Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Source-to-exposure assessment with the Pangea multi-scale framework – Case study in Australia – Electronic Supporting Information (ESI)

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Section S1 Receptor perspective – block of FF

Receptor perspective and source apportionment – For simplifying the source apportionment, we computed the block of FF, the matrix of fate factors (section 2.1 main text), that corresponds to transfers from the first layer of the atmospheric grid to itself (Figure 5 main text). This block defines the contribution of each cell of this layer to the mass at steady state in each other cell.

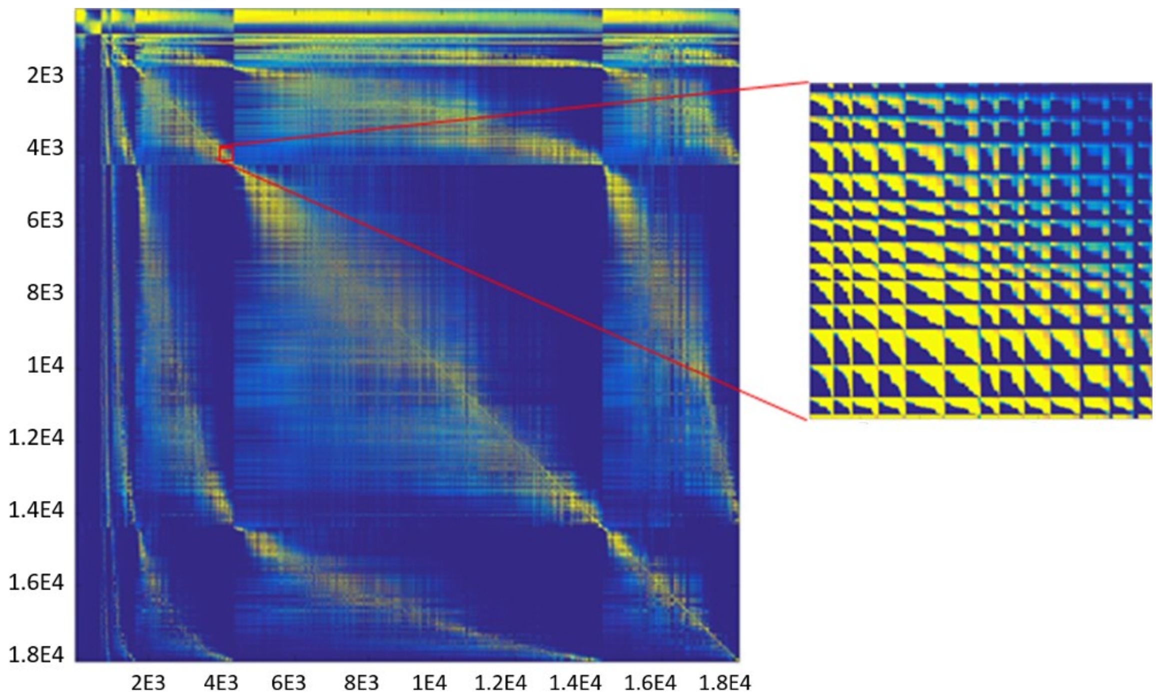


Figure S1 – **Left**: 18,107×18,107 dense block of a FF matrix (for benzene and the geometry of the Australian project), associated with transfers from the atmospheric layer #1 to itself. **Right**: zoom on a small part, detail of fine pattern. The full 109,766 × 109,766 FF matrix cannot be computed fully/directly.

