approaches (Table 1) and discuss how these characteristics influence model applicability domain.

Multimedia models (MMs) and highly resolved atmospheric transport models (HRATMs). Simple MMs (or “box” models) have been extensively used to study the fate of chemicals which can distribute between two or more environmental compartments.^{4, 5} Assuming well-mixed environmental compartments (e.g. air, water and soil), they predict uniform concentrations in those compartments.^{6, 7} A notable example of a simple MM used for LRTP assessment is The OECD P_{ov} and LRTP Screening Tool (“The OECD Tool”) by Wegmann et al (2009).²

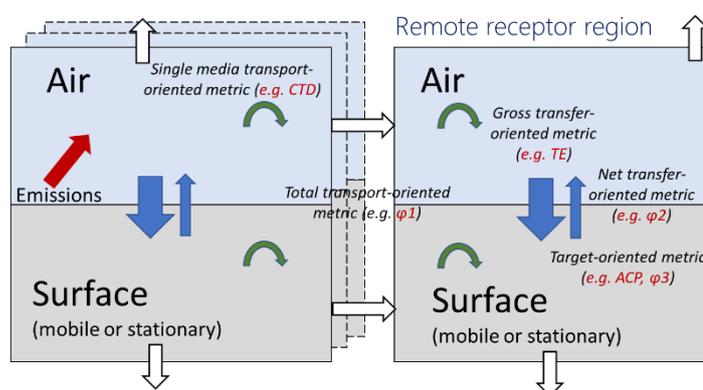


Figure 1: Simplified diagram of bulk processes which are typically included in LRTP models which include surface compartments. Red, green, blue and white arrows illustrate emissions, degradation, air-surface exchange and transport processes, respectively. Existing LRTP metrics included in The OECD Tool², including the alternative LRTP metrics developed during the ECO53, are indicated with abbreviations (see Appendix B).

In spite of their relative simplicity, The Tool and similar simple MMs are capable of integrating information on partitioning, transport and transformation to assemble a picture of the overall chemical fate in the environment following chemical emissions.^{4, 5, 7-9} Since the 1990s, MMs have gradually evolved to account for spatial and temporal variability (see also next paragraph). Unlike MMs, HRATMs are based on spatially and temporally variable atmospheric dynamics. Initially developed and used to study the dispersion of “classical” air pollutants, such as sulfur dioxide SO₂, HRATMs are now used for POPs on an operational basis under CLRTAP.¹⁰ We note that the distinction between what constitutes a highly resolved MM and what constitutes a HRATM is not always clear, as they have evolved to become more similar.¹¹ However, two different types of “classical” HRATMs can be discriminated. Lagrangian models follow the motion of parcels in air over time, whereas Eulerian models divide the environment into different grid cells (boxes). Spatially resolved MMs may therefore be classified as Eulerian models too. Not all HRATMs include compartments other than air,¹²⁻¹⁴ although several Eulerian ATMs have been expanded to include descriptions of chemical exchange with, and fate in, surface compartments.¹⁵⁻²⁰ The latter type of model is occasionally referred to as a Multicompartment Chemistry Transport Model (MCTM). Lagrangian and Eulerian HRATMs often differ from the highly resolved MMs as they have a higher spatial and temporal resolution. A feature of some Lagrangian HRATMs is that they can be operated in both forward and backward modes. This facilitates both predicting²¹ and interpreting long-range atmospheric transport events.²² In the ECO53 project, we have made extensive use of the Nested Exposure Model (NEM)²³, which may be classified as a highly resolved MM (**Appendix A**), as well as FLEXPART, which is a Lagrangian HRATM.^{13, 21, 22, 24}

Non-spatially resolved versus spatially resolved models. Non-spatially resolved MMs all describe the environment as a one-dimensional single box (1-D). Spatially resolved models may be classified as two- to three dimensional. An example of a 2-D model is SimpleBox, which is a nested MM describing regional, continental and global scales.^{25, 26} Globo-POP represents another example of a 2-D model, targeting zonal transport of chemicals on a global scale (Table 1). Some of the more recent MMs and all HRTMs are 3-D models, accounting for both vertical, zonal and meridional transport (Table 1). The spatial resolution varies considerably among LRT models, and some models may be operated with different spatial resolutions. For example, the BETR-Global model has versions with a resolution of either 15° x 15° (lat/long) or 3.75° x 3.75° (lat/long).²⁷ Choosing a non-spatially resolved model rather than a spatially resolved model may influence predictions of LRAT considerably.²⁸ Some nested models listed in Table 1, such as the Nested Exposure Model (NEM)²⁹ (**Appendix A**), can be operated with variable user-defined spatial resolutions, and thereby facilitate assessing the impact of spatial variability on LRAT.²⁹ In ECO53, we have used NEM to demonstrate how a low spatial resolution can lead to an overestimation of LRTP into remote regions (**Appendix A**). The spatial variability required for a realistic assessment of LRATP is chemical specific. A fine spatial resolution may be required for substances which are more rapidly removed from the atmosphere, either because of a relatively short reaction half-life in air and/or rapid atmospheric deposition. The latter applies to involatiles (“single-hoppers”) and superhydrophilics. Conversely, substances with a limited potential for deposition and long-half life in air (e.g., hexachlorobenzene, HCB) may not require a model with very high spatial resolution (persistent volatiles or “flyers”). The desirable spatial resolution of higher-tier LRATP models additionally depends on the emission scenario. This is particularly relevant for LRATP assessments involving highly “heterogeneous” emission scenarios, e.g., those dominated by one or a few point sources. The impact of variable spatial resolution on LRATP was presented and discussed at the **kick-off meeting**, using NEM to calculate various LRATP metrics for a diverse set of chemicals and emission scenarios.

Steady-state versus non-steady state. Environmental concentrations of chemicals in steady-state models (such as Mackay-type Level II to Level III MMs) do not change in time, whereas non-steady state models (Level IV models) account for dynamic (time-variant) conditions⁶. Most of the MMs used for LRTP assessment, including the OECD Tool, are steady-state Level III models. While it has been argued that (pseudo) steady-state conditions may prevail on shorter time-scales (e.g. if a non-persistent chemical is emitted at a constant rate), this is unlikely to be true over longer time-scales⁷. Steady-state models are not designed to predict the environmental response to changes in emissions over time, e.g., as a result of control measures aiming to reduce emissions. This intrinsic limitation is particularly relevant for long-lived chemicals residing in compartments which may respond slowly to changes in emissions, such as sediments and soils.³⁰⁻³³

Surface media. Inclusion of surface media in LRTP models is important when assessing the atmospheric transport of chemicals with a potential for reversible atmospheric deposition, and for estimating the potential for exposure (other than inhalation). The number of surface compartments included in MMs and MCTMs varies. For example, while the OECD Tool contains soil and water only, the BETR Global model additionally includes freshwater and vegetation.³⁴

Transport in media other than air. Several MMs, including The OECD Tool, also account for transport in water. This feature is particularly relevant when assessing LRTP of chemicals that partition into (low K_{AW} and low K_{OW}), and are persistent in, water. For such chemicals, referred to as superhydrophilics or “swimmers”, the half-life in air is inadequate as the sole criterion to assess LRTP, and models that include transport in water are required.³⁵

Generic vs geographically explicit multimedia models. Several early MMs were developed for the purpose of predicting the environmental fate of chemicals in a hypothetical, or generic environment. This feature has proven to be useful for comparing the relative LRATP behavior for multiple chemicals in screening exercises.^{2, 7, 11} Some non-spatially resolved evaluative models are parameterized to represent a specific region based on geo-referenced data (often a country, jurisdiction or continent) or, as the OECD Tool, the entire globe.² Most spatially resolved MMs and all HRTMs tend to be geo-referenced models (Table 1). The latter applies to NEM and FLEXPART used in ECO53.

Intermittent precipitation Early steady-state and non-steady state MMs all applied a constant, low rain rate, sometimes referred to as the “constant drizzle assumption”⁷. Hertwich³⁶ demonstrated that this assumption leads to the concentrations in air, and therefore LRAT, being underestimated for chemicals with a low K_{AW} (“superhydrophilics”). This potential for error was later demonstrated to also apply to chemicals which sorb to particles in air in appreciable amounts, i.e., chemicals with a high K_{OA} (“involatiles”).^{37, 38} These limitations were also discussed in studies by Lammel et al.³⁹ and Hansen et al.,⁴⁰ comparing model predictions by MMs and HRTMs. Many steady-state MMs have been updated to account for this process,²⁵ implementing the approximation proposed by Jolliet and Hauschild.^{38,34, 41, 42} During the ECO53 project, we implemented a description of intermittent precipitation in the Level III EFA model, which is otherwise identical to the Level III model within The Tool (**Appendix B**).

Table 1: Characteristics of selected LRTP models. The listed characteristics may not always be up-to-date as several models have been subject to development over several years.

Model name	Temporal resolution	Model domain	Spatial resolution	Intermittent precipitation	Type	Dimensions	Surface media	Key metrics ^D	Key references
OECD Tool	No	Generic (global)	No	No	MM	1-D	Yes	CTD, TE	2, 43, 44
EFA Model	No	Generic (global)	No	Yes	MM	1-D	Yes	EFA	(Appendix B)
CEMC LII/LIII	No	Regional	No	No ^{B)}	MM	1-D	Yes	CTD	6, 7
TaPL3	No	Generic	No	No	MM	1-D	Yes	CTD	45-49
ELPOS	No	Regional	No	No ^{B)}	MM	1-D	Yes	CTD	49, 50
CaITOX	No	Regional	No	No ^{B)}	MM	1-D	Yes	CTD	51-54
Impact 2002	No	W. Europe	2°x2.5°	No ^{B)}	MM	3-D	Yes	OR	55, 56
SimpleBox ^A	No ^{B)}	Nested	Yes	No ^{B,C)}	MM	1-D	Yes	OR	25, 57-59
ADEPT	No	Europe	Yes	No	HRTM	3-D	No	CTD _{east}	28, 60, 61
MPI-MBM ^A	Yes	Generic	No	No	MM	1-D	Yes		62
CoZMo-POP	Monthly	Regional	No	No	MM	1-D	Yes	CTD	37, 63, 64
ChemRange	No	Global	Yes	No ^{B)}	MM	1-D	Yes	SR	65, 66
CliMoChem	Yes	Global	Yes		MM	2-D	Yes	SR, cc	67
Globo-POP	Monthly	Global	Yes	No ^{C)}	MM	2-D	Yes	ACP	3, 41, 49, 64, 68-70
G-CIEMS	Monthly	Flexible	5km x 5km		MM	3-D	Yes	OR	71-74
BETR-N America ^A	Monthly	North America	Yes	No	MM	3-D	Yes	TE	75-77
EVn-BETR ^A	Monthly	Europe	5° x 5°	No	MM	3-D	Yes	CTD	40, 78-80
BETR-Global ^A	Monthly	Global	15° x 15°	Yes	MM	3-D	Yes	TE	34, 81-88
BETR-Global hr ^A	Monthly	Global	3.75° x 3.75°	Yes	MM	3-D	Yes		27, 82, 89
NEM	Monthly	Global (nested)	max 0.5° x 0.5°	Yes	MM	3-D	Yes		29 (Appendix A)
FATE	Yes	Global	2.5° x 2.5°		MM	3-D	Yes	GIF (S-R)	90
GEOS-Chem	Yes	Global	4° x 5°	Yes	HRTM	3-D	No ^{C)}	S-R	91-94
MPI-MCTM	Yes	Global	100 km x 250 km ⁹⁵	Yes	HRTM	3-D	Yes	PD / SS	16, 39, 96
MSCE-POP	Yes	N. Hemisphere (nested)	50 km x 50 km	Yes	HRTM	3-D	Yes	TD	15, 97, 98
GLEMOS	Yes	Global (nested)	0.1° x 0.1° ^{E)}	Yes	HRTM	3-D	Yes	S-R	10, 99
DEHM-POP	Yes	N. Hemisphere	Yes	Yes	HRTM	3-D	Yes	S-R	18, 40, 100, 101
GEM/POPs	Yes	Global	Variable (2°x2°)	Yes	HRTM	3-D	Yes		17, 102
FLEXPART	Yes	Global	Yes	Yes	HRTM	3-D	No	S-R	13, 21, 22, 103-108
HYSPLIT	Yes	Flexible	Yes	Yes	HRTM	3-D	No	S-R	12

^A May be operated in steady-state and non-steady state mode. ^B Version used in the model inter-comparison by Fenner et al.¹⁰⁹ ^C Accounted for in one or more recent versions of the model. ^D Some higher-tier models may be capable of predicting metrics introduced and/or explored using lower-tier models. ⁹⁰ See Table 2 for a description of individual metrics. ^E Finer resolutions are possible.

2. LRTP metrics

Several metrics have been developed to quantify a chemical's long-range transport potential (LRTP), e.g. ^{109, 110} (Table 2). It has become common to distinguish between *transport- and target focused metrics*. Transport-oriented metrics generally seek to quantify the extent of a compound's dispersal within a medium, e.g. in the atmosphere, whereas target-oriented metrics account for transfer of chemicals to a particular target, e.g. surface media in a specific geographic region.¹⁰⁹ For decision making contexts aiming to address the “*transfer to a receiving environment in locations distant from the sources of its release*”, transport-oriented metrics may have a limited stand-alone utility and target-focused metrics will be a more appropriate choice.³⁵

Several of the existing metrics suffer from limitations. For example, the CTD which is implemented as the transport-oriented metrics in The OECD Tool does not allow for coherent assessment of the long-range environmental transport in air and water combined. This restriction does not only have implications for the assessment of transport alone. It also restricts opportunities to carry out any assessment of transfer to, and accumulation in, remote surface media for chemicals which may undergo LRT via pathways other than air (**Appendix B**).

In the ECO53 project, we have developed a set of three coherent LRTP metrics which quantify the extent to which the chemical (i) reaches a remote region (dispersion, ϕ_1), (ii) is transferred to surface media in a remote region (transfer, ϕ_2), and (iii) accumulates in surface media in a remote region (accumulation, ϕ_3) (**Appendix B**). We have furthermore proposed that the term “target-oriented” should be restricted to metrics predicting delivery to a target region (e.g. TE, ϕ_{2A}), whereas metrics which additionally account for a chemical’s potential for accumulation in the target region should be referred to as accumulation-oriented (e.g. ACP, ϕ_3) (**Appendix B**).

Table 2: Existing LRTP metrics and related model endpoints.

