Supplementary materials

Integrating modes of transport in a dynamic modelling approach to evaluate urban population exposure to NO_2 and $PM_{2.5}$ pollution

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Supplement S1—Emission downscaling procedure:

We used CAMS-REG-AP v3.1 (Granier et al. 2019) emission data downscaled the data from a horizontal resolution of ca. a 0.1° x 0.05° (lon × lat) to a ca. 1 km x 1 km resolution. CAMS-REG-AP v3.1 (Granier et al. 2019) follows the sector classification GNFR, which is an aggregated version of the NFR (Nomenclature For Reporting, (EEA 2016)) and provides sectoral annual emission totals for Europe. We applied sector specific proxies to downscale the regional emission inventories to a resolution of 1 × 1 km^2. The downscaling procedure followed a generalized framework:

1. First, a set of suitable proxies for the sector of interest was defined.

2. Second, the proxy grids were gridded and resampled (nearest neighbor) to match extent and resolution of the EPISODE-CityChem modeling domain. We applied proxy data from different sources, such as Corine Land Cover 2018 (CLC2018, (Copernicus Land Monitoring Service 2018)), the Global Human Settlement Population Grid (GHS-Pop, (Florczyk et al. 2019)), and source information of the European Pollutant Release and Transfer Register (E-PRTR). A full list of applied proxies can be found in Supplement Table S-T2.

3. Third, the chosen proxy was normalized by calculating a factor that is indicating the proportion of each proxy data type in one high-resolution grid cell within one coarse grid cell.

4. In a fourth and last step, these factors are used to downscale the respective emissions in the respective area top-down. Thus, the spatial distribution information of the coarse CAMS-REG-AP grid is considered in downscaling to the high-resolution grid (1 km x 1km). This framework was applied to downscale all gridded sector emissions of the CAMS-REG-AP emission inventory to create are emissions for the urban domain.

Table S1-1. Applied proxies to downscale regional emissions to the urban-scale.

Type	GNFR	SNAP	Proxy / Downscaling Approach	Emission Data Source
Point	A_PublicPower	1	If reported emissions are lower than CAMS-REG-AP v3.1: residual emissions are distributed to SNAP 1, 3 or 4 area emissions (see below)	11. BImSchV & CAMS-REG-APv3.1
Point	B_Industry	3, 4		11. BImSchV & CAMS-REG-APv3.1
Line	F_RoadTransport	7	Downscaled with GHS-Pop, weighted with factor of 3 (Kuik et al. 2018 (Kuik et al. 2018)) in areas defined as urban center by GHS Urban Centre database and distributed to major Open Street Map road types.	CAMS-REG-APv3.1
Area	A_PublicPower	1	CLC2018 code 121	CAMS-REG-APv3.1
Area	B_Industry	3, 4	CLC2018 code 121	CAMS-REG-APv3.1
Area	C_OtherStationaryComb	2	GHS-Pop	CAMS-REG-APv3.1
Area	D_Fugitives	5	CLC2018 code 121	CAMS-REG-APv3.1
Area	E_Solvents	6	GHS-Pop	CAMS-REG-APv3.1
Area	G_Shipping	8	CLC2018 codes 123, 523, 522	CAMS-REG-APv3.1
Area	H_Aviation	11	CLC2018 codes 124	CAMS-REG-APv3.1
Area	I_Offrad	11	CLC2018 codes 121, 211, 212, 213, 221, 222, 223, 231, 241, 242, 243, 244, 123, 124, 521, 522,523, 131, 132, 133	CAMS-REG-APv3.1
Area	J_Waste	9	E-PRTR information combined with CLC2018 polygons.	CAMS-REG-APv3.1
Area	K_AgriLiveStock	10	CLC2018 codes 211, 212, 213, 221, 222, 223, 231, 241, 242, 243, 244	CAMS-REG-APv3.1
Area	L_AgriOther	10		CAMS-REG-APv3.1

Supplement S2—Meteorological and Air Quality monitoring stations

Table S2-1. Stations by the DWD used in wind field assimilation of TAPM runs.

DWD Station Code	UTM Zone 32N Easting	UTM Zone 32N Northing
DWD_Hamburg	565337	5943161
DWD_Hannover	546054	5812908
DWD_Bremen	486451	5877295
DWD_Cuxhaven	480655	5969242
DWD_Kiel	582485	6039856
DWD_Schwerin	657803	5946395
DWD_Luechow	643524	5871337
DWD_Hamburg	565337	5943161

Table S2-2. Hamburger Luftmessnetz (HaLM) measurement stations with type of stations and measured pollutants.