Section S2 Emitter perspective, formaldehyde

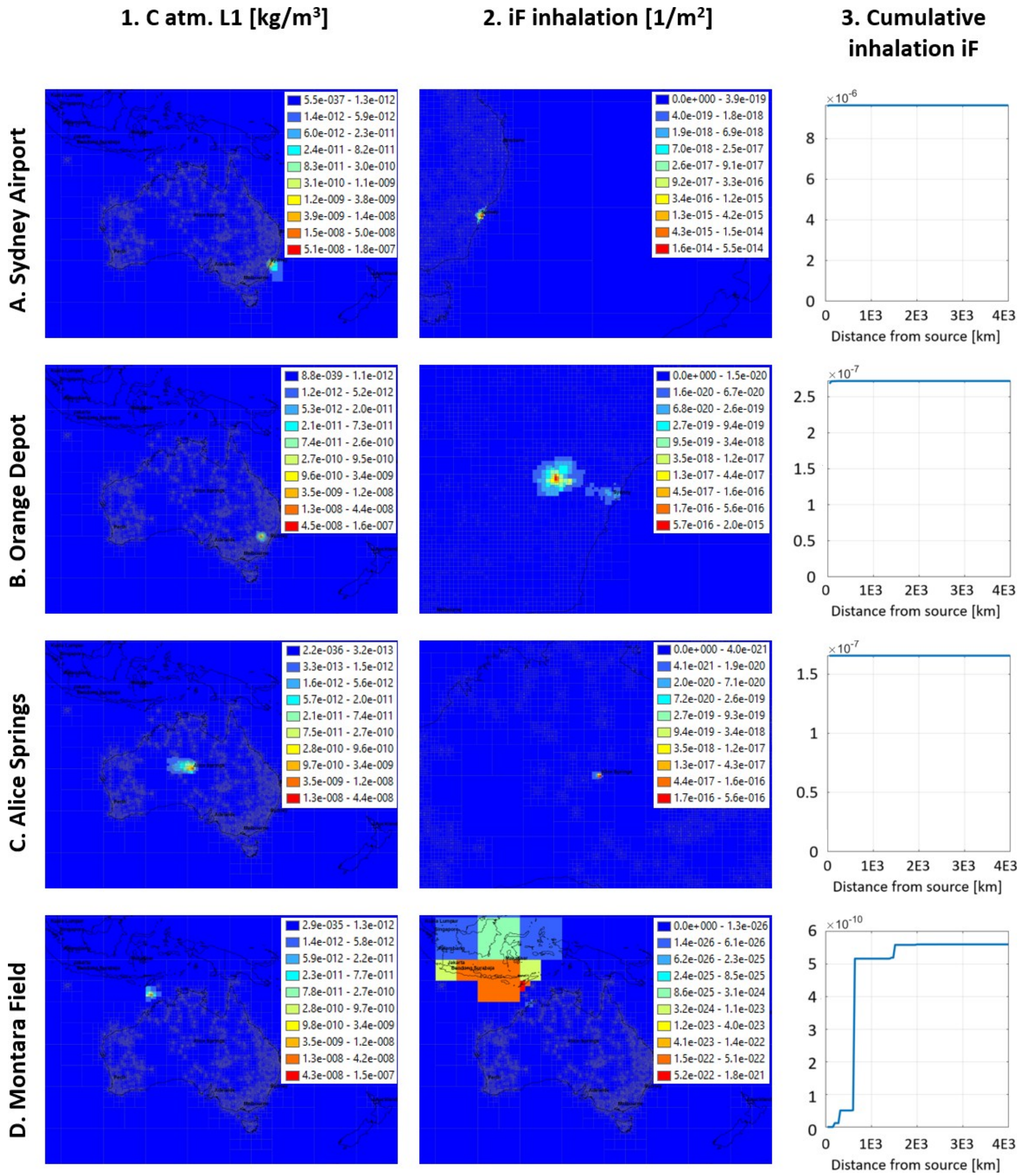
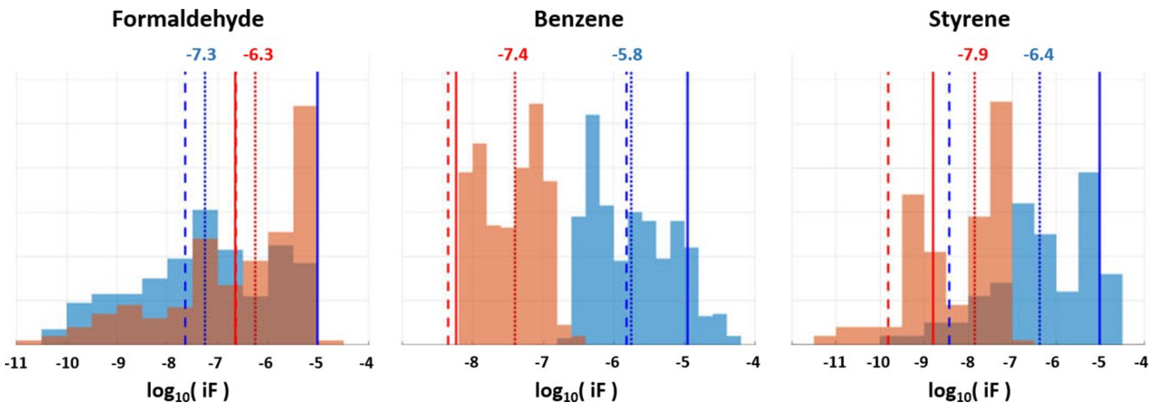


Figure S2 – **1.** Atmospheric concentration in layer #1, **2.** Inhalation iF per square meter, and **3.** Cumulative radial statistics of inhalation iF, for a unit emission flow of 1 kg/s of formaldehyde in **A.** Sydney airport (urban), **B.** Orange depot, 200 km North-West of Sydney (rural), **C.** Alice Spring, desert, and **D.** the Montara Field oil platform (remote, sea).

Section S3 Emitter perspective, comparison of global intakes with USEtox

We generalize the study of iFs through inhalation and ingestion to all substances and all emission points, and we compare distributions of global iFs computed by *Pangea*, with pairs of iFs computed with USEtox and associated with the urban and continental (rural) archetypes. To illustrate this comparison with benzene, the 755 emissions (reports) of benzene defined by NPI are located in 552 atmospheric cells; we compute global iFs associated with these cells, and we get a distribution. We then extract urban and continental iFs for Australia (zone 10) from USEtox, and we compare. Figure S3A shows results for formaldehyde, benzene, and styrene. We observe a good match for inhalation iFs (blue), where USEtox values for urban emissions (plain blue line) and for continental emissions (dashed blue line) are well within *Pangea*'s distributions (median value marked by dotted blue line with value). Intake fractions through total ingestion are generally an order of magnitude (max) higher in *Pangea* than in USEtox.

Figure S3B shows a comparison for all substances. Points are defined by USEtox mean value (x axis) and *Pangea* mean value (y), bars are defined by urban/continental values for USEtox, and one standard deviation for Pangea. We observe a relatively good match between *Pangea* and USEtox for inhalation iFs (Pearson r = 0.5, RMSE = 0.61) with generally less than an order of magnitude difference between the two models, and a good match for ingestion iFs, where *Pangea* provides consistently larger values than USEtox (Pearson r = 0.94, RMSE = 0.89). The inhalation iF of the USEtox urban archetype is constant ($\log_{10}$iF = -5, iF = 10 ppm) for most substances, unless persistence in air is high, whereas *Pangea* is able to account for the effective population density around sources.



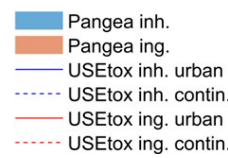


Figure S3A – Comparison of distributions of intake fractions computed with *Pangea* and pairs of intake fractions associated with rural and continental archetypes computed with USEtox. Dotted blue lines mark median values of *Pangea* distributions.