Metric	Metric type	General description	Unit	Key references
Characteristic Travel Distance (CTD)	Transport	CTD represents the distance it takes to reduce the chemical concentration by 63% in a plug-flow system.	Distance	37, 111, 112
Spatial Range (SR)	Transport	The distance that contains 95% of the area under the curve of concentration as a function of place	Fraction (perimeter of earth)	65, 66
Outflow Ratio (OR)	Transport	A measure of export out of the (regional) model domain as a result of advection	Fraction	28
Transport Distance (TD)	Transport	Average distance from the source at which the mean annual concentration in air is reduced to 0.1%	Distance	28
Global average of imported fractions (GIF) ^A	Transport (other)	GIF is derived from source-receptor analyses and represents the global average of imported fractions in the atmospheric boundary layer of receptor regions weighted by the contents.	Fraction	90
Plume displacement (PD) and spatial spreading (SS)	Accumulation	Tendencies of substance distributions to migrate in two geographic directions	Fraction (total environmental burden)	96
Transfer Efficiency (TE)	Target (transfer)	TE represents the mass flux into a target compartment divided by the emission mass flux in a source region with the aim to quantify how much chemical reaches a remote region.	Percentage	2, 76
Spatial and temporal remote state (R^{TS})	Target (accumulation)	A conservative measure of the distribution of the chemical in the environment far from sources and long after emissions have ceased, independent of the mode of emissions. The maximum LRTP calculated for different modes of emissions has been suggested as a useful surrogate.	Fraction	35, 113, 114
-	Target (accumulation)	Fraction of amount released that is transported to the Arctic and remains there (time-integrated net flux into the Arctic divided by M_0)	Fraction	109
Arctic Contamination Potential (ACP)	Target (accumulation)	Potential for chemical enrichment in the Arctic, expressed as the fraction of the total amount in global surface media that is in the Arctic after a specified time period	Fraction	3, 115
Source-Receptor Analyses (S-R)	Transfer	Source-receptor analyses are calculated to assess so-called “blame matrices” under CLRTAP	Mass per time	10
The Emissions Fractions Approach	Transport, Transfer and Accumulation	A set of three new metrics which quantify the extent to which the chemical (i) reaches a remote region (dispersion, ϕ_1), (ii) is transferred to surface media in a remote region (transfer, ϕ_2), and (iii) accumulates in surface media in a remote region (accumulation, ϕ_3). (Appendix B)	Fraction of emissions	29

3. The chemical categorization approach

We developed a chemical categorization approach that discriminates between four categories of chemicals that differ fundamentally in their LRAT characteristics on the basis of the dominant physical process governing their removal from the atmosphere: (i) wet deposition of dissolved chemical; (ii) wet or dry deposition of aerosols; (iii) gaseous deposition; (iv) loss to the stratosphere. This categorization approach is most useful for understanding the LRAT of persistent organic molecules.

The chemical categories are determined by the partitioning properties of the chemical, namely the: (i) K_{AW} (air-water partitioning ratio), (ii) K_{OW} (octanol-water partitioning ratio), and (iii) K_{OA} (octanol-air partitioning ratio), each of which can be determined experimentally or estimated theoretically. Only two of these three partitioning ratios are independent as $K_{OW} = K_{OA} \times K_{AW}$ (or $\log K_{OW} = \log K_{OA} + \log K_{AW}$), if the effect of the mutual solubility of octanol and water on partitioning can be neglected. Model features can be plotted on chemical partitioning space plots (e.g., a plot of low K_{AW} versus $\log K_{OA}$) to illustrate what combinations of partitioning properties leads to that particular model feature. The scientific basis of this approach is well documented e.g. ^{1, 116, 117, 118}.

Figure 2 illustrates a chemical partitioning space plot of the chemical categorization approach. The plot was constructed from the dominant physical removal process as determined from chemical fate calculations with the Globo-POP model for chemicals with different combinations of K_{AW} and K_{OA} .¹¹⁵ Chemicals for which wet deposition of dissolved chemical (category (i)) was the dominant process are characterized by low K_{AW} and low K_{OA} and were labeled *Superhydrophilics*. Aerosol deposition (ii) dominated for chemicals with high K_{OA} , and these were labeled *Involatiles*. Chemicals for which loss to the stratosphere (iv) was the dominant process have high K_{AW} and high K_{OA} and were labeled *Volatiles*. Finally, chemicals for which gaseous deposition was dominant (iii) had partitioning

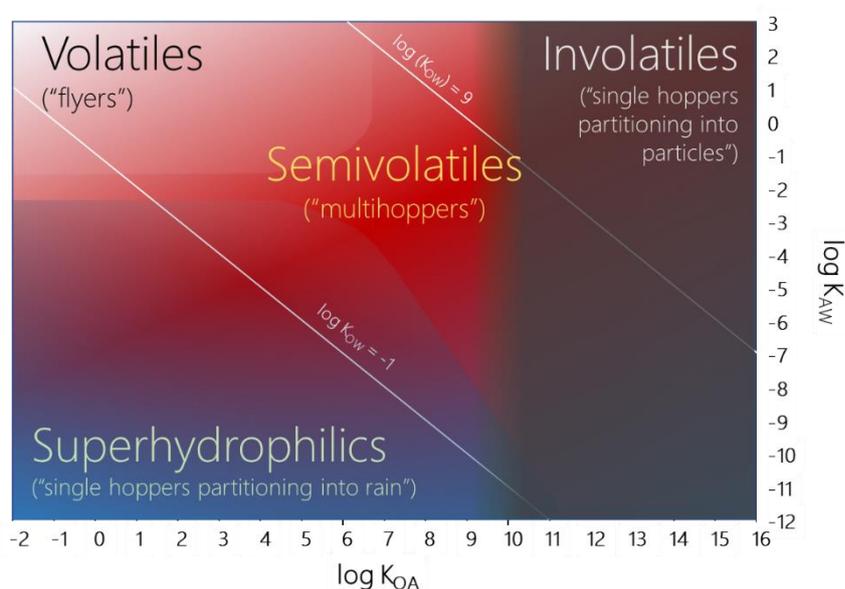


Figure 2: Chemical partitioning plot ¹ showing modes of environmental transport on a global scale for perfectly persistent chemicals modified from ³. The zones of transition depicted are illustrative and not definitive.

properties in between the other categories and were labeled *Semivolatiles*. Without the need to run any model simulations, it is possible to place chemicals based on their partitioning ratios in this plot to anticipate key aspects of their LRT behavior including temporal and spatial variability.^{119, 120}

Ignoring environmental variability could have implications for the assessment of LRTP as many models, including the OECD Tool, assume steady-state conditions

within a generic fixed environment and hence account for neither spatial nor temporal variability. The impact of accounting for environmental variability was explored using a series of model calculation

experiments in the ECO53 project, using NEM, FLEXPART and The OECD Tool. These results were presented and discussed during the 3rd project meeting.

4. Inter-comparison of models

Model inter-comparison studies provide important information to guide the selection of models for LRTP assessment. Some of the most comprehensive studies comparing different LRTP-assessment models e.g. ^{35, 39, 40, 49, 95, 109, 113, 121} have paid particular attention to assessments motivated by regulatory needs. There are the OECD/UNEP activities^{35, 109, 122, 123} which led to the development of The Tool² and there is CLRTAP,¹²¹ which relies on operational HRATMs at the EMEP centers in their work.^{28, 95, 124} Within ECO53, we have engaged in relevant activities in both the OECD/UNEP and CLRTAP regulatory contexts as summarized at the closing meeting. Whereas The Tool is a consensus-based MM which was designed by a group of experts to represent the state-of-science at the time, the HRATMs operated under CLRTAP (MSCE-POP and, more recently, GLEMOS) are maintained and refined by scientists at the EMEP Meteorological Synthesizing Centre – East in Moscow. The latter are not consensus-based models but are judged by independent international experts to represent the state-of-the-science and therefore to be fit for purpose for the activities under CLRTAP. The MSCE-POP model has been used to assess the LRTP of a wide range of candidate substances under CLRTAP.¹²⁵⁻¹³⁶ In these assessments, benzo[a]pyrene (B[a]P) and HCB serve as benchmarks to identify chemicals of regional and global concern, respectively.¹²¹

In the context of OECD/UNEP activities, Fenner et al.¹⁰⁹ compared estimates of different LRTP metrics using nine different multimedia models (Level II and Level III, CalTOX, ChemRange, ELPOS, SimpleBox, BETR North America, Globo-POP and Impact 2002) for a wide range of hypothetical chemicals. The ranges of partition ratios were $\log K_{AW}$ (-11 to 2) and $\log K_{OW}$ (-1 to 8), with $\log K_{OA}$ (calculated as $\log K_{OW} - \log K_{AW}$) spanning from -1 to 15. A number of generic half-life combinations in various compartments were furthermore explored (e.g., with reaction half-lives in air ranging from 4 hr to 1 yr), leading to a set of 3175 hypothetical chemicals. Whereas general agreement between transport-oriented LRTP metrics from different models for large parts of the chemical space was noted, differences were apparent for flyers in Globo-POP which, unlike the other models, calculated a target-oriented metric. Model results also varied across the chemical space according to whether certain fate processes were included or not (aquatic transport, degradability for the aerosol-bound fraction among involatiles, and the particle-bound settling to the deep sea for hydrophobic low-volatile substances). Overall, the model inter-comparison by Fenner et al.¹⁰⁹ clearly illustrated that the domain of applicability of models varies across the chemical space, i.e. for different chemical categories. A follow-up study by Klasmeier et al.³⁵ classified chemicals according to P_{OV} and LRTP characteristics, calculated using the same MMs as in Fenner et al.¹⁰⁹ (except the Level II model). Both studies informed the development of The OECD Tool, which was assessed to be representative of the state-of-the-art as its predictions were consistent with those of other steady-state MMs (BETR North America, CalTOX, CEMC Level III, ChemRange, ELPOS, Impact 2002 and SimpleBox) ².

Hollander et al.²⁸ published a summary of a major study, initiated by the Executive Body of the CLRTAP and coordinated by EMEP MSC-E,^{137, 138} comparing seven models with respect to rankings of predicted LRTP and overall persistence. This study included several MMs (CliMoChem, SimpleBox, EVn-BETR, G-CIEMS, The OECD Tool) and two HRATMs (MSCE-POP, ADEPT) and focused on 14 chemicals spanning a wide range of fate properties (aldrin, atrazine, benzo[a]pyrene, two polybrominated diphenyl ethers [BDE-47 and BDE-99], biphenyl, carbon tetrachloride (CCl₄), hexachlorobenzene (HCB), hexachlorobutadiene (HCBd), α -HCH, *p*-cresol and selected PCBs [PCB-28, PCB-153, PCB-180]).

Notable differences in rankings were found to reflect the choice of LRTP metrics. A similar finding was reported by Wegmann et al.² For example, large differences occurred for volatile chemicals (CCl₄ and HCB) with high LRTP rankings in models calculating various transport-oriented metrics and low scores in CliMoChem which uses an accumulation-oriented metric (Table 2). The situation was reversed for heavier PCBs as CliMoChem predicted a relatively high fraction being retained in the Arctic. Other reasons for differences in LRTP rankings were differences in spatial resolution (highly resolved models *versus* SimpleBox and the OECD Tool) and the inclusion of chemical loss to the deep ocean.

The EMEP study also included a more detailed model inter-comparison for PCB-153 for the period 1981-2000. This revealed a tendency for the more highly resolved models (MSCE-POP, EVn-BETR and G-CIEMS) to predict a higher LRATP than SimpleBox and CliMoChem. Lammel et al.³⁹ similarly found that MMs may overestimate atmospheric particle deposition and therefore underestimate atmospheric transport. Hollander et al. noted that substantial variation in atmospheric height between models (12,000 m in MSC-East vs. 1,000 m in SimpleBox), led to a higher export via air (i.e. LRATP) for models with the larger atmospheric heights.²⁸ The assumed atmospheric height in MMs had earlier been shown to influence the relative importance of atmospheric degradation and deposition.^{35, 49} This, along with the constant drizzle assumption, may explain why Hollander et al. found that the total deposition flux was predicted to be higher using MMs, compared to MSCE-POP.²⁸

Taken together, the model intercomparison studies show how the choice of model could have a significant impact on the outcome of any LRTP assessment.

5. Monitoring programs and databases

5.1 Major long-term regional air monitoring programs

Monitoring programs and databases provide information that can be used to evaluate models of LRT. The research and monitoring programs that measure POPs and related organic contaminants in air were reviewed during the **1st progress review meeting**. Many of the more recent efforts have been catalyzed by the SC.¹³⁹ Only a few long-term air monitoring networks (i) have operated for ~30 years and, (ii) are using active air sampling strategies. These are described below.

EMEP. The integrated approach of the EMEP program consists of three interconnected science-based pillars: emission estimation, atmospheric transport modeling and monitoring. The EMEP monitoring program is defined by a strategy.¹⁴⁰⁻¹⁴² The network of EMEP monitoring sites includes a range of components in a multi-pollutant, multi-effect approach.^{142, 143} POPs are mandatory observations in the EMEP program at so-called Level 2 sites. EMEP furthermore encourages submission of measurement data from more research-driven observations (Level 3 sites), e.g., data from measurement campaigns or observations of non-regulated organic contaminants of emerging interest. POPs in air and deposition have been reported to EMEP since 1999, but the EMEP database contains historical monitoring data as far back as 1991. During a workshop in 1997,¹⁴⁴ a set of selected POPs was recommended in a stepwise implementation approach, which was later included in the UNECE strategic long-term plans on POPs.¹⁴⁵ It was recommended to restrict measurements in the start-up phase to one 24 (48) hour high volume air sample per week, with particle filter and polyurethane foam (PUF) as sampling media¹⁴⁴ as detailed in the EMEP Manual for Sampling and Analysis.¹⁴⁶ The EMEP POP monitoring program collaborates with the marine (OSPAR, HELCOM) and

Arctic (AMAP) Conventions to develop harmonized sampling and quality assurance procedures as well as a shared data reporting, repository and dissemination system. EMEP sampling sites are defined as regional and should be representative of a large area. This classification is harmonized with the European Environment Agency (EEA).¹⁴⁷ The number of sites in the EMEP POPs network remains limited, with stations mainly located in Northern and Central Europe.¹⁴³ To meet the challenge of limited geographical coverage, passive air sampling campaigns supplement the long-term monitoring activities.¹⁰³

Table 3: Studies based on results from individual EMEP / AMAP long-term air monitoring sites using active air sampling techniques.

Site	Network(s)	Coordinates	Years	Chemicals
Alert (CA)	AMAP/NCP	82°30'N,62°20'W	1991 -	POPs ¹⁴⁸⁻¹⁵⁰ , PFAS ¹⁰⁴ , OCPS ¹⁵¹ , PAHs ¹⁵²
Andøya (N)	EMEP	69°16'N,16°00'E	2009 -	PFAS ¹⁰⁴
Aspvreten (S)	EMEP	58°48'N,17°23'E	1995 -	PCDD/Fs ¹⁵³
Birkenes (N)	EMEP	58°23'N,08°15'E	2003 -	POPs ¹⁵⁴ ,PCBs ^{13, 21}
Kosetice (CZ)	EMEP	49°34'N,15°04'E	1998 -	POPs ¹⁵⁴⁻¹⁵⁷ , PAHs ¹⁵²
Villum (DK)	AMAP	81°36'N,16°40'W	2009 -	POPs ^{148, 149, 158}
Pallas (FI)	AMAP/EMEP	68°00'N,24°15'E	1995 -	POPs ^{148, 149, 154, 159} , PAHs ¹⁵²
Råö (S)	EMEP	57°23'N,11°54'E	2001 -	POPs ^{154, 159}
Stórhöfði (IS)	AMAP/EMEP	63°24'N,20°17'E	1994 -	POPs ^{148, 149, 154}
Zeppelin (N)	AMAP/EMEP	78°54'N,11°53'E	1991 -	POPs ^{148, 149, 154} , PCBs ^{13, 107} , VMS ¹⁶⁰ , PFAS ¹⁰⁴

Measurements reported to EMEP are carried out by national laboratories, and quality assurance is performed on both the national level and by the EMEP Chemical Coordination Centre (CCC) to ensure satisfactory data quality and spatial and temporal comparability. In addition to reference methods and standard operation procedures,¹⁴⁶ EMEP conducts occasional laboratory intercomparisons in cooperation with other programs like AMAP and the Canadian Northern Contaminants Program (NCP).^{161, 162} Schlabach et al.¹⁶¹ concluded that participating laboratories had a satisfactory level of performance, with an uncertainty between 10% and 25% for most of the compounds. However, inter-laboratory differences of up to a factor of 2 have been reported when analyzing atmospheric SVOCs,¹⁶² Uncertainties in measurement data are therefore not merely dictated by analytical uncertainties alone.

The data checking routines in EMEP includes procedures for identifying outliers, comparison with other observations and application of knowledge on spatial and temporal variations. It is also recommended to compare measurements with model predictions and to use air mass trajectories to check if elevated concentrations are consistent with the sampled air mass having passed through known source areas. In addition, regularly station and laboratory audits at the national level are part of the applied accreditation systems.

In the ECO53 project, we did not only review the EMEP monitoring data, but we also used measurement data for PCB-153 at nine long-term monitoring sites to evaluate the ability of the NEM model to reproduce observations. The model evaluation showed that differences between observations and model predictions are comparable to the variability anticipated from differences in sampling and analytical methods applied across laboratories taking part in the EMEP POPs program (**Appendix A**). This study was highlighted as part of the **2nd progress review meeting**.

AMAP. The presence of POPs at unexpectedly high concentrations in the Arctic was recognized in the late 1970s.¹¹⁹ The Arctic Monitoring and Assessment Programme (AMAP) was established in 1991 to address various environmental issues of concern, including contamination with POPs.¹⁶³ Over the years, AMAP has issued a number of comprehensive assessment reports.¹⁶³ AMAP early on initiated the development of a coordinated air monitoring program across Arctic countries, including efforts to harmonize measurements across sites.¹⁴⁹ Some of the AMAP air monitoring stations (Alert on Ellesmere Island in Canada, Zeppelin at Svalbard – Norway, Pallas in Arctic Finland and Stórhöfði on Iceland) have been operational since ~1990. Additional sites have been reporting, or are more recently reporting, POPs air monitoring data to AMAP. Hung et al. have summarized long-term temporal trends of POPs in Arctic air in several reviews.^{148, 149} AMAP has recently also issued a report on the assessment of chemicals of emerging concern, which reviews data on the occurrence in air of a number of regulated, non-regulated and naturally occurring chemicals.¹⁶⁴

IADN. (Great Lakes Integrated Atmospheric Deposition Network) is a joint US-Canadian monitoring network which consists of five master stations, each located near one of the Laurentian Great Lakes.¹⁶⁵ Measurements at a few of the master stations started as early as 1990. An extensive number of scientific studies based on IADN has been published in the literature, including several reviews.¹⁶⁶⁻¹⁷⁴ Table 4 contains an overview of selected studies targeting specific chemical groups, although the total number of studies extends beyond those listed.

TOMPS. The UK TOMPS (Toxic Organic Micro-Pollutants) network consists of six urban and rural air monitoring stations measuring selected POPs (PCBs, PCDD/Fs, PBDEs and, more recently, HBCDD) using active air sampling. TOMPS is operated by Lancaster University and funded by the UK Department of Environment, Food and Rural Affairs (DEFRA). An overview of the main results from the TOMPS program over more than 20 was recently published by Graf et al..¹⁷⁵

Table 4: Major studies from long-term air monitoring networks including active air sampling of POPs.

Network	Number of stations	Years	Chemicals
AMAP (Arctic) ¹⁶³	8 sites (5 sites >10 years)	1990 -	POPs ^{148-150, 176} , PCBs ^{13, 107} , PFAS ¹⁰⁴ , OCPs ¹⁵¹ , PAHs ¹⁵²
EMEP (Europe) ^{177, A)}	38 sites (70% report PAHs only)	1990 -	POPs ¹⁴³ , PAHs ^{143, 178}
IADN (US-Canada) ¹⁶⁵	5 master stations plus satellite stations	1990 -	OCPs ¹⁷⁹⁻¹⁸⁶ , CUPs ^{187, 188} , PAHs ^{178, 189-191} , PCBs ¹⁹²⁻²⁰⁰ , PCDD/Fs ²⁰¹ , PBDEs ²⁰²⁻²⁰⁴ , Other HFRs ²⁰⁵⁻²¹⁶ , OPEs ^{217, 218}
TOMPS (UK) ²¹⁹	6 monitoring stations	1991 -	PCBs ²²⁰⁻²²² , PCDD/Fs ²²³⁻²²⁵ , PBDEs ²²⁶ , PAHs ^{227, 228}

A) Data from the EMEP program are summarized in annual data reports (see www.emep.int)

5.2 Major databases, data repositories and other large-scale studies

EBAS. The observations from the EMEP and AMAP programs are reported to the EBAS database infrastructure (<http://ebas.nilu.no/>) operated by the Norwegian Institute for Air Research (NILU). EBAS has been in operation since the 1970s and includes observations from many national and international monitoring programs and from several different research projects. The data center activities are closely linked to research and quality assurance activities in the networks. An important part of the data reporting is documenting methods and data quality along with the data to help the data users in interpreting the observations. In the reporting templates for POPs (<https://ebas->

submit.nilu.no) important metadata about field method (i.e., the filter setup) and laboratory procedures (i.e., absorbent used and sample preparation) are included. In addition, several flags are to be used to indicate problems with the instrument or procedure, or possible local influence from various sources.

GMP. The Global Monitoring Plan (GMP) supports the implementation and effectiveness evaluation of the SC by collecting comparable, harmonized and reliable information on POPs in core environmental matrices, including air.²²⁹ EMEP, AMAP as well as several other monitoring programs and networks contribute to the GMP. A Global Guidance document has been developed for Parties to the SC to implement the GMP.²³⁰ It is recommended to have a network of a few active high-volume sampling and several passive sampling stations to obtain sufficient geographical coverage. Several QA/QC procedures are defined to ensure comparability between methods.²³⁰ Observations from contributing networks are collected in the POPs GMP Data Warehouse (GMP DWH, <https://www.pops-gmp.org/>).

Passive air sampling. A number of passive air samplers (PAS) have been developed over the last 25 years e.g.^{231, 232}. PASs have in common that they measure chemicals that had been taken up from the atmosphere by a sorbent without the aid of a pump. PASs have found wide-spread use for semi-volatiles, including many POPs. Several studies have used PASs to sample in both source regions and at background sites.²³³ Studies that may be particularly relevant in the context of LRAT are listed in Table 5, and include PAS studies of urban-rural gradients,²³⁴ and studies on a national,²³⁵ regional,²³⁶⁻²³⁹ continental,^{48, 240-244} or global scale.^{245, 246} We refer to Wania and Shunthirasingham for a comprehensive overview.²³³

Table 5: Major large-scale and/or long-term monitoring efforts using passive air samplers, targeting or including measurements in background air (>10 sites).

Geographical region	Year(s)	Type of sampler ¹	Number of sites	Chemicals ²	References
UK-Norway transect	1994 - present	SPMDs / SIPs	11	PCBs, PBDEs, PAHs, OCPs	236, 238, 247-250
North America (5 countries)	2000-2001	XAD	40	PCBs, OCPs, PBDEs	48, 242, 251
Europe (22 countries)	2002	PUF	71	PCBs, OCPs, PBDEs, PAHs, PCNs	240, 252
Europe (19 countries)	2002	POG	38	PCBs, OCPs, PBDEs, PAHs, PCNs	244
Great Lakes (USA/Canada)	2002-2003	PUF	15	PCBs, OCPs, PBDEs, PCNs	253-255
Czech Republic	2003-2015	PUF	14	PCBs, OCPs, PAHs	256
Asia (China, Japan, S. Korea, Singapore)	2004	PUF	77	PCBs, OCPs, PBDEs	241
Global (GAPS)	2004/2005	PUF	41	PCBs, OCPs, PBDEs, PCNs, FRs	245, 246, 257
China	2005	PUF	97	PCBs, DP	258, 259
Global (GAPS)	2005 (seasonal)	PUF	41	PCBs, OCPs, PBDEs	260
Global (GAPS)	2005-2006	XAD	39	PAHs	261
Global (GAPS)	2005-2008	XAD	34-46 each year	OCPs, CUPs	262
Europe / EMEP (34 countries)	2006	PUF	86	PCBs, OCPs, PAHs	103
Central / Eastern Europe (22 countries)	2006-2008	PUF	155	PCBs, OCPs, PAHs	239
Global (GAPS)	2006-2011	XAD	17-46 each year	nPFAS	263
East Asia (China, Japan and S. Korea)	2008	PUF	45	PBDEs	264
East Asia (5 countries)	2008	PUF	106	PCBs, PCNs	265
Africa (15 countries)	2008	PUF	26	PCBs, OCPs, PAHs, PCDD/Fs	243, 266
Asia (China, India, Japan)	2009	SIPs	46	PFCs	267
GAPS	2009	SIPs	20	PCBs, PFCs, PCBs, PBDEs, OCPs, CUPs, VMS	268-270
GAPS	2009/2013/2015	SIPs	21	PFAS, VMS	271
Asia (5 countries)	2012/2013	PUF	176	PAHs	272
Global (GAPS)	2014	PUF	48	FRs	273
Australia	2016	XAD	15	SCCPs and MCCPs	274

- 1) Semipermeable membrane devices (SPMDs) e.g. ²³⁸, polyurethane foam disks (PUF) e.g. ²³¹, Polymer-coated glass (POG) e.g. ²⁷⁵, XAD-2 e.g. ²³², Sorbent-impregnated PUFs (SIPs) e.g. ²⁷⁶
- 2) Polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs), polybrominated diphenyl ethers (PBDEs), currently used pesticides (CUPs), polychlorinated naphthalenes (PCNs) flame retardants (FRs), Dechlorane Plus (DP), neutral per- and polyfluoroalkyl substances (nPFAS), dioxins and furans (PCDD/Fs), polyfluoroalkyl substances (PFCs – neutral and ionic), volatile methyl siloxanes (VMS), short- and medium-chain chlorinated paraffins (SCCPs / MCCPs).

6. Closing remarks and outlook

Partway through the project two conditions converged to change its course: we had the inspiration for novel and improved LRTP metrics, and we were invited to participate in the OECD process to update the OECD P_{ov} and LRTP Screening Tool (“The Tool”). This resulted in the development of the Exposure Fraction Approach (EFA) to LRTP assessment, which is the major achievement of this project. EFA provides three distinct yet coherent measures of LRTP, elegantly integrates LRAT and LRWT, and reduces the risk of false negative and false positive outcomes. We have published a paper on the EFA approach in a leading journal (**Appendix B**), launched publicly accessible software to calculate the EFA metrics, and advocated for the inclusion of EFA in “The Tool” within the OECD working group. This represents a major advance in the ability to assess the LRTP of organic chemicals.

Before embarking on the development of EFA, we achieved many of the objectives of the initial project plan, as documented in this report. This included characterizing the features of different models and metrics used for LRTP assessment and analyzing their strengths and limitations. The chemical categorization approach proved to be a valuable framework for this work. In addition, we documented the monitoring programs and data sources that could be of use in evaluating models used for assessment of LRTP.

However, pursuing the EFA came at the expense of not completing key elements of the planned research. One such element was exploring discrepancies between what is needed for sound LRTP assessments and what is available in terms of scientific knowledge, and identifying strategies for how these discrepancies could be overcome. Particularly large discrepancies are already apparent for involatile chemicals. At present, The Tool and similar simple models are not capable of discriminating between the LRATP of different involatile chemicals; all are classified identically. To overcome these discrepancies, existing knowledge of atmospheric reactions on/in aerosols and of particle deposition as a function of emission source needs to be synthesized and assessed. The second major research element that we could not pursue in full detail was an assessment of the spatial and seasonal variability in LRTP, which we intended to pursue using the chemical categorization framework while employing the modeling tools selected in the first part of the project. We hope that future research will address these questions, as we believe that they will lead to further improvement in LRTP assessment.

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Introducing a nested multimedia fate and transport model for organic contaminants (NEM)[†]

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Some organic contaminants, including the persistent organic pollutants (POPs), have achieved global distribution through long range atmospheric transport (LRAT). Regulatory efforts, monitoring programs and modelling studies address the LRAT of POPs on national, continental (e.g. Europe) and/or global scales. Whereas national and continental-scale models require estimates of the input of globally dispersed chemicals from outside of the model domain, existing global-scale models either have relatively coarse spatial resolution or are so computationally demanding that it limits their usefulness. Here we introduce the Nested Exposure Model (NEM), which is a multimedia fate and transport model that is global in scale yet can achieve high spatial resolution of a user-defined target region without huge computational demands. Evaluating NEM by comparing model predictions for PCB-153 in air with measurements at nine long-term monitoring sites of the European Monitoring and Evaluation Programme (EMEP) reveals that nested simulations at a resolution of $1^\circ \times 1^\circ$ yield results within a factor of 1.5 of observations at sites in northern Europe. At this resolution, the model attributes more than 90% of the atmospheric burden within any of the grid cells containing an EMEP site to advective atmospheric transport from elsewhere. Deteriorating model performance with decreasing resolution ($15^\circ \times 15^\circ$, $5^\circ \times 5^\circ$ and $1^\circ \times 1^\circ$), manifested by overestimation of concentrations across most of northern Europe by more than a factor of 3, illustrates the effect of numerical diffusion. Finally, we apply the model to demonstrate how the choice of spatial resolution affect predictions of atmospheric deposition to the Baltic Sea. While we envisage that NEM may be used for a wide range of applications in the future, further evaluation will be required to delineate the boundaries of applicability towards chemicals with divergent fate properties as well as in environmental media other than air.

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Environmental significance

Within a regulatory context, the model-based assessment of the potential of organic chemical for long range atmospheric transport (LRAT) is often accomplished with extremely simple models lacking spatial resolution. The computational efficiency of the nested global modelling approach introduced here should enable future model applications of regulatory and scientific interest that previously were not possible with highly spatially resolved models. This includes (i) the performance of stochastic sensitivity and uncertainty analyses, (ii) the screening of large numbers of organic chemicals for LRAT, (iii) the modelling of complex contaminant mixtures comprised of numerous constituents with divergent LRAT potential, and (iv) model investigations requiring the simulation of large numbers of scenarios.

Introduction

Persistent Organic Pollutants (POPs) have been banned and restricted globally because of possible harm to environment

and human health.^{1,2} The justification for their regulation on a global scale is their ability to undergo long-range environmental transport (LRT),^{1,3,4} which has been established through numerous measurements at remote background sites^{5,6} on national,⁷⁻⁹ regional,^{10,11} continental¹²⁻¹⁵ and global scales.¹⁶ Field measurements across geographical scales have been paralleled by regulatory efforts across national boundaries and jurisdictions. At the same time a wide variety of mathematical models has been developed to predict the environmental dispersal of POPs, because a comprehensive understanding of their long-range atmospheric transport (LRAT) and the consequences for sound control strategies cannot be achieved on the basis of measurements alone.