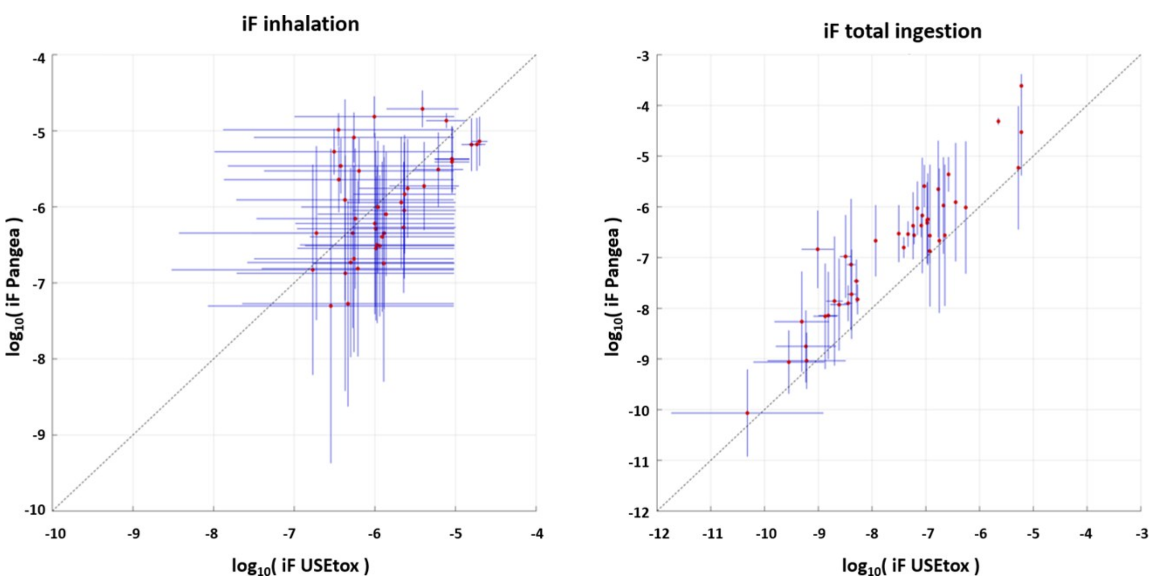


Figure S3B – Comparison of inhalation and ingestion intake fractions from USEtox (x axis, mean, continental, urban) and *Pangea* (y axis, one standard deviation), for the 43 substances.

Section S4 Overall population exposure to the 4,101 sources of the NPI inventory

S4.1 Full sector names for Figure 4 (main text)

Table S1 – Full sector names corresponding the labels in Figure 4.

A1

1701	Petroleum Refining and Petroleum Fuel Manufacturing
1709	Other Petroleum and Coal Product Manufacturing
3321	Petroleum Product Wholesaling
2110	Iron Smelting and Steel Manufacturing
1821	Synthetic Resin and Synthetic Rubber Manufacturing
1811	Industrial Gas Manufacturing
700	Oil and Gas Extraction
2921	Waste Treatment and Disposal Services
1812	Basic Organic Chemical Manufacturing
2132	Aluminium Smelting
2121	Iron and Steel Casting
600	Coal Mining
1813	Basic Inorganic Chemical Manufacturing
2611	Fossil Fuel Electricity Generation
5309	Other Warehousing and Storage Services

A2

1811	Industrial Gas Manufacturing
700	Oil and Gas Extraction
2110	Iron Smelting and Steel Manufacturing
801	Iron Ore Mining
3321	Petroleum Product Wholesaling
1701	Petroleum Refining and Petroleum Fuel Manufacturing
1709	Other Petroleum and Coal Product Manufacturing
2393	Railway Rolling Stock Manufacturing and Repair Services
1813	Basic Inorganic Chemical Manufacturing
2132	Aluminium Smelting
2921	Waste Treatment and Disposal Services
1821	Synthetic Resin and Synthetic Rubber Manufacturing
600	Coal Mining
802	Bauxite Mining
5309	Other Warehousing and Storage Services

B1

2619	Other Electricity Generation
1701	Petroleum Refining and Petroleum Fuel Manufacturing
2812	Sewerage and Drainage Services
2090	Other Non-Metallic Mineral Product Manufacturing
600	Coal Mining
2032	Plaster Product Manufacturing
2131	Alumina Production
1811	Industrial Gas Manufacturing
5211	Stevedoring Services
1494	Reconstituted Wood Product Manufacturing
2921	Waste Treatment and Disposal Services
1499	Other Wood Product Manufacturing n.e.c.
2021	Clay Brick Manufacturing
5212	Port and Water Transport Terminal Operations
2010	Glass and Glass Product Manufacturing

B2

1701	Petroleum Refining and Petroleum Fuel Manufacturing
2619	Other Electricity Generation
2132	Aluminium Smelting
1494	Reconstituted Wood Product Manufacturing
1499	Other Wood Product Manufacturing n.e.c.
600	Coal Mining
801	Iron Ore Mining
804	Gold Ore Mining
2090	Other Non-Metallic Mineral Product Manufacturing
1812	Basic Organic Chemical Manufacturing
700	Oil and Gas Extraction
2921	Waste Treatment and Disposal Services
2700	Gas Supply
2032	Plaster Product Manufacturing
5211	Stevedoring Services