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Because POPs reach remote areas mainly by LRAT, networks have been set up to measure their concentrations in air in support of national, regional and global policy objectives.¹⁷ The most comprehensive network in Europe is the European Monitoring and Evaluation Programme (EMEP),¹⁸ which collates POP measurements carried out by national laboratories as part of national programs. Similar arrangements apply to other regional long-term monitoring programs, such as the Arctic Monitoring and Assessment Programme (AMAP),¹⁹ and the joint US-Canadian Great Lakes Integrated Atmospheric Deposition Network (IADN).²⁰ These programs have generated long-term time trends of POPs in the atmosphere based on data reaching back to the early 1990s. On a global scale, the Global Monitoring Plan (GMP) supports the implementation and effectiveness evaluation of the Stockholm Convention on POPs^{4,21} by synthesizing data on ambient concentrations in air from national, regional and global air monitoring programs.^{16,22}

Monitoring data do not only inform regulatory efforts but are also essential for model evaluations. In the context of LRAT, the utility of multimedia fate and transport models (MFTMs) targeting national scales^{23,24} is limited. Continental-scale MFTMs such as Impact 2002 (ref. 25) or the Berkley-Trent models for North America^{26,27} and Europe²⁸ suffer from difficulties in accurately defining the advective import of POPs from outside the model domain.²⁹ This is not an issue for global MFTMs which range from relatively simple evaluative models *e.g.*³⁰ to dynamic geo-referenced three-dimensional models.³¹ Whereas MFTMs rely on simplified descriptions of environmental transport, such as meteorological conditions averaged over the time scale of a month, global high resolution transport models (HRTMs) account for the spatially and temporally highly dynamic nature of atmospheric mixing and removal processes. HRTMs are, for example, capable of realistically predicting the short-term variability in concentrations in air of remote regions,^{32–34} such as the occurrence of LRAT episodes causing temporary concentration peaks.³⁵ At the same time, HRTMs tend to be computationally too demanding to simulate a wide range of POPs over the decadal time scales reflective of their environmental persistence.

Major challenges therefore still remain in terms of formulating models which have global coverage, high spatial resolution, and the capability of performing a large number of simulations (*e.g.* for multiple chemicals and scenarios or for sensitivity analyses). In other words, LRAT models which operate across scales to serve multiple needs are largely missing. We propose to address this need through a flexible, nested modelling approach which is (i) less computationally demanding, enabling numerous simulations at coarse resolution, (ii) able to individually target, yet also integrate regional, continental and global scales, (iii) offers higher spatial resolution of receptor regions of scientific and/or regulatory interest, and (iv) dynamic, allowing one to address the environmental response to changes in emissions.

The main objectives of this study are to (i) introduce a new Nested Exposure Model (NEM) for organic contaminants that is designed to meet those requirements, (ii) evaluate different geometries of the model by comparing predictions of variable

resolution with each other and with measurements, (iii) apply the model to quantify the relative importance of local sources and advective atmospheric import from elsewhere to the concentrations observed in areas in which European background monitoring stations are located, and (iv) explore how the choice of spatial resolution affect predictions of atmospheric deposition to a specific receiving environment. PCB-153 is used as an example.

Materials and methods

Points of departure

NEM builds upon CoZMo-POP2 (ref. 36) and BETR-Global,³⁷ two existing dynamic MFTMs for predicting the long-term behavior of persistent semi-volatile organic contaminants in the physical environment. Whereas CoZMo-POP2 is not spatially resolved,³⁶ the resolution of the global-scale BETR-Global was originally $15^\circ \times 15^\circ$ (lat/long),³¹ but recently increased to 3.75° .³⁸ As all resolutions used here are identical in latitude and longitude we used X° when referring to $X^\circ \times X^\circ$ in the remainder of the paper. NEM adopts major parts of the code from CoZMo-POP2 and supplements it with parts of the parameterization of BETR-Global.³⁸ An advantage of this approach is that these models have undergone extensive evaluations over the years lending credibility to the underlying process descriptions built into NEM. CoZMo-POP2 has been evaluated for PCBs³⁹ and short-chain chlorinated paraffins.⁴⁰ Similarly, BETR-Global has been previously evaluated and applied.^{37,38} By relying on previously documented models and process descriptions, we can restrict the model documentation to those elements of NEM that are novel and make it different from CoZMo-POP2 and BETR-Global.

Nesting

A key feature of NEM is the opportunity to operate the model across different spatial scales and resolutions to focus on a specific region of the globe. By nesting up to five different model domains with user-defined geometries and resolutions, NEM offers increasing resolution with increasing proximity to a given target region of interest. Through sequential calculations, model output obtained from simulating a larger domain with coarse resolution serves to define the boundary conditions for a smaller domain with finer resolution, nested within the larger domain. The nested domain needs to be fully incorporated within the larger domain and rely on boundary conditions derived from one preceding domain alone. This nested approach not only reduces computational demands by limiting simulations at the finest resolution to a user-defined region of interest, but it also allows for the assessment of the impact of spatial resolution on model predictions, all within a consistent modelling framework. The BETR-Global model relies on spatially variable environmental input data.⁴¹ Most of these data sets have been replaced by new global databases as described and summarized below.

Transport

Chemical transport between adjacent grid cells within NEM may occur by air, fresh water and seawater, and is defined on

the basis of data with a monthly temporal resolution and a 0.5° spatial resolution (Fig. S1†). NEM linearly interpolates monthly into daily values. Monthly means of fresh water discharge for the years 2000–2009 are taken from the global freshwater model WaterGAP 2.2c.⁴² Freshwater flow from a grid cell may occur in eight directions (N, NE, E, SE, S, SW, W, NW) and vertically (inland sinks). Inland sinks are treated as an input of chemical to terrestrial surface media. Outflow cells for run-off into the sea are based on Döll *et al.*⁴³ Ocean circulation is based on the third version of the Simple Ocean Data Assimilation (SODA3) ocean reanalysis⁴⁴ for which the horizontal velocities in four directions (N, E, S, W) within the ocean mixing layer and the vertical velocity out of the mixing layer were retrieved (SODA 3.4.2, 30th May 2019). The parameterization of atmospheric transport is re-gridded from BETR-Research.³⁸ The finest resolution in NEM is constrained by the resolution of freshwater transport.

Compartmentalization

NEM includes compartments describing a two-layer atmosphere (representing the boundary layer and the free troposphere),³⁷ forest canopies (needleleaf and broadleaf), soils (forest soils, agricultural soils and uncultivated soils), permanent snow/ice, fresh water and freshwater sediment, as well as seawater and marine sediment (Fig. S1†). NEM additionally includes a seasonal sea ice cover, parameterized on basis of the SODA3 ocean reanalysis. Area fractions for terrestrial compartments are based on the land cover classification from IGBP at 0.5° resolution.⁴⁵ Some minor adjustments were necessary to align the IGBP database with the requirements of WaterGAP and SODA3. A marine sediment compartment is assumed to be present in all coastal grid cells with run-off into the sea, and for marine grid cells with a seawater depth less than 600 meters. NEM is also integrated with the ACC-Human bioaccumulation model.⁴⁶

Other environmental input parameters

Long-term monthly means of temperature in air at 2 meters above the surface, wind speed at 10 m and daily precipitation at 0.5° resolution were taken from ECWMF (European Centre for Medium-Range Weather Forecasts – www.ecwmf.int). The durations of dry and wet periods were also calculated with data from ECWMF. Soil organic carbon content at 0.5° resolution was derived from global soil maps.^{47,48} Monthly seawater temperatures and bathymetry were taken from SODA3.⁴⁴ The remaining environmental input parameters were adopted from CoZMoPOP2 (ref. 36) or BETR-Global.³⁸

Physical-chemical properties and emissions

For this initial study, we selected PCB-153 (2,2',4,4',5,5'-hexachlorobiphenyl) because (i) it is representative of the larger group of non-polar organic contaminants (ii) environmental and human exposure to PCBs remains high and toxicologically relevant^{49,50} (iii) empirical data critical for model evaluation are plentiful, and (iv) a global historical emission inventory for PCBs is available.^{51,52} Partition coefficients at 25°C (octanol-water K_{OW} , octanol-air K_{OA} and air-water K_{AW}) and energies of

phase transfer for adjusting those coefficients to temperatures other than 25°C were adopted from Li *et al.*⁵³ The rate constant for the gas-phase reaction of PCB-153 with OH radicals at 25°C , along with the corresponding activation energy, were taken from Brubaker and Hites.⁵⁴ For reaction rates and activation energies in media other than air, we used the values by Wania and Su.⁵⁵ The most recent global historical emission inventory for PCB-153 was used to predict environmental concentrations (default scenario).⁵⁶ Reflecting the observed seasonal variability at a monitoring station influenced by local sources,⁵⁷ emissions are assumed to vary seasonally following a sine function with the warmest month in each grid cell having emissions 50% above the annual average.

Model scenarios exploring impact of spatial resolution outside a nested European domain

It is well established that numerical diffusion in Eulerian box models with coarse resolution leads to overestimated concentrations in remote areas and underestimated concentrations in source regions.^{58,59} Global HRTMs employ a fine spatial resolution to overcome this problem, but a fine spatial resolution may not necessarily be required on a global scale when the area of interest is a single confined region. Nevertheless, a coarse spatial resolution outside of a nested region could have an appreciable “edge effect” on model predictions within that region. The maximum number of grid cells in a single simulation in NEM is restricted to ~ 3000 . The number of grid cells is constrained by the available memory within the software used. This limits the finest spatial resolution for a global simulation to 5° . To explore this edge-effect, we performed three global simulations at resolutions of 30° , 15° and 10° , each of which was followed by the simulation of a nested European domain (90°N , 30°W to 30°N , 30°E) at 5° (Fig. S2A†). We also included an additional simulation for the European domain at 5° , in which inflow of PCB-153 was set to zero (Fig. S2B†). The results obtained from a global simulation at 5° then served as the benchmark to which results from the four nested simulations were compared (Fig. S2C†).

Model scenarios exploring impact of spatial resolution within a northern European domain

For model evaluation and application, we studied the impact of spatial resolution within northern Europe (85°N , 30°W to 45°N , 30°E) using three global simulations: non-nested with 15° resolution throughout; non-nested with 5° resolution throughout; and nested with 1° resolution in the nested northern European domain and 5° outside of it (Fig. 1 and S3†).

In all cases, simulations covered the years from 1930 to 2020 at time-steps of 6 hours ($30^\circ/15^\circ$), 3 hours (10°), 1 hour (5°) and 15 minutes (1°) with results stored six times per year.

Monitoring data

The nested domain was chosen because of the relatively large number of long-term monitoring stations in northern Europe. We used monthly mean concentrations in background air from AMAP^{19,60} and EMEP^{18,61} (ebas.nilu.no) to evaluate the predicted

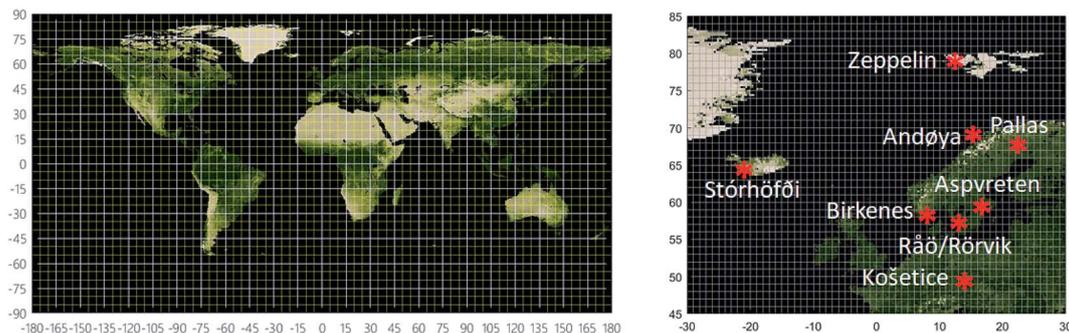


Fig. 1 The map on the left shows the global NEM model domains at the 15° and 5° resolutions. The map on the right shows the location of the air monitoring sites within the nested northern European domain at the 1° resolution.

spatial and temporal trends of PCB-153. The list of stations, station coordinates and years covered is included in Table S1.† The model evaluation is restricted to air because the focus of the model application is on LRAT.

Visualizations

The Normalized Difference Vegetation Index (NDVI) image generated by NASA based on MODIS satellite observations was used for mapping results.

Results and discussion

Spatial resolution outside a nested domain

The advective chemical inflow *via* air, fresh water and seawater into a nested domain will depend on the emissions outside the nested domain as well as the spatial resolution selected for this external (to the nest) domain. Fig. S4† displays maps of the ratio of PCB-153 concentrations in air obtained for late summer 2015 from each of the nested simulations and those from the global benchmark simulation. If the global simulation was done at 30°, the predicted concentrations in the nested domain were higher than the benchmark simulation by factor of 1.96–1.26 over the entire year. When the global simulation was done at 15° and 10° resolution, that factor was reduced to 1.46–1.05 and 1.09–0.93, respectively. While predictions thus were still higher than in the global benchmark simulation, the increased spatial resolution outside the nested domain reduced the effect of numerical diffusion into the nested domain. Concentrations predicted with the scenario without advective inflow were always lower than in the benchmark simulation, although the difference relative to the benchmark simulation was minor (as low as 2%) in the major source region in central parts of Europe (*e.g.* UK, Germany and France)^{51,52} (Fig. S3†).

Larger deviations were predicted for individual grid cells within the nested domain. The maximum ratios over the year were particularly large (up to 15.8) when relying on the coarsest resolution (30°) and declined with finer resolution (8.7 at 15°; 3.1 at 10°). The tendency to overpredict concentrations relative to the benchmark simulation was largest at the highest latitudes (Fig. S4†). For example, when the nested simulation relied on the finest outside resolution (10°), the average ratio over the

year for the 5° grid cell in which the Zeppelin monitoring station on Svalbard is located was 1.23 (1.00–1.80).

There were also circumstances where predictions based on the global benchmark simulation exceeded predictions from any of the nested simulations, as seen over ocean areas near the western border of the domain (Fig. S4†). However, these minimum ratios were very similar for all of the nested simulations (0.36 at 30°, 0.33 at 15°, 0.36 at 10°). Such results may be anticipated when the global benchmark simulation resolves confined plumes extending outwards from source regions towards the boundaries of the nested domain (see also Fig. 4C). The ability to perform quantitative evaluations within a consistent modelling framework illustrates that this feature can be explored to inform tiered modelling strategies. While inflow into a nested domain from global sources will always be over-estimated because of numerical diffusion outside the boundaries, the initial analysis shows that any “edge effect” as seen at the highest latitudes (Fig. S4†) can be greatly reduced by increasing the spatial resolution of the preceding domain (*e.g.* by further nesting).

Model evaluation

Numerical diffusion can be further minimized by reducing the spatial resolution within the nested domain. Fig. 2 compares predicted and observed concentration time trends of PCB-153 in air at the nine monitoring sites with the longest time series at a spatial resolution down to 1° (Table S1,† results for four further sites with shorter time series are found in Fig. S5†). The extent of model-measurement agreement is further evaluated by comparing geometric means of observed and predicted concentrations for the period for which measurement data were available for each site (Fig. 3A). The geometric mean concentrations varied over more than an order of magnitude between sites, from 3.7 pg m⁻³ at Košetice to 0.1 pg m⁻³ at Andøya, and this range is largely captured by the model (Fig. 3A). The three model simulations predict a decline in concentrations at all sites (Fig. 2), consistent with the decline in primary emissions in the model domain over the time-period for which monitoring data are available. The predictions are broadly in line with temporal trends observed at most, but not all, of the sites. For example, a review of long-term temporal trends in Arctic air did

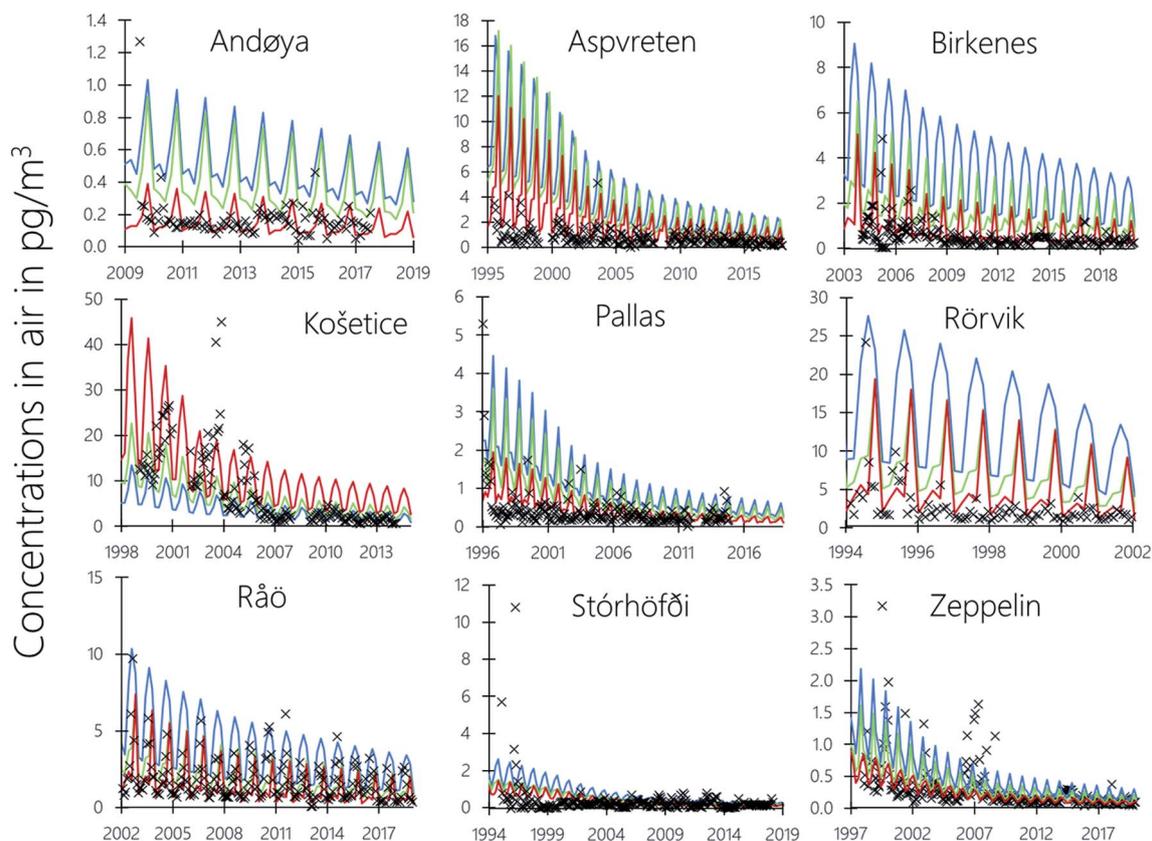


Fig. 2 Observed long-term trends of PCB-153 in air (black markers) and predictions with NEM at 15° (blue), 5° (green) and 1° (red) spatial resolution.

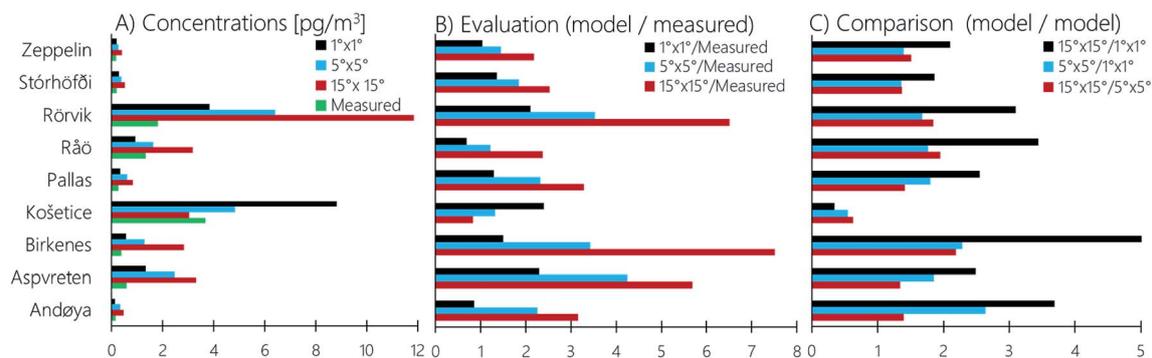


Fig. 3 Observed and predicted geometric mean concentrations (A), ratios between model predictions and measured concentrations (B) as well as ratios across model predictions (C) simulated using different resolutions. The predicted mean concentration is calculated from data for the grid cell in which the station is located for those years for which observed data are reported.

not identify any downward trend in concentrations of PCB-153 at Stórhöfði,¹⁹ but NEM predicts such a decline.

The extent of agreement between predicted and observed temporal trends varies between sites (Fig. 2 and 3). This could be because sampling frequency and duration vary between sites. At the Swedish and Finnish stations weekly samples are collected throughout the year and analyzed in monthly pools,⁶² which yields concentrations more directly comparable to the results of a model that relies on variables with a temporal

resolution of a month. At Košetice, Stórhöfði, and the Norwegian sites air sampling is intermittent. For example, samples at Andøya were collected over 48 hours once per month in 2018. Such intermittently measured concentrations are subject to short-term fluctuations that cannot be captured by NEM. Some variability in model-measurement agreement between sites could also arise because they were generated in different laboratories. Differences of up to a factor two are plausible when trace amounts of PCBs are quantified in air samples.⁶³

The extent of agreement between predicted and observed concentrations generally improved with increasing spatial resolution (Fig. 2 and 3). The predicted geometric mean concentration divided by the corresponding measured concentration at 15°, 5° and 1° across all nine sites was, on average, 3.8 (range: 0.8–7.5), 2.4 (range: 1.2–4.3) and 1.5 (range: 0.7–2.4), respectively (Fig. 3B). That these average ratios are larger than 1 implies that the model tended to overestimate observed concentrations of PCB-153 in air, irrespective of the spatial resolution. The tendency for better agreement with increasing spatial resolution was consistent for 7 out of 9 sites, with the notable exception of Košetice for reasons discussed below. There was a slightly better agreement for Råö at intermediate resolution compared to the finest resolution. Overall, the model evaluation shows that differences between observations when averaged over a month and model predictions at higher resolution are comparable to the variability anticipated from differences in sampling and analytical methods used across laboratories involved in the EMEP POPs programme.

Comparison of model results at different resolution

NEM allows for a direct comparison of predictions obtained at different spatial resolution. Predictions at coarse resolution (15°) were 2–5 times higher than those at the finest resolution (1°) at all sites except Košetice (Fig. 3C). As the comparison of air concentrations at different resolution in Fig. 3C is restricted to grid cells containing monitoring stations, Fig. 4 displays maps with concentrations predicted with NEM, exemplified for late summer 2015, with three different resolutions as well as a map of the ratio between predictions obtained at the coarsest and the finest resolution. Similar plots for other points in time during 2015 are included in the ESI (Fig. S6–S8†). The three first panels in Fig. 4 highlight the effect of numerical diffusion on the predicted spatial dispersion of PCB-153. Concentrations in the northwestern part of the domain are clearly elevated at coarse resolution, whereas at fine resolution higher concentrations are predicted in central Europe in the southeastern part of the model domain, comprising the major PCB source regions (Fig. S3†).

The maps in Fig. 4 suggest that a coarse resolution overpredicts atmospheric dispersion and thereby overestimates concentrations in air across much of the domain. Concentrations predicted at 15° resolution were, on average, 3.4 times higher than those at 1° (Fig. 4), with the factor varying from 2.2 to 3.4 over the year (Fig. S9†). The difference between predictions obtained at 5° *versus* 1° was less pronounced, varying by a factor between 1.5 and 1.8 during 2015 (Fig. S10†).

In sharp contrast to most of the model domain, concentrations predicted for the southeastern PCB source regions at the 1° resolution were much higher than predictions at 15°. This is also a result of overestimated atmospheric dispersion from major source regions at coarse resolution. Košetice is the only sampling site that falls into that part of the model domain, which explains why it is also the only one where an increasing spatial resolution does not improve model-measurement agreement (Fig. 3B). The apparent better model-measurement

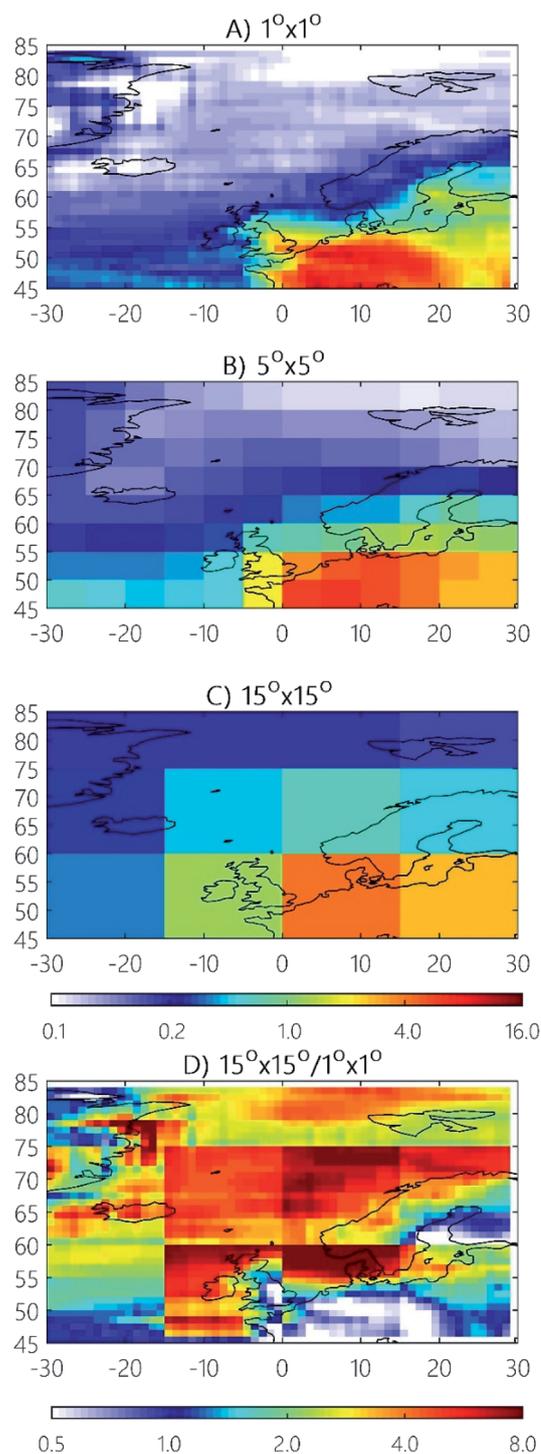


Fig. 4 Predicted concentrations of PCB-153 in air (pg m^{-3}) during late summer 2015 using NEM with different spatial resolutions (upper three panels). The lower panel shows the ratio of the concentrations predicted at a resolution of 15° and 1°.

agreement for Košetice at coarse resolution (Fig. 2 and 3B) may suggest that emissions affecting the 1° grid cells containing Košetice are overestimated. We attribute this to uncertainties in the emissions inventory which spatially distributes national emissions using population densities. While this might be a fair

approximation for models operating at a coarse resolution, it becomes increasingly unrealistic at finer spatial scales, *e.g.* because facilities treating PCB-containing waste are not necessarily located in the most densely populated areas of a nation. This suggests that the ability of the model to resolve spatial variability in concentrations in or close to PCB source regions is constrained by the accuracy of the emission inventory at finer resolution.⁵² Furthermore, an accurate description of atmospheric dispersion becomes increasingly important when a station is located close to an area with strong emissions.

Fig. 4 also helps to explain the tendency towards better agreement with increasing spatial resolution, especially if the emissions inventory at 1° reveals a high spatial variability in emission strength within a 15° cell (Fig. S3†). A notable example is the 15° grid cell containing Birkenes in southern Norway (Fig. 1), for which predictions exceeded measurements by almost a factor of 8 (Fig. 2 and 3). This is because the same 15° grid cell also comprises major source regions in Central Europe. Simulations at higher resolution result in stronger modeled concentration gradients within that 15° cell (Fig. 4, lower panel). The map in Fig. 4 (lower panel) helps to identify regions where increasing spatial resolution is expected to be particularly important. These are regions with steep concentration gradients. Reducing model resolution reduces concentration gradients by two mechanisms: averaging out gradients within a cell, and flattening out gradients between cells (Fig. S9 and S10†).

Applying NEM to compare inflow *versus* in-cell emissions

It is of interest to assess whether atmospheric burdens within a certain region are mainly controlled by atmospheric inflow from the outside or primarily dictated by emissions within the region. The ratio of these two flows is a measure of the remoteness of the location from sources, assessed on the scale of the grid cell. Whether this grid cell is remote or not will therefore depend on the resolution of the model. How to choose suitable measurement sites has been a topic within EMEP for many decades, dating back to assessments of LRAT for acidifying pollutants.^{18,64} EMEP aims to sample at background sites where the air and precipitation quality parameters are representative of a larger region.^{18,65} Since an important objective of monitoring in EMEP is also to evaluate atmospheric transport models, the size of the area a site is expected to represent has historically been constrained by the spatial resolution of early EMEP models. Compared to air pollutants that are more easily measured, monitoring sites for POPs are very limited in number (Fig. 1).¹⁸ We therefore applied NEM to assess how representative a site is in terms of remoteness within the area (grid cell) in which they are located.

We calculated the fraction of the total PCB-153 input to a grid cell that is originating from outside this cell, *i.e.* is not attributed to local emissions within that cell. We caution that this measure assumes that any molecule which were initially emitted in a grid cell never returns. This calculation was done for the grid cells containing air monitoring sites and for different spatial resolution (Fig. 5). If a site is located within a cell without emissions, atmospheric inflow is the only source

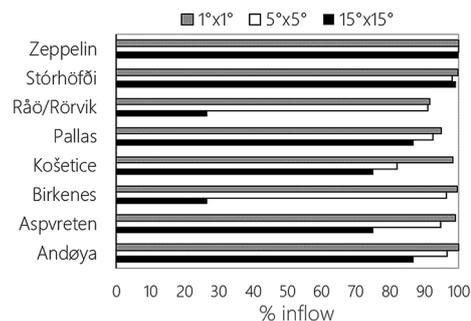


Fig. 5 Relative significance of atmospheric inflow to the sum of atmospheric inflow plus emissions for grid cells in which a monitoring station is located, simulated using three different spatial resolutions, 1930–2020.

of PCB-153. This is the case for Zeppelin, irrespective of the spatial resolution, as well as for Andøya at 1°. For most sites and resolutions, inflow from the outside dominates, making a contribution of more than 75% (Fig. 5). The notable exception was the 15° grid cell containing Birkenes and Råö/Rörvik, where only ~27% of the PCB-153 is estimated to originate from outside. This occurs because this cell comprises areas with high emissions in central Europe as well as parts with comparatively low emissions, including the regions where Birkenes and Råö/Rörvik are located (Fig. 4 and S3†). This leads to a high estimated contribution from local emissions at coarse resolution, which is not representative of the actual situation at those sites. At increasing resolution, these and many other sites become increasingly influenced by atmospheric inflow. At 1°, atmospheric inflow is predicted to contribute close to 100% of PCB-153 to the cell containing Birkenes, and ~92% to that containing Råö/Rörvik. The general pattern of decreasing influence of local emissions with increasing resolution indicates that the sites are generally remote within the larger region in which they are located. The exception is Stórhöfði where local sources make the highest contribution at a resolution of 5°. This is because at that resolution Stórhöfði shares a cell with Reykjavik (61°14'N, 21°94'E), where most of the Icelandic PCB emissions are assumed to occur, but at a 1° resolution they are in separate cells. At a resolution of 15° (Fig. 4), Icelandic emissions become negligible when distributed within a very large grid cell. A similar situation to Stórhöfði is evident for High Muffles (coastal site in N. England) and Westerland (west coast of Germany, close to the Danish border) which both are seen as remote at a resolution of 1°, but not 5° (Fig. S5†).