S4.2 Analysis per sector for styrene and dichloroethane

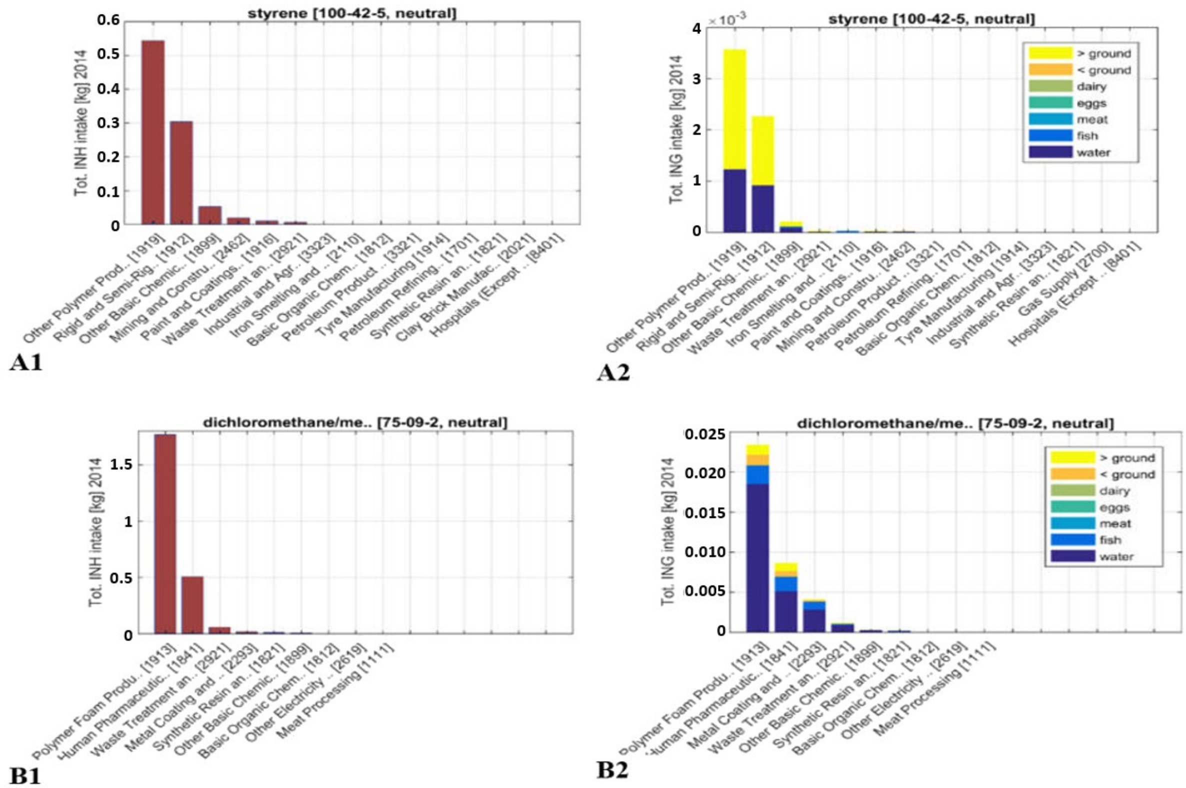


Figure S4 – Total inhalation (column 1) and ingestion (column 2) intakes per NPI Australian sector, for A. styrene and B. dichloromethane. Full sector names are available in Table S2, ESI.

Figure S4A shows that for styrene and dichloromethane, most contributing sectors are the chemical and pharmaceutical industry. In term of exposure pathway for the ingestion route, none of these substances tends to bioaccumulate, so that the direct water ingestion is dominant for all substances but styrene, for which the ingestion via aboveground crops is dominant but nevertheless low compared to inhalation.

S4.3 Full sector names for Figure S4

Table S2 – Full sector names corresponding the labels in Figure S4.

A1

1919	Other Polymer Product Manufacturing
1912	Rigid and Semi-Rigid Polymer Product Manufacturing
1899	Other Basic Chemical Product Manufacturing n.e.c.
2462	Mining and Construction Machinery Manufacturing
1916	Paint and Coatings Manufacturing
2921	Waste Treatment and Disposal Services
3323	Industrial and Agricultural Chemical Product Wholesaling
2110	Iron Smelting and Steel Manufacturing
1812	Basic Organic Chemical Manufacturing
3321	Petroleum Product Wholesaling
1914	Tyre Manufacturing
1701	Petroleum Refining and Petroleum Fuel Manufacturing
1821	Synthetic Resin and Synthetic Rubber Manufacturing
2021	Clay Brick Manufacturing
8401	Hospitals (Except Psychiatric Hospitals)

A2

1919	Other Polymer Product Manufacturing
1912	Rigid and Semi-Rigid Polymer Product Manufacturing
1899	Other Basic Chemical Product Manufacturing n.e.c.
2921	Waste Treatment and Disposal Services
2110	Iron Smelting and Steel Manufacturing
1916	Paint and Coatings Manufacturing
2462	Mining and Construction Machinery Manufacturing
3321	Petroleum Product Wholesaling
1701	Petroleum Refining and Petroleum Fuel Manufacturing
1812	Basic Organic Chemical Manufacturing
1914	Tyre Manufacturing
3323	Industrial and Agricultural Chemical Product Wholesaling
1821	Synthetic Resin and Synthetic Rubber Manufacturing
2700	Gas Supply
8401	Hospitals (Except Psychiatric Hospitals)

B1

1913	Polymer Foam Product Manufacturing
1841	Human Pharmaceutical and Medicinal Product Manufacturing
2921	Waste Treatment and Disposal Services
2293	Metal Coating and Finishing
1821	Synthetic Resin and Synthetic Rubber Manufacturing
1899	Other Basic Chemical Product Manufacturing n.e.c.
1812	Basic Organic Chemical Manufacturing
2619	Other Electricity Generation
1111	Meat Processing

B2

1913	Polymer Foam Product Manufacturing
1841	Human Pharmaceutical and Medicinal Product Manufacturing
2293	Metal Coating and Finishing
2921	Waste Treatment and Disposal Services
1899	Other Basic Chemical Product Manufacturing n.e.c.
1821	Synthetic Resin and Synthetic Rubber Manufacturing
1812	Basic Organic Chemical Manufacturing
2619	Other Electricity Generation
1111	Meat Processing

Section S5. Exposure to severity and damage on human health

Using USEtox human effect and dose-response factors (in DALYs/kg_{intake}), we compute cancer and non-cancer effects, and then cancer and non-cancer Disability Adjusted Life years (DALYs). Figure S5 shows all 43 simulated substances, ranked based on total DALYs. It suggests that the nation-wide human health footprint of the considered point source emissions of organic compounds is restricted, with a maximum of 30 DALYs per year for formaldehyde. Even considering the high uncertainty associated with the USEtox effect factors – typically a geometric squared deviation of a factor 60 on the dose response of case of non-cancer per kg_{intake}, and a factor 13 for the severity factor of DALY per case of non-cancer[1] – this level of health impact associated with the considered NPI source emission of organic compounds is much lower than the 38,000 DALY/year estimated in the Global Burden of Disease for fine particulate matter impacts in Australia[1]. It is however important to emphasize that the NPI emission inventory only considers emissions related to industry, and does not account for most of e.g. traffic related emissions[2].

[1] https://vizhub.healthdata.org/gbd-compare/

[2] It provides estimates of emissions related to motor vehicles only for a limited set of air sheds that were not included in this study.