A numerical threshold is required to define a grid cell as remote or not. If an (arbitrary) cut-off of less than 95% is chosen, then the cells containing Stórhöfði and Zeppelin will be defined as remote at all model resolutions. The cells containing Andøya and Birkenes are remote at 5°, Aspvreten and Košetice at 1°, while Pallas falls just below this threshold at 1° (94.9%). The cell containing Råö/Rörvik which always includes Göteborg, the second largest city in Sweden, is not defined as remote at any spatial resolution (91.5 at 1°). To what extent a grid cell containing a specific background monitoring station is remote

therefore depends on the model resolution, while a threshold is required to identify whether the cell is remote or not. If the site is close to source regions, the desirable spatial resolution is likely to be higher and will depend on the grid cell's source proximity.

The simple approach used herein, calculating emissions *vs.* inflow to assess whether and to what extent a site monitors LRAT, resembles attempts to establish measures of remoteness using generic emission scenarios *e.g.*³⁸ If realistic emission scenarios are available, NEM can help to plan where to collect air samples.^{14,66} Fig. S11† shows results across the model domain at 15°, 5° and 1°. Applying the cut-off at 95% for model predictions at 15°, 7 out of 16 grid cells will qualify as remote. The two adjacent grid cells which include the UK (60°N, 15°W to 45°N, 0°E) and Germany/France (60°N, 15°W to 45°N, 0°E) are predicted to be predominantly controlled by emissions within these grid cells. The percentage inflows are predicted to be 41.9% and 26.6% respectively. Råø/Rörvik and Birkenes fall into the latter grid cell (Fig. 5). At a resolution of 5°, 34 grid cells qualify as remote. Two out of the 108 grid cells in total are predicted to be mainly controlled by emissions, rather than inflow. These two cells cover central parts of England (55°N, 5°W to 50°N, 5°W) and central parts of France (50°N, 0°W to 45°N, 5°E) with predicted inflows of 33.2% and 48.3%, respectively. At a resolution of 1°, emissions occur in 666 out of 2400 grid cells in total whereby 463 grid cells classify as remote. Only a single grid cell (London) is predicted to be predominantly influenced by local emissions (48.8% inflow) at 1° resolution. The results presented in Fig. S11† summarize how numerical diffusion both leads to overestimated atmospheric dispersion from major source regions and LRAT into remote northern sites.

Applying NEM to resolve spatial gradients in atmospheric deposition to the Baltic Sea

The occurrence of PCBs in the Baltic Sea has been extensively studied for more than half a century, *e.g.*^{67,68} Previous studies have highlighted the role of atmospheric deposition and air–sea exchange in controlling the levels and fate of PCBs in the Baltic Sea. *e.g.*^{69–71} A review of temporal trends of PCBs in biota over ~30 years across the Baltic Sea suggested that airborne transport has been the key factor controlling contaminant burdens in the Baltic Sea, with local and regional sources and LRAT all having played a role.⁷² It follows that accurate quantification of atmospheric input is crucial for understanding the fate of PCBs in the Baltic Sea. Because this body of water extends from PCB source regions in the Southwest to more remote regions in the North, predictions of atmospheric inputs are likely to depend strongly on spatial resolution. We have therefore compared the atmospheric deposition of PCB-153 to the Baltic Sea in 2015, when calculated with NEM at resolutions of 1°, 5° and 15° (Fig. S12–S14†). The atmospheric deposition predicted at 15° generally exceeded predictions at the finest resolution by a factor of about ~2 (1.5–2.2) for the Baltic Sea as a whole during most of the year except for the late fall (0.7) (Table S2†). Differences are much more pronounced for individual sub-

basins, with discrepancies in estimated deposition for the Bothnian Sea (defined as the Baltic Sea >60°N) during winter, and the Kattegat/Skagerrak region (<15°E) during summer, occasionally exceeding one order of magnitude for individual grid cells (Fig. S15†). This example highlights the significance of numerical diffusion for compartments other than air, and the need for fine resolution when targeting specific receiving environments of interest.

Conclusions

The NEM model allows for both targeting and integrating regional, continental and global scales through nesting. The effect of numerical diffusion has been explored by varying the spatial resolution selected outside of a nested European region simulated at 5°. This “edge effect” was found to be most pronounced for grid cells at the boundaries of the highest latitude, but it was greatly reduced by increasing the spatial resolution outside the nested domain from 30° to 10°. The evaluation for the nested northern European model domain, simulated at 1°, demonstrated a very good agreement with concentrations of PCB-153 in air measured at nine long-term monitoring sites. While there was a slight tendency to overestimate observed concentrations in air, the deviations between predictions are comparable to the variability expected from differences in sampling and analytical methods alone. The model predicted that ~92% or more of the atmospheric burden within the grid cells containing an EMEP site can be attributed to advective atmospheric transport from outside those cells at a resolution of 1°. However, the ability of the model to resolve spatial variability in concentrations within or adjacent to PCB source regions becomes increasingly constrained by the accuracy of the emission inventory at finer resolution.

An advantage of NEM is the ability to flexibly select spatial resolutions and nesting strategies in a way that matches the resolution of the emission inventory, rather than operating HRTMs on the basis of emissions data that are more coarsely resolved than the model. The latter approach may lead to an overparameterization with respect to the available emission inventory. For some countries and regions more highly spatially resolved emission inventories^{73,74} may be available than what is available at coarser scales (*e.g.* globally). Under such circumstances, NEM allows for a harmonized nesting strategy which matches the model resolution to the available emission inventories at various scales.

An obvious advantage of a nested modelling strategy is that its computational efficiency allows for simulations of large number of substances (*e.g.* complex mixtures or groups of chemicals). Multimedia models have been shown to be suitable for simulating the fate of complex compound mixtures based on defining input parameters for blocks of closely related substances, *e.g.* hydrocarbon mixtures in gasoline^{75,76} or different formula groups of the short-chain chlorinated paraffins.⁴⁰ However, while previous efforts of this type were restricted to relatively simple non-spatially resolved models, NEM opens up the possibility to simulate spatial and temporal aspects of the fate of contaminant mixtures, *e.g.* by investigating

how mixture composition will change in space and time. This is feasible because a nested approach allows for spatially resolved, dynamic simulations of dozens of mixture components with reasonable computational resources. It should also be feasible to screen large numbers of chemicals for LRAT with a NEM-based approach, when spatial variability in LRAT is an important aspect of the assessment.^{77–79}

The scope of testing and applying the model in this study was restricted to PCB-153 in air within northern Europe. There clearly is a need for future efforts to test the model for other regions, contaminants and compartments to help identify areas for further improvements as well as to characterize the domain of applicability. However, the flexible model geometries and resolutions make us optimistic that NEM will prove useful in future studies of scientific and/or regulatory interest. The choice of resolution will depend on the purpose and requirements of a model simulation as illustrated for the Baltic Sea. Ideally, a simulation using NEM should be done at the finest resolution achievable with the available computing resources (*i.e.* model and CPU time). As those resources will always be restricted, NEM offers a rationale for targeting most of the limited resources towards those aspects of a model simulation which are of greatest interest.

Conflicts of interest

There are no conflicts to declare.

Author contributions

Knut Breivik: conceptualization, data curation, formal analysis, funding acquisition, investigation, methodology, project administration, resources, software, validation, visualization, writing – original draft. Sabine Eckhardt: conceptualization, data curation, formal analysis, investigation, methodology, software, visualization, writing – review & editing. Michael S. McLachlan: conceptualization, funding acquisition, supervision, methodology, writing – review & editing. Frank Wania: conceptualization, funding acquisition, supervision, methodology, writing – review & editing.

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The Emissions Fractions Approach to Assessing the Long-Range Transport Potential of Organic Chemicals

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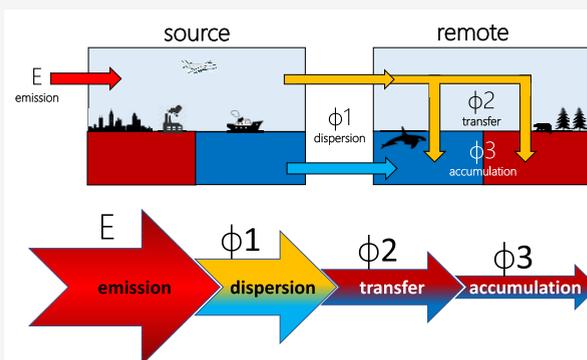
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Supporting Information

ABSTRACT: The assessment of long-range transport potential (LRTP) is enshrined in several frameworks for chemical regulation such as the Stockholm Convention. Screening for LRTP is commonly done with the OECD Pov and LRTP Screening Tool employing two metrics, characteristic travel distance (CTD) and transfer efficiency (TE). Here we introduce a set of three alternative metrics and implement them in the Tool's model. Each metric is expressed as a fraction of the emissions in a source region. The three metrics quantify the extent to which the chemical (i) reaches a remote region (dispersion, ϕ_1), (ii) is transferred to surface media in the remote region (transfer, ϕ_2), and (iii) accumulates in these surface media (accumulation, ϕ_3). In contrast to CTD and TE, the emissions fractions metrics can integrate transport via water and air, enabling comprehensive LRTP assessment. Furthermore, since there is a coherent relationship between the three metrics, the new approach provides quantitative mechanistic insight into different phenomena determining LRTP. Finally, the accumulation metric, ϕ_3 , allows assessment of LRTP in the context of the Stockholm Convention, where the ability of a chemical to elicit adverse effects in surface media is decisive. We conclude that the emission fractions approach has the potential to reduce the risk of false positives/negatives in LRTP assessments.

KEYWORDS: Stockholm Convention, long-range environmental transport, hazard, metrics, multimedia model, screening



1. INTRODUCTION

Concern related to long range atmospheric transport (LRAT) of pollution dates back 50 years¹ and has led to international agreements such as the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP)^{2,3} and the Stockholm Convention (SC) on Persistent Organic Pollutants (POPs).⁴ Over time, the regulatory interest has evolved toward a broader view of long-range transport (LRT), with long-range transport via water (LRWT) becoming an important consideration. The potential to undergo long-range transport (LRTP) to remote regions is a key hazard criterion to be met for an organic chemical to be listed under CLRTAP and the SC.⁴ In the SC, the requirement for listing is “the chemical is likely as a result of its long-range environmental transport to lead to significant adverse human health and/or environmental effects”, i.e., the chemical must not only be transported to remote regions, it must also accumulate in surface media there to an extent sufficient to cause harm.

Mathematical models play an important role in the scientific support of regulatory efforts, and a number of model-derived metrics has been developed for LRTP assessment,^{5–10} whereby transport- and target-oriented metrics are distinguished. The former address the potential of a chemical for widespread dispersal in air and/or water.⁵ Examples include the character-

istic travel distance (CTD),^{11–13} the spatial range (SR),^{14,15} and the outflow ratio (OR).⁷ Because adverse effects of POPs arise from dietary uptake and transfer in food webs and rarely, if ever, from respiratory exposure, the SC considers the “transfer to a receiving environment in locations distant from the sources of its release” an integral part of LRT.¹⁶ Examples of target-focused metrics that seek to explicitly account for transfer of chemicals to surface media include the transfer efficiency (TE in %)^{17,18} and the Arctic contamination potential (ACP).¹⁹

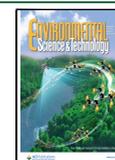
A wide range of models with different levels of sophistication have been developed over the years to calculate these LRTP metrics.^{5,7} Following initiatives by the Organisation for Economic Co-operation and Development (OECD) and the United Nations Environment Programme (UNEP),²⁰ an expert group was established in 2001 to provide guidance on how to use multimedia models in assessments of LRTP and

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overall persistence (P_{OV}).²¹ A consensus model for LRTP and P_{OV} assessments, the OECD Overall Persistence and Long-Range Transport Potential Screening Tool (“the Tool”) was developed to support decision making for chemical management.¹⁷ The Tool calculates the CTD and the TE and it has found wide use in scientific research and regulatory practice, e.g., refs 22–25. For these calculations it employs a multimedia model which was deliberately designed to be as simple as possible, e.g., with respect to the number of compartments and the use of the steady-state assumption.

Transport- and target-oriented metrics are clearly related: only chemicals dispersed widely can be transferred to, and accumulate in, remote surface media. Curiously, no attempt has so far been made to establish metrics where that relationship is made explicit or quantified. Similarly, although the possibility for dispersal in air and water, and even the interaction of the two dispersal paths,^{5,7} is generally acknowledged, separate metrics for LRT in air and water are usually defined and rarely integrated. For example, there is no apparent way to combine CTDs in air and water to characterize overall LRT, and the TE, as implemented in the Tool, does not allow for the possibility that a chemical is transferred to the remote environment in water. Clearly, there is room to improve on the existing metrics for LRTP assessment.

The objectives of this study were to develop and introduce a coherent and integrated mechanistic approach to LRTP assessment that builds on a set of new transport- and target-oriented metrics that overcomes many of the limitations of the existing approaches. While these metrics should be intuitive and have an easily grasped meaning, one of them should explicitly assess a chemical’s potential for LRT in the sense of the SC. The point of departure was the desire to express quantitatively the relative extent to which a chemical can (i) reach remote regions, (ii) be transferred to surface media in remote regions, and (iii) accumulate in surface media in remote regions. Importantly, the relevant metrics should not be separate entities, but relate to, and complement, each other in a mechanistically meaningful manner. While the new metrics can be obtained with a variety of models of different levels of sophistication, a guiding principle during the design of the new approach was the need to have it implementable in very simple, steady-state models. Therefore, we introduce it here while relying on the fate model and parametrization in the OECD Tool.

2. MATERIALS AND METHODS

2.1. OECD Tool Model Environment. The Tool, which has been described in detail by Wegmann et al.,¹⁷ is a steady-state multimedia mass balance model, classified as a level III fugacity model.^{26–28} Its three compartments are parametrized to represent the global environment: the troposphere, the soil surface layer, and the seawater surface layer (Figure 1). Intermedia transport occurs by diffusion and advection, and degradation can occur within each compartment. Bulk degradation is assumed for water and soil, whereas degradation in air is restricted to the gas phase. While equilibrium is assumed within each compartment, chemicals need not be in equilibrium between them.¹⁷ Consequently, the predicted chemical distribution within the model environment and the LRTP metrics depend on the mode of emissions. The Tool makes predictions for three individual emission scenarios, which are 100% emissions to air, water, or soil.

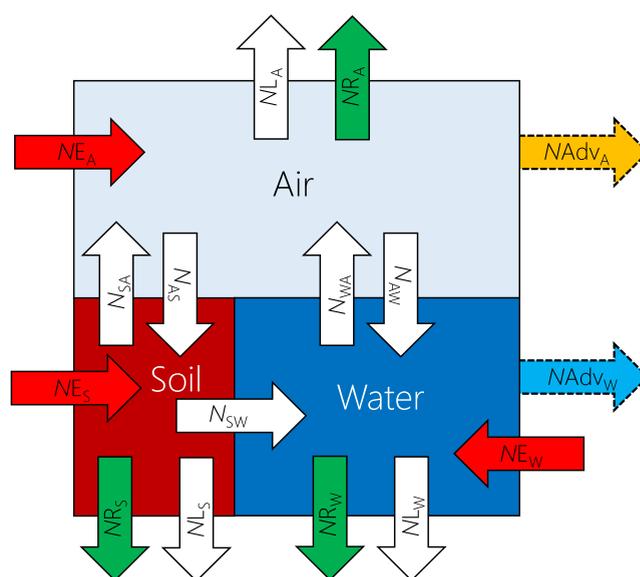


Figure 1. Compartments and processes of the level III steady state mass balance model in the Tool. A white arrow represents a transport flux, either occurring between air (A), soil (S), or water (W) within the model domain or from compartment X to the rest of the world (NL_X). A red arrow represents an emission flux into compartment X (NE_X). A green arrow represents a permanent loss rate by reaction in a compartment (NR_X). The advective flux of chemical via air ($NAdv_A$ in yellow) and water ($NAdv_W$ in blue) are calculated from the results of the steady state mass balance as described in the text.

Whereas the Tool describes a closed world, Wegmann et al. apply a scheme to nevertheless quantify the advective outflow of chemical in air ($NAdv_A$), which is required for the calculation of the TE.¹⁷ They assume that the wind in the Tool, which has a speed of 4 m/s or 14400 m/h, is blowing across a hypothetical area $Aadv_A$. This allows for an estimate of the amount of chemical which leaves the model region in air, without this flux becoming a part of the model’s mass balance equations. We have adopted a similar strategy to calculate outflow of chemical in water. The rationale and equations used to calculate CTD and TE in the Tool are included in Section S1.

Most process descriptions and many of the key environmental input parameters in the Tool trace their origin back to pioneering work by Mackay and co-workers.²⁷ While this does not imply that there might not be a need to reassess some of them, we chose to largely rely on the existing parametrization as this allowed us to more readily explore how our model predictions align with the Tool. However, as the assumption of constant drizzle in the Tool underestimates the potential for LRAT of both highly water-soluble chemicals and chemicals sorbed to particles during periods without precipitation²⁹ which could lead to false negative categorizations, we included a parametrization of intermittent precipitation in the Tool³⁰ (see Section S2).

2.2. Metrics. The emission fractions approach to LRTP assessment consists of a set of three coherent metrics. The environmentally dispersed fraction (ϕ_1) quantifies the relative extent to which a chemical can reach remote regions. The remotely transferred fraction (ϕ_2) expresses to what relative extent a chemical can reach surface media in remote regions. By accounting for degradative loss in surface media, the remotely accumulated fraction (ϕ_3) assesses the fraction of

chemical emissions accumulating in surface media of remote regions. Each metric is a fraction of the total amount emitted in the model environment as well as a fraction of the preceding metric. ϕ_2 is a fraction of ϕ_1 because only chemicals dispersed to a remote region can be transferred to the surface media there, and ϕ_3 is a fraction of ϕ_2 , because only chemicals transferred to those media can accumulate in them. **Figure 2**

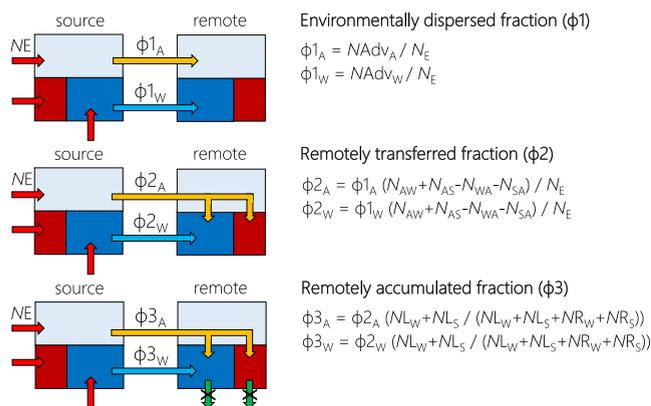


Figure 2. Representation of the three metrics in graphical terms. By only displaying elements crucial to the definition of the emission fractions, this model representation is simplified. For example, intermedia transfer in the source region and the reversibility of air surface exchange are not shown. See **Figure 1** for all considered processes and an explanation of the colored arrows.

highlights how the three fractions can be conceptually represented in a steady-state model environment, along with the simplified equations. Please note that following an idea first proposed for the derivation of the TE in a simple steady-state mass balance model,¹⁷ we use the same model environment and parametrization for the source and the remote region, i.e., **Figure 2** shows two environments that are represented by the same set of equations and input parameters. ϕ_1 is a transport-oriented metric resembling the CTD. ϕ_2 and ϕ_3 are target-oriented metrics having similarities with the TE and the ACP, respectively.

2.2.1. Environmentally Dispersed Fraction (ϕ_1). The relative potential for atmospheric dispersion is expressed as the fraction of emissions to all three media of the source region that is entering a remote region by air (A):

$$\phi_{1A} = N_{Adv_A} / N_E \quad (1)$$

where N_{Adv_A} (mol/h) is the outbound flux of chemical by advection into a hypothetical remote environment via the atmosphere, and N_E (mol/h) is the emission flux. N_{Adv_A} is the product of the chemical concentration in air (mol/m³), the cross-sectional area across which atmospheric advection occurs (A_{Adv_A} in m²) and wind speed (m/h). The numerical value for A_{Adv_A} was selected in such a way that the chemicals with the highest atmospheric dispersion potential are assigned a ϕ_{1A} of 1 (i.e., $N_{Adv_A} = N_E$). The maximum occurs for highly volatile chemicals emitted entirely into air (ϕ_{1A}) which neither react in air nor net-deposit from the atmosphere to surface media. Using partitioning properties of $\log K_{OA}$ of 2 and $\log K_{AW}$ of 4 for this inert “flyer”, an area of 2.27×10^9 m² causes ϕ_{1A} to adopt a value of 1. This area is used for calculating ϕ_{1A} for any other chemical of interest.

The relative potential of a chemical to undergo environmental dispersion by water (ϕ_{1W}) can be expressed using a similar equation:

$$\phi_{1W} = N_{Adv_W} / N_E \quad (2)$$

where N_{Adv_W} (mol/h) is the outbound flux of chemical by advection into a hypothetical remote environment via water. Here we use the existing water flow velocity in the Tool (0.02 m/sec or 72 m/h). The cross-sectional area for advective outflow via water A_{Adv_W} was derived from A_{Adv_A} using

$$A_{Adv_W} = A_{Adv_A} \times (h_W / h_A) \times 0.71 \quad (3)$$

where h_W and h_A are the heights of the water (100 m) and air compartment (6000 m) in the Tool, respectively, while 0.71 is the fraction of the global surface area covered by ocean. This results in an A_{Adv_W} of 2.68×10^7 m². This implies that a chemical would need to have a concentration in water 4.23 orders of magnitude higher than in air for air and water advection to be of similar importance.

Finally, the relative potential for a chemical to undergo environmental dispersion is calculated as

$$\phi_1 = \phi_{1A} + \phi_{1W} \quad (4)$$

2.2.2. Remotely Transferred Fraction (ϕ_2). ϕ_2 expresses the relative extent to which a chemical is (net) transferred to surface compartments following environmental dispersion to a remote region. The relative extent to which a chemical can be transferred from air (A) to surface compartments (S = soil, W = water) following atmospheric dispersion (LRAT) to a remote region is calculated using

$$\phi_{2A} = \phi_{1A} \times (N_{AS_a} + N_{AW_a} - N_{SA_a} - N_{WA_a}) / N_{E_a} \quad (5)$$

Note that the fluxes (N -values) in eq 5 refer to model results for a scenario with emissions to air only, irrespective of what the mode of emission to the source environment had been. The equation for transfer to both surface media after dispersion in water (ϕ_{2W}) is

$$\phi_{2W} = \phi_{1W} \times ((N_{E_w} - N_{WA_w} + N_{AW_w} - N_{SA_w} + N_{AS_w}) / N_{E_w}) \quad (6)$$

The fluxes in eq 6 always refer to a model scenario with 100% emissions to water (N_{E_w}) as only long-range transport with water (LRWT) into the remote region is being targeted.

The relative potential for a chemical to reach remote surface compartments following environmental dispersion is summarized as

$$\phi_2 = \phi_{2A} + \phi_{2W} \quad (7)$$

2.2.3. Remotely Accumulated Fraction (ϕ_3). Accumulation in both surface compartments following atmospheric dispersion and net atmospheric deposition within the remote region (ϕ_{3A}) is calculated using

$$\phi_{3A} = \phi_{2A} \times (N_{LS_a} + N_{LW_a}) / (N_{LS_a} + N_{LW_a} + N_{RS_a} + N_{RW_a}) \quad (8)$$

where N_{LS_a} and N_{LW_a} describe soil burial and transfer to the deep sea, respectively, and N_{RS_a} and N_{RW_a} represent reaction in soil and water, respectively. Eq 8 expresses the fraction of deposited chemical that is retained in the respective medium (soil or water) but transferred to deeper layers. In this manner,

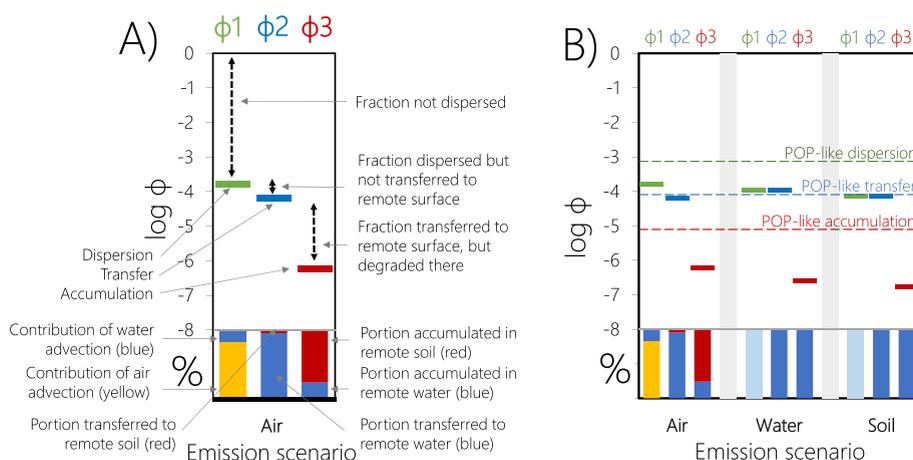


Figure 3. Visualization used to summarize the main results of the emissions fractions approach, using TCEP as an illustrative example. Panel A provides a description of the different elements of the figure using the example of the emission to air scenario. Panel B shows the complete visualization for all three emission scenarios and includes illustrative thresholds for POP-like L RTP behavior for each metric.

accumulation tendency can be estimated using a steady state model. ϕ_{3A} is equal to ϕ_{2A} for persistent chemicals but is much smaller for chemicals that readily degrade. ϕ_{3A} thus quantifies not only the extent to which a chemical can reach remote surface compartments but also the extent to which it persists there. Again, the fluxes in eq 8 refer to model results for a scenario with emissions to air only. Using similar reasoning for dispersion in seawater (except that it references results for a scenario with 100% emissions to water) yields

$$\phi_{3W} = \phi_{2W} \times (NLS_w + NLW_w) / (NLS_w + NLW_w + NRS_w + NRW_w) \quad (9)$$

The relative potential for a chemical to accumulate in surface compartments following environmental dispersion is then summarized as

$$\phi_3 = \phi_{3A} + \phi_{3W} \quad (10)$$

2.2.4. Transfer and Accumulation in Soil or Water. The approach introduced above not only allows for calculations of emission fractions ϕ_2 and ϕ_3 where both surface compartments are lumped together. For more in-depth analyses, it is possible to quantify the relative potential for transfer to, and accumulation in, individual surface compartments. All equations employed for implementation in the Tool are included in Section S3.

Sometimes, the concern is not restricted to the LRT of the originally emitted chemical but comprises any persistent environmental transformation product(s). Building on earlier approaches advocating for “joint” assessment metrics,^{31,32} it would be straightforward to estimate the fraction of the emission of a chemical that is transferred, deposited, or accumulated in either its original form or as its persistent degradation product(s).

2.3. Visualizing Results. The coherency of the emission fractions approach, along with the additivity of the equations, allows for the display of the main results in a format that fosters a comprehensive understanding of the processes which lead to dispersion, transfer, and accumulation. Specifically, we propose to use graphs of the type shown in Figure 3 to summarize and display the results of the emissions fraction approach. Figure 3A uses TCEP [ethanol, 2-chloro-, phosphate

(3:1)] as an illustrative example (Table S1) for the model scenario with 100% emissions to air. The three emissions fractions are designated by three colored markers in the upper part of the graph: green for the environmentally dispersed fraction, blue for the remotely transferred fraction, and red for the remotely accumulated fraction. The very wide range of values for emissions fractions necessitates the use of a logarithmic scale with an upper bound of $\log_{10} \phi$ of 0, i.e., a ϕ of 1. For TCEP emitted to air, ϕ_1 equals 0.016% ($\log_{10} \phi_1 = -3.8$), ϕ_2 equals 0.006% ($\log_{10} \phi_2 = -4.2$), and ϕ_3 is 0.00006% ($\log_{10} \phi_3 = -6.2$). The difference between the top of the graph and the position of the green marker corresponds to the fraction not dispersed, the difference between blue and green markers represents the fraction dispersed but not transferred to remote surface compartments, and the difference between red and blue markers is the fraction transferred but not accumulated. The stacked colored bars with a scale of 0 to 100% in the lower parts of Figure 3A,B provide additional information. Those shown below ϕ_1 indicate the relative importance of air and water advection to the chemical’s dispersal, i.e., LRAT (yellow) and LRWT (blue). The bars placed below ϕ_2 and ϕ_3 designate the relative portions transferred to, and accumulated in, the soil (red) and water (blue) of the remote region, respectively.

The three panels of Figure 3B further allow for a comparison of the results for the three modes of emissions. For illustration and to offer context for the numerical results for each of the three metrics, Figure 3B includes lines designating thresholds that separate chemicals that have POP-like L RTP from those that do not. Following Wegmann et al.,¹⁷ we defined these lines based on the lowest ϕ -value obtained for a subset of 14 discrete chemicals belonging to the initial “dirty dozen” POPs. These compounds have well characterized physical-chemical properties and fulfill the SC’s half-life criteria for persistence (see Table S2). We emphasize that what constitutes LRT is not primarily a scientific question but depends on the regulatory context. By using SC POPs to define the lines, they designate thresholds for global scale LRT. In a different regulatory context, different thresholds may apply, which could be defined on an expanded set of compounds that are deemed to satisfy the criterion of LRT on a smaller scale.

3. RESULTS AND DISCUSSION

3.1. Application 1: Exploring LRTP Behavior of a Chemical. The Tool was used to calculate the emission fractions for four selected chemicals with highly divergent LRTP behavior using three modes of emission (Figure 4).

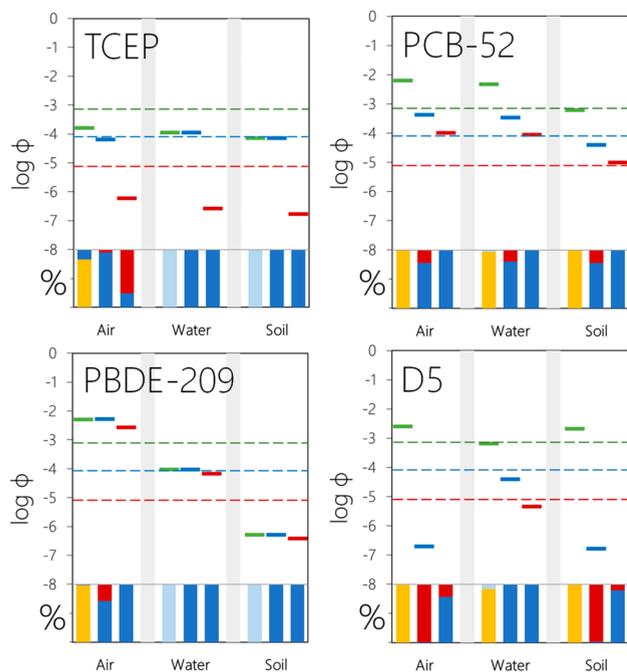


Figure 4. Results for a selection of chemicals with highly different LRTP behavior for three emission scenarios (see Figure 3 for explanation of legends).