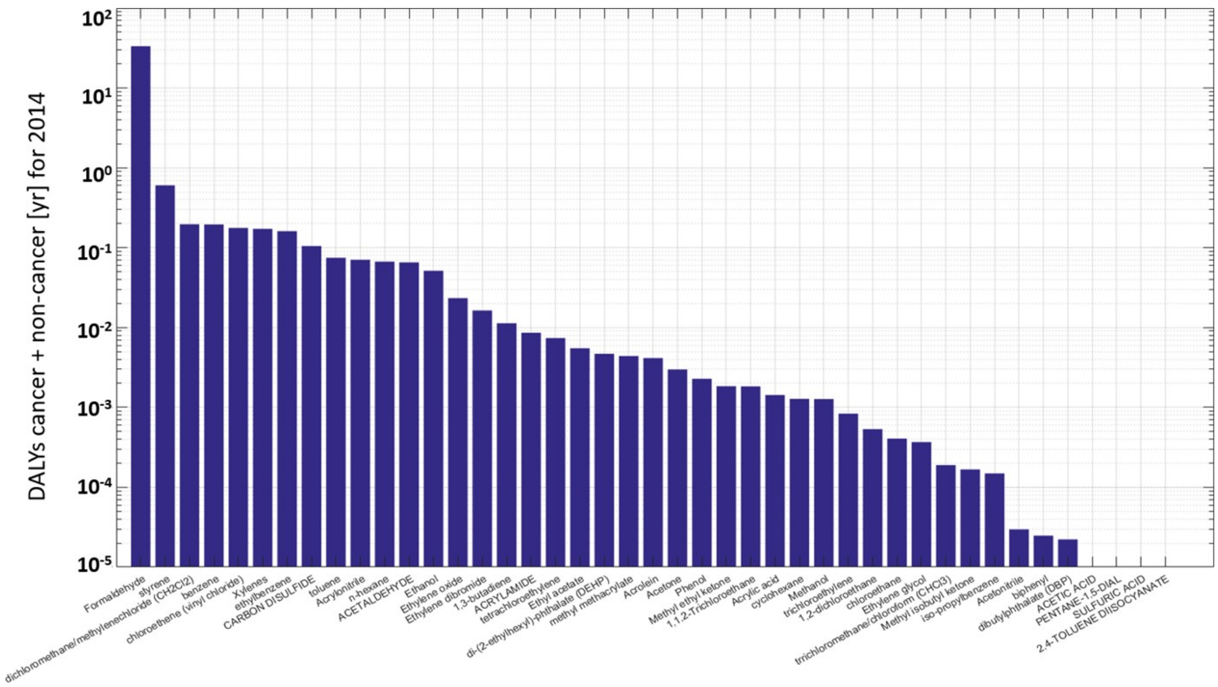


Figure S5 – Estimates of total cancer and non-cancer DALYs associated with the overall emissions of 43 organic compounds of the NPI inventory, ranked from highest to lowest total DALY.

Section S6 Fate factor maps of formaldehyde

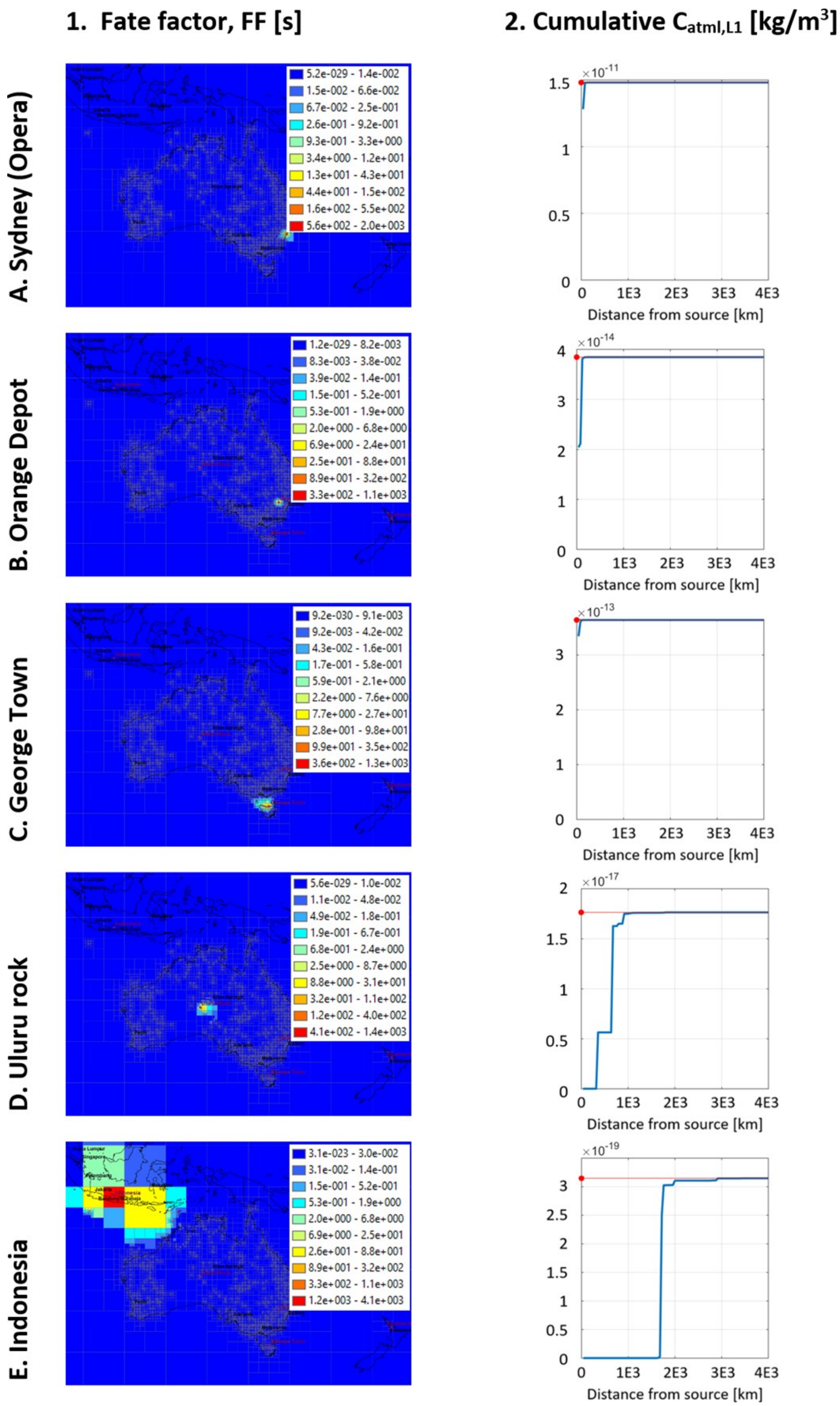


Figure S6 – **1.** Maps of fate factors (increase in receptor concentration due to emission of 1 kg/s in the considered cell) and **2.** radial statistic of contributing Australian sources for benzene, for receptors in **A.** Sydney opera, **B.** Orange depot, 200 km North-West of Sydney, **C.** George Town, Tasmania, **D.** Uluru Rock, and **E.** Indonesia. The sum of remote contributions to the local concentration, computed with source apportionment, converges towards the concentration computed based on total emissions.

Section S7 – Extra material for contrasting emitter vs receptor perspectives

Figure S7 shows locations of emission sources that contribute the most to the concentration at the Sydney opera. Most of the contributing sources are in the Sydney agglomeration itself, especially for formaldehyde, with very limited contributions from sources outside of Sydney. For benzene, however, we observe substantial contributions from more distant sources.

It is only in the case of Uluru rock, with no important close sources, that contributions from distant sources represent a dominant share (Section S8). From a receptor perspective, the contribution of local sources substantially contributes to the total concentration even for a persistent substance such as benzene, unless we are in a really desert area (Uluru rock) without local source or if we disregard the local emissions (Indonesia).

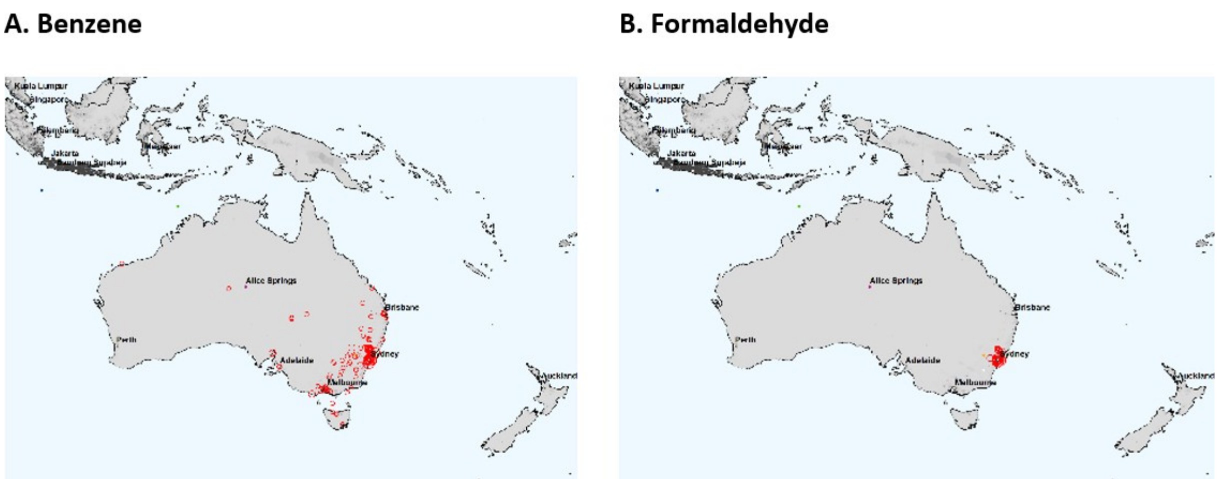


Figure S7 – Maps of locations of main sources contributing to concentration of **A.** benzene and **B.** formaldehyde at the Sydney Opera.

Section S8 Source apportionment at Uluru Rock

Tracing further major contributors – Using lists of largest contributing cells, we can extract NPI reports and rank them by emission. Taking the Uluru rock, for example, we find that the major contribution to the local atmospheric concentration of benzene is cell 7183, North-West of Adelaide (red arrow, Figure S8), which encompasses emissions from NPI reports {2469, 2946, 3752}. Table S3 shows the ranking of these reports by total emission of Benzene during year 2014, differentiated by industry sector to identify the main contributing industries.

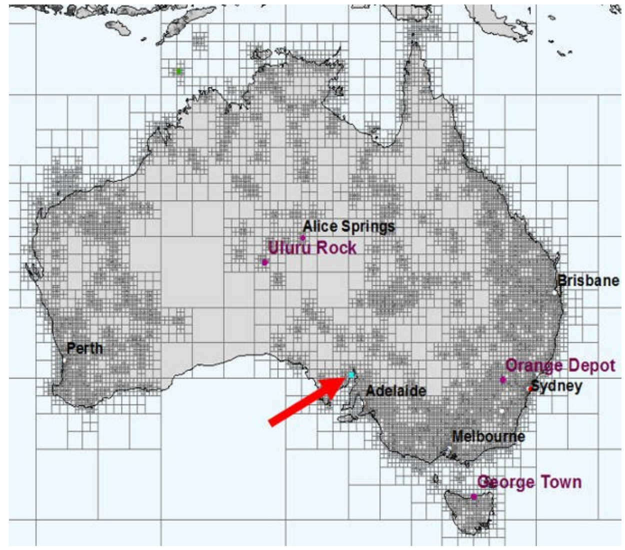


Figure S8 – Identification of the cell that contributes the most to the atmospheric concentration of benzene at Uluru Rock.

Table S3 – Ranking of reports associated with cell 7183, major contributor to the intake of benzene at the Uluru rock.