TCEP ($\log K_{AW} = -7.5$, $\log K_{OW} = 1.7$) is a highly water-soluble chemical (or “swimmer”). PCB-52 ($\log K_{AW} = -1.96$, $\log K_{OW} = 6.26$) is a semivolatile organic contaminant (SVOC or “multi-hopper”). PBDE-209 ($\log K_{AW} = -6.6$, $\log K_{OW} = 8.7$) is an involatile chemical (or “single-hopper”) with a $\log K_{OA}$ of 15.3. Whereas PBDE-209 is predicted to be completely sorbed to particles in air, D5 ($\log K_{AW} = 3.16$, $\log K_{OW} = 6.78$) is a highly volatile chemical (or “flyer”) which occurs as a vapor in the atmosphere.

3.1.1. TCEP. If this chemical is emitted to water or soil, ϕ_1 equals ϕ_2 , i.e., all of the dispersed chemical is deposited to surface media in the remote region (Figure 4). From the stacked bars in the lower part of the graph, we see that this is because TCEP is predicted to reach the remote region by advection in water (>99.9%). This result supports the study by Sühling et al.²² who recently suggested modifications to the Tool to expand its utility for chemicals which are more prone to undergo LRWT.²² Only if TCEP is emitted to air is there a difference between ϕ_1 (dispersion) and ϕ_2 (transfer), whereby ~83% of the outflow from the source region is predicted to occur in air and the remainder in water. The latter contribution reflects the potential of TCEP to undergo wet deposition in the source region, followed by LRWT into the remote region. Notably, only a very small fraction of deposited TCEP accumulates in the remote region (ϕ_3).

3.1.2. PCB-52. The potential of PCB-52 for dispersion is attributed to LRAT irrespective of the mode of emission. As a persistent SVOC, PCB-52 is transferred quite readily between the three compartments in comparison to the other three

chemicals. In particular, PCB-52 volatilizes from surface media to which it has been emitted in source regions, allowing for LRAT (ϕ_1), followed by transfer (ϕ_2) and accumulation (ϕ_3). This explains why the relative distribution in terms of modes of transport, transfer, and accumulation show very minor differences across emission scenarios (stacked bars in Figure 4).

3.1.3. PBDE-209. If an involatile chemical like PBDE-209 is emitted to media other than air, then the mode of dispersion into the remote region, transfer, and accumulation will all be associated with the water compartment (i.e., $\phi_1 = \phi_{1W}$, $\phi_2 = \phi_{2W}$, and $\phi_3 = \phi_{3W}$), see stacked bars in Figure 4. If PBDE-209 is emitted to air, it has a high ϕ_1 (0.5%) with only a minor fraction being dispersed in water (1.3%). ϕ_2 equals ϕ_1 for PBDE-209 because the fraction dispersed in air is predicted to deposit on surface media in the remote region ($\phi_{2A} = \phi_{1A}$; eq 5), whereas the fraction dispersed in water is identical to the fraction which enters water in the remote region ($\phi_{2W} = \phi_{1W}$; eq 6). The relative portion transferred from air to each of the surface compartments of an involatile chemical as PBDE-209 is therefore by and large a reflection of the area fractions for soil and water in the model (71% water, 29% soil) as seen from the stacked bar in Figure 4. Given the default model assumption in the Tool, namely that particle-sorbed chemicals are persistent in air, the LRAT of PBDE-209 (as well as any other involatile chemical) is identical to the LRAT of the atmospheric particles. Reactions on particle surfaces have been shown to markedly reduce the atmospheric residence times of some involatile organic contaminants.³³ Given the large number of chemicals in commerce which fall into this category,^{23,34,35} this model assumption may lead to a significant risk for false positive LRATP categorizations. Opportunities to improve descriptions of processes that affect the LRAT behavior of involatile chemicals should therefore be pursued in future work.

3.1.4. D5. D5 is highly volatile and LRT is largely a result of LRAT, irrespective of mode of emissions (Figure 4, yellow bars). It has been shown to undergo LRAT where and/or when phototransformation is slow, such as at higher latitudes and/or during winter.³⁶ D5 is predicted to have a very limited potential for atmospheric removal by deposition (ϕ_{2A}), and the relative potential for D5 to accumulate in surface media as a result of atmospheric deposition is minuscule ($\log \phi_3 < -9.5$). Because $\log K_{AW}$ of D5 is very high (3.16), its potential for transfer from air to water is even smaller than that for transfer from air to soil (stacked bars in Figure 4). The potential for environmental dispersion (ϕ_1) is lowest if D5 is emitted to water where the potential for volatilization is mitigated by sorption of D5 to solids ($\log K_{OW} = 6.78$). The latter emission scenario also leads to a trapping effect as D5 is more persistent in water than air. The highest potential for both transfer (ϕ_2) and accumulation (ϕ_3) is therefore predicted when D5 is emitted to water. We highlight the divergence of the LRTP categorization of D5 based on the CTD (or ϕ_1) vs ϕ_3 (or ϕ_2). One could argue that judging D5 as having high LRTP in the sense of the SC based on its high CTD is a false positive decision because of its failure to accumulate in remote surface media to an extent sufficient to cause harm.

Impact of Mode of Emissions. Model scenarios involving 100% emissions into air usually represent the “worst-case” when it comes to a chemical’s potential for environmental dispersion (ϕ_1). This is the case for the four chemicals in

Figure 4. However, as the emission fractions approach, unlike the CTD and TE, explicitly accounts for chemical outflow with water, 100% emissions to air may not necessarily be the scenario which also leads to the highest ϕ_2 and ϕ_3 as noted for D5. Interestingly, the potential of TCEP for dispersion (ϕ_1) and accumulation in remote surface media (ϕ_3) are highest if emitted to air, whereas the potential for transfer to remote surface compartments (ϕ_2) is highest if emitted to water. For ϕ_1 and ϕ_3 in the air emission scenario, a significant portion of the transport to the remote region occurs via water (17% and 24%, respectively, Figure 4). These observations illustrate the need to both account for LRWT^{19,22} and consider emissions to air in LRTP assessment of swimmers.

3.2. Application 2: Comparing the LRTP Behavior of Different Chemicals with Each Other and with Threshold Values. When using the emission fractions approach to compare several chemicals with respect to their LRTP, it may be advisable to focus on selected results, such as the maximum values for ϕ_1 , ϕ_2 , and ϕ_3 obtained for any of the three emission scenarios. Figure 5 shows those values for eleven

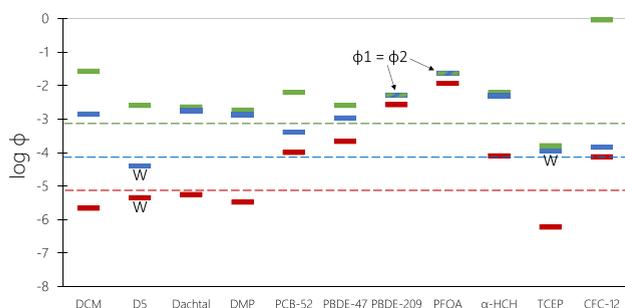


Figure 5. Predicted maximum values from the three emission scenarios for each of the three metrics for eleven selected chemicals (see Table S1 for explanation of acronyms). The dotted lines represent illustrative thresholds for potential POP-like dispersion (ϕ_1 , green), transfer (ϕ_2 , blue), and accumulation (ϕ_3 , red). The letter “W” indicates that the maximum value is predicted for the model scenario with 100% emissions to water. For all other values, the maximum values occur when the chemical is emitted to air, or they are identical across these two emission scenarios.

selected chemicals, together with the illustrative thresholds for POP-like LRTP behavior. Standard figures for the seven additional chemicals which were not included in Figure 4 are shown in Figure S2.

On the basis of the worst-case emission scenario, ten out of eleven chemicals are above the threshold for POP-like dispersion (ϕ_1), ten are also above the threshold for POP-like transfer (ϕ_2) although not the same chemicals, while six remain above the threshold for POP-like accumulation (ϕ_3). With ϕ_3 values above the red threshold line, Figure 5 attributes POP-accumulation behavior to chemicals listed under the SC (PCB-52, PBDE-47, PBDE-209, PFOA, and α -HCH), whereas those which are not (DCM, D5, Dacthal, DMP, and TCEP) have a ϕ_3 below that line. An exception is CFC-12, which is assigned POP-like accumulation even though it is not listed under the SC. Figure 5 suggests that ϕ_3 has the potential to identify chemicals for potential listing under the SC. It also highlights that neither a metric for dispersion (ϕ_1 or CTD) nor transfer (ϕ_2 or TE) is sufficient to do that. For example, an LRTP assessment based solely on a transport-oriented metric such as ϕ_1 would indicate POP-like dispersion for D5, although its transfer to remote surface media is very limited. Similarly, a CTD and a TE-based assessment will assign LRTP above POP-like thresholds to many chemicals that are too degradable to accumulate in remote surface compartments. Overall, the results in Figure 5 are consistent with expectations based on existing knowledge and illustrate that the emission fractions approach has the power to differentiate between different kinds of LRTP behavior across a diverse set of chemicals.

3.3. Comparison of the Approach with Earlier Metrics. The choice of metrics has consequences for the outcome of an LRTP assessment. In Table 1, we have listed the main advantages and disadvantages of the existing and alternative metrics as implemented in the Tool with emphasis on risks for false positives/negatives, decision making contexts, and research needs.

3.3.1. ϕ_1 versus CTD. CTD describes the potential for transport in the mobile media with simultaneous exchange with other media and predicts the distance from a point source at which the concentration of a chemical has been reduced to $\sim 37\%$. ϕ_{1A} is equal to CTD/CTD_{max} (where CTD_{max} is the CTD for an inert chemical), and hence ϕ_{1A} and CTD contain the same mechanistic information. CTD has dimensions of length. While a distance traveled by a chemical is intuitive and easily understood, the predicted distances should not be confused with actual transport distances in the real world. CTDs are not readily amenable to evaluation by higher-tier models that offer a more realistic representation of the environment. A metric which references emissions and estimates a fraction leaving a source region (ϕ_1) aligns better

Table 1. Advantages and Disadvantages of the Existing and Alternative Metrics

	OECD tool metrics	emissions fractions approach
transport-oriented metrics (CTD vs ϕ_1)	same mechanistic information	
	has intuitive dimension	has intuitive meaning
	requires definition of wind speed	requires definition of wind speed and cross-sectional area
	combined assessment of LRAT and LRWT not possible	LRAT and LRWT are additive
transfer-oriented metrics (TE vs ϕ_2)	gross deposition (for some chemicals TE > 100%)	net deposition
target-oriented metrics (ϕ_3)	includes transfer to remote region via air	includes transfer to remote region via air and water
	not included	allows distinction between transfer to versus accumulation in remote surface media
combined metrics	not coherent	coherent and multiplicative, allowing quantitative comparison of different LRTP metrics

with spatially resolved models predicting dispersion. For example, ϕ_1 mirrors the outflow ratio (OR) which predicts the export out of a model domain as a result of advection.⁷ However, the major disadvantage of the CTD is that it is not able to express the combined dispersion in air and water. This is unlike ϕ_1 which predicts the relative potential for dispersion in air, water, and both media combined. As the transport of chemical by either medium to a remote region should merit attention in the context of LRTP, we see this as a significant advantage.

3.3.2. ϕ_2 versus TE. The aim of both ϕ_2 and TE is to quantify the relative potential of chemicals for transfer to surface media in remote regions. TE was originally introduced as a metric to identify chemicals which are prone to undergo gross atmospheric deposition to the Great Lakes following LRAT, using the spatially resolved BETR North America model.¹⁸ TE as implemented in the Tool is also restricted to consideration of gross atmospheric inputs. As recognized by Wegmann et al.,¹⁷ values for TE could therefore exceed 100% for chemicals undergoing repeated air-surface exchange (see also Figure S3). TE also does not account for transfer with seawater to the Tool's remote region. This leads to a significant risk for false negatives for chemicals which are dispersed in water (LRWT) and therefore also in both air and water combined (LRT). ϕ_{2A} quantifies the net atmospheric deposition which is more relevant for LRTP assessments of SVOCs.^{11,14,37,38} ϕ_2 furthermore accounts for transfer to surface compartments in remote regions as a result of both LRAT and LRWT (eq 7). The choice of the transfer-oriented metric has implications for ranking chemicals according to LRTP. Two examples demonstrate the shortcomings of the TE relative to ϕ_2 . PFOA has a low TE (ranked no. 9 out of the 11 chemicals in Figure 5; data not shown) but a high ϕ_2 (ranked no. 1) because the latter metric correctly accounts for the considerable potential of PFOA for LRWT.³⁹ By being based on the gross atmospheric deposition flux, the TE for CFC-12 is a nonintuitive 975%, whereas the ϕ_2 for CFC-12 is 1.4×10^{-04} , which correctly indicates that most of this chemical remains airborne and only a small fraction is transferred to surface media.

3.3.3. ϕ_3 versus ϕ_2 and TE. Existing LRTP metrics have been classified as being either transport- or target-oriented. Here we deliberately distinguish between metrics quantifying transfer to (ϕ_2), and accumulation (ϕ_3) in, the remote region, and we think that this needs to be reflected in an expanded classification system of transport-, transfer-, and target-oriented metrics. While TE was introduced as a target-oriented metric, we believe this metric is better classified as a transfer-oriented metric along with ϕ_2 . ϕ_3 provides an assessment of LRTP in the context of listing chemicals in the SC by offering an estimate of a chemical's potential to be retained in the surface compartment(s) within a remote region, where the ability of a chemical to elicit adverse effects in surface media is decisive.

3.4. Strength and Limitations of the Emissions Fraction Approach. We used here the OECD Tool to introduce and illustrate the new system of LRT metrics because this widely accepted consensus model constitutes the current state-of-the-art in regulatory LRT assessment, as is for example evident in its frequent use in nominations of chemicals for listing in the SC. This should not be interpreted as an uncritical endorsement of this simple model and its current parametrization or as suggesting that there is no role for more complex models to play when assessing LRT. In fact,

the emissions fraction approach to LRT assessment is not tied to a particular model but could be implemented with a variety of models of different spatial, temporal, and process resolution.

We have shown that the emission fractions approach introduced herein offers several advantages over the metrics currently implemented in the Tool. We therefore recommend that screening models such as the Tool include the emission fractions approach for coherent, transparent, and more reliable LRTP assessments. A clear advantage of the emission fractions approach in comparison to CTD and TE is that it helps identify and even quantify the more influential processes affecting LRT. We believe this aspect is important because there are some obvious, albeit deliberate, limitations to what a simple screening model as the Tool can do, such as accounting for temporal and spatial variability. It is thus important that simple screening models provide mechanistic insight to help guide higher-tier LRTP assessment. In future work, we will therefore explore how the emission fractions approach can be applied to gain more detailed insight into the pathways of transfer to, and accumulation in, individual surface compartments in remote regions.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.2c03047>.

Additional model equations, input parameters, and results (PDF)

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Notes

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