Rank	Report ID	Company sector	Total emission [kg] for 2014
1	3752	Iron Smelting and Steel Manufacturing	$4.7{\times}10^4$
2	2946	Rail Freight Transport	0
2	2469	Other Non-Metallic Mineral Mining and Quarrying	0

Section S9 Source apportionment at Uluru Rock

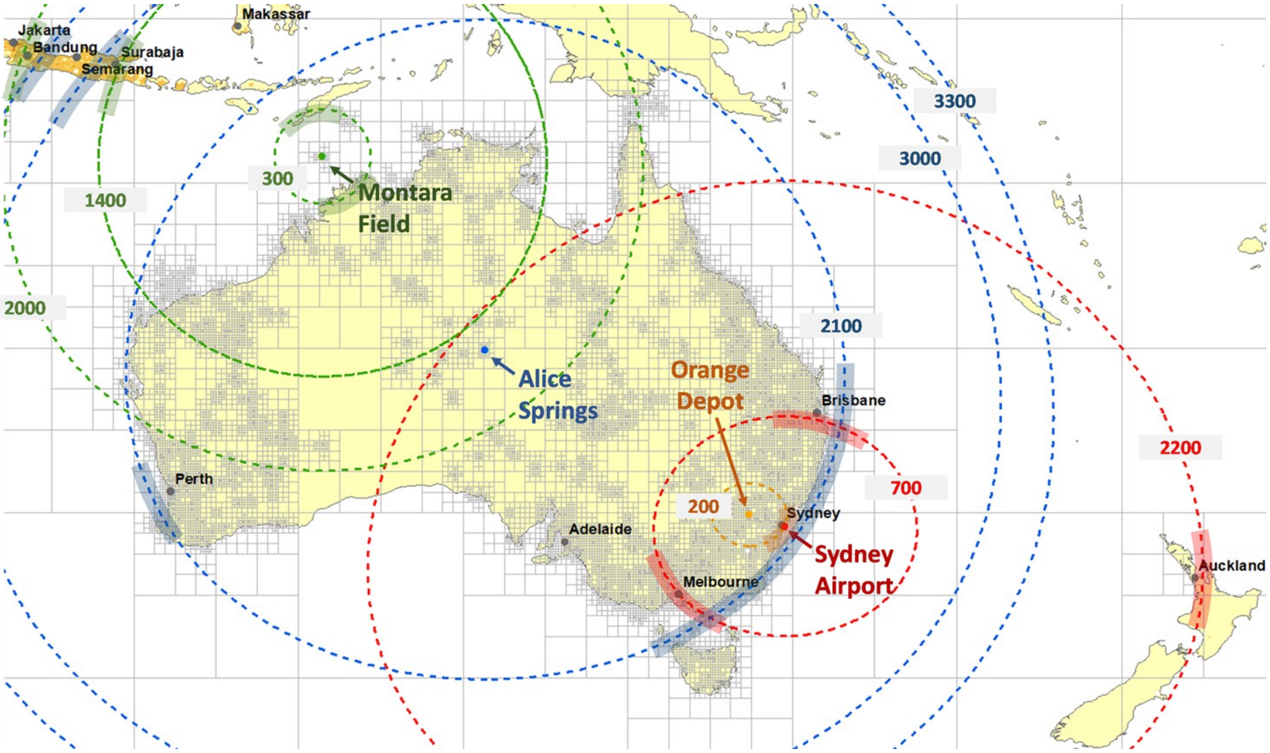


Figure S9 – Circles around emission source, approximately at the distances of the steps in the cumulative radial statistics of inhalation iF. Highlights along the circles identify main cities or regions corresponding to these steps. Sydney airport, red circles at 700 km and 2,200 km. Alice Springs, blue circles at 2,100 km, 3,000 km and 3,300 km. Montara Field, green circles at 300 km, 1,400 km and 2,000 km. The Orange Town: orange circle at 200 km.

Section S10 Pangea framework: geometric and virtual systems

Two of the main challenges in Pangea are to build a global 3D multi-scale geometry made of grids and grid cells, and then to transform this system into a mathematical compartmental system. This is illustrated in Figure S10:

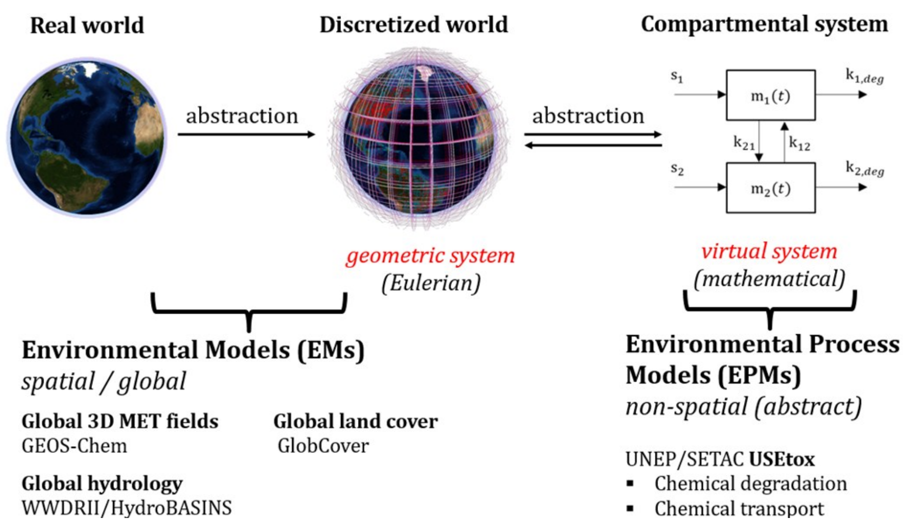


Figure S10 – Abstraction layers in Pangea, the geometric and virtual systems.

The set of grids and grid cells, which discretizes the world spatially, is called the *geometric system*. Geo-referenced data are associated with this system by projection. Figure S11 schematizes a portion of this system:

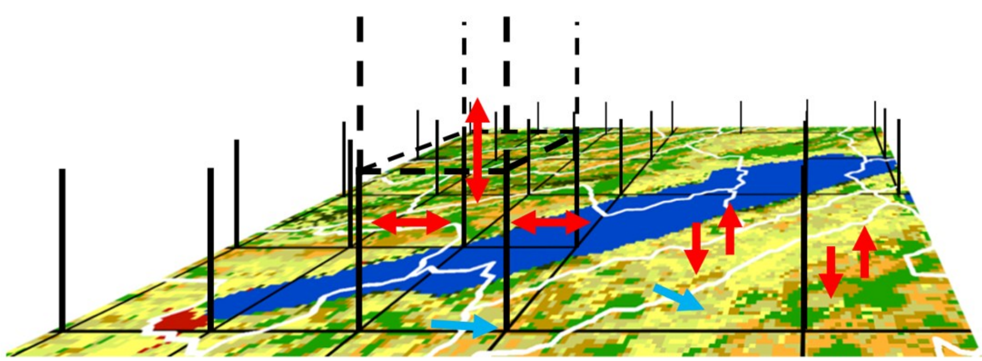


Figure S11 – The geometric system: complex geometry of cells with irregular boundaries and connections, and heterogeneous content (in terms of media).

Plain black lines represent the first layer of the atmospheric grid (made of 17 layers by default), plain white lines represent a terrestrial grid made of hydrological catchments defined by HydroBASINS, and the raster (pixels at ground level) represent a data set of land cover (here GlobCover). This small examples illustrates the complexity of connections between grids, and the fact that the terrestrial grid delineated regions with heterogeneous content. Pangea transforms this system into a compartmental system called the *virtual system*. This system is made of homogeneous compartments with simple connections. Environmental process models (e.g. from USEtox and IMPACT 2002) are associated with this system, in the sense that they describe elimination and transfer within and between the compartments.

Section S11 Observed data and comparison with predicted increase at 7 km × 7 km resolution

The National Environment Protection Council published in 2010 a mid-review document "The National Environment Protection (Air Toxics) Measure"[2] made in 2004, reporting monitored concentrations for benzene, formaldehyde, toluene and xylene in Australia. Table 5 summarized the observed concentrations for four archetypical locations: roadside, Central Business District, residential and industrial.

Since monitored concentrations result from overall emissions, and not just from industrial sources as the predicted concentration from Pangea in the present study, monitored data are likely to represent an upper limit of the predicted concentration. Keeping this in mind, we performed a short indicative comparison of our predicted concentrations from industrial sources with the total observed concentrations of benzene, formaldehyde, toluene and xylene in Australia. Of special interest is a comparison of concentration monitored on industrial sites with the Pangea predicted concentration on the 7 km × 7 km grid cells that encompass industrial sources.

As expected from prediction derived from a partial emission inventory, it is only the maximum predicted concentrations that fall close to the range of observed data, and the predicted concentrations are lower than the total monitored concentrations accounting for all sources. Interestingly, for benzene and xylene, the residential concentrations are substantially higher than the industrial ones, whereas for toluene the highest concentrations are observed on roadside. This suggests that sources other than industrial ones might play a dominant role, e.g. linked to consumer usage in residential areas. For formaldehyde, concentrations are higher in industrial areas and Pangea predictions underestimate the concentrations in these areas. This could be due to missing emission sources (emphasizing the need for improved input data), or is related to the 7 km × 7 km resolution.

To identify the cause behind such differences, a proper validation would necessitate to account for all sources, with a careful sampling strategy and corresponding model resolution.

Table S4 – Monitored annual concentrations of all samples according to NEPM (2010) for 4 locations and Pangea predicted concentrations for cells with individual sources of benzene, toluene, formaldehyde and xylene.

Data source	Substance	Location	No of samples	No of Sites	Mean	50th %tile	95th %tile	max
			-	-	µg/m3	µg/m3	µg/m3	µg/m3
Monitoring	Benzene	ROADSIDE	361	6	3.2	3.2	9.6	-
Monitoring	Benzene	CBD[1]	91	4	0.0	0.00	3.2	-
Monitoring	Benzene	RESIDENTIAL	785	11	28.7	0.00	137.3	-
Monitoring	Benzene	INDUSTRIAL	3603	35	0.0	0.00	3.2	-
Pangea predicted	Benzene	IND. SOURCES	-	552	0.019	0.002	0.05	3.96
Monitoring	Toluene	ROADSIDE	374	5	15.1	7.5	45.2	-
Monitoring	Toluene	CBD[1]	295	3	7.5	3.8	15.1	-
Monitoring	Toluene	RESIDENTIAL	2600	19	7.5	3.8	18.8	-
Monitoring	Toluene	INDUSTRIAL	1446	28	3.8	0.0000	7.5	-
Pangea predicted	Toluene	IND. SOURCES	-	651	0.037	0.003	0.17	5.15
Monitoring	Formaldehyde	ROADSIDE	195	2	4.9	4.9	13.5	-
Monitoring	Formaldehyde	CBD[1]	396	2	3.7	3.7	4.9	-
Monitoring	Formaldehyde	RESIDENTIAL	823	13	4.9	2.5	16.0	-
Monitoring	Formaldehyde	INDUSTRIAL	456	6	9.8	11.0	23.3	-
Pangea predicted	Formaldehyde	IND. SOURCES	-	349	0.037	0.006	0.16	0.68
Monitoring	Xylenes	ROADSIDE	174	4	4.3	4.3	8.7	-
Monitoring	Xylenes	CBD[1]	136	3	4.3	4.3	4.3	-
Monitoring	Xylenes	RESIDENTIAL	599	10	21.7	4.3	208.3	-
Monitoring	Xylenes	INDUSTRIAL	1446	32	0.0	0.0000	4.3	-
Pangea predicted	Xylenes	IND. SOURCES	-	735	0.020	0.001	0.09	1.80

CBD[1]=Central Business District

Bibliography

1 P. Fantke, R. Friedrich and O. Jolliet, Health impact and damage cost assessment of pesticides in Europe, *Environ. Int.*, 2012, **49**, 9–17.

2 National Environment Protection Council, *The National Environment Protection (Air Toxics) Measure*, 2010.