Station Code	Station Name	Station Type	Altitude	Lon	Lat	Pollutants Measured
13ST	Sternschanze	Urban	3.5	9.96834	53.56449	NO_2, O_3, PM_{10}, $PM_{2.5}$, SO_2
17SM	Stresemannstrasse	Traffic	1.5	9.957387	53.56086	NO_2, O_3, PM_{10}
20VE	Veddel	Urban	3.5	10.02197	53.52291	NO_2, PM_{10}, $PM_{2.5}$, SO_2
21BI	Billbrook	Urban	3.5	10.08214	53.52943	NO_2, PM_{10}, SO_2
27TA	Tatenberg	Urban	3.5	10.08365	53.48814	NO_2, O_3
51BF	Bramfeld	Urban	3.5	10.1105	53.63089	NO_2, O_3
54BL	Blankenese	Background	3.5	9.786191	53.56806	NO_2, O_3, SO_2
52NG	Neugraben	Background	3.5	9.857199	53.48098	NO_2, O_3
61WB	Wilhelmsburg	Urban	3.5	9.990543	53.50792	NO_2, PM_{10}, $PM_{2.5}$, SO_2
64KS	Kielerstrasse	Traffic	1.5	9.944616	53.56437	NO_2, $PM_{2.5}$
68HB	Habichtstrasse	Traffic	1.5	10.05372	53.59235	NO_2, PM_{10}
70MB	MaxBrauerAllee	Traffic	1.5	9.943065	53.55573	NO_2, PM_{10}
72FI	Finkenwerder West	Urban / Industry	3.5	9.844191	53.53622	NO_2
73FW	Finkenwerder Airbus	Urban / Industry	3.5	9.831633	53.53136	NO_2, PM_{10}
74BT	Billstedt	Urban	3.5	10.10286	53.53851	NO_2, PM_{10}
80KT	AltonaElbhang	Urban	3.5	9.944915	53.54524	NO_2, O_3, PM_{10}, SO_2

Supplement S3—Diurnal activity profile for weekdays

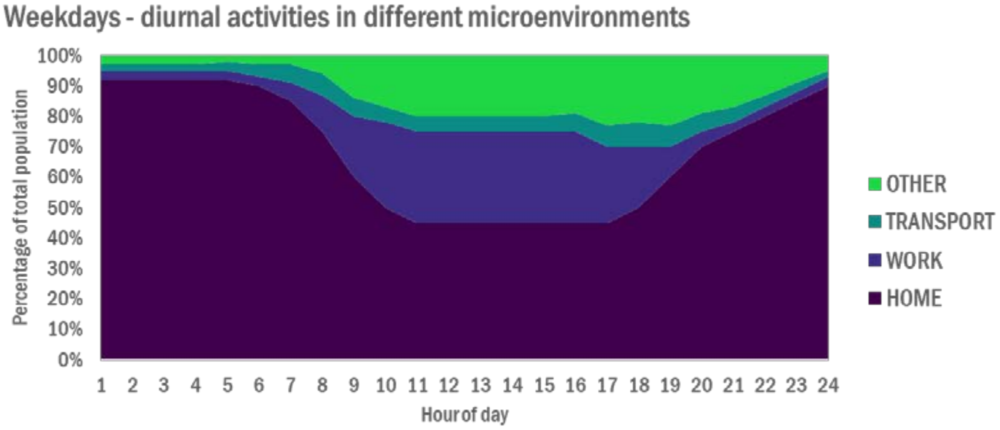


Figure 3. Diurnal activity profile for weekdays as derived from (Ramacher et al. 2019..)

Supplement S4—Indoor infiltration factors for different microenvironments

While there is a solid foundation for experimentally derived infiltration factors for PM2.5 in almost all microenvironments and modes of transport (S4-1), there are major knowledge gaps in terms of infiltration factors for NO2 (S4-2), especially in terms of different modes of transport. Thus, we made reasonable assumptions for missing values based on existing literature values. To identify the uncertainty that arises from the chosen infiltration factors for different modes of transport, we performed a sensitivity analysis in chapter 3.4 in the manuscript.

Table S4-1. Infiltration ratios for $PM_{2.5}$ in different microenvironments from literature

Microenvironment	Min	Mean	Max	Location	Season	Reference
Transport	-	1	-	London	annual	Smith et al. (2016), Singh et al. (2020)
Car	0.2	-	0.9	Hangzhou	winter	Tong et al. (2019)
Car	0.6	0.92	1	Los Angeles	spring	Fujita et al. (2014)
Car	0.3	-	0.98	North Raleigh	summer	Jiao and Frey (2013)
Car	0.43	-	0.99	NA	NA	Liu and Frey (2011)
Bus	-	0.91	-	Nanjing	annual	Shen and Gao (2019)
Subway		0.37		Beijing	spring	Jia et al. (2018)
Subway	0.56	-	0.66	Hongkong	winter	Li et al. (2018)
Subway	-	0.94	-	Taipei	NA	Shen and Gao (2019)
Subway	-	0.81	-	Seoul	NA	Shen and Gao (2019)
Subway	-	0.82	-	Los Angeles	NA	Shen and Gao (2019)
Subway	-	0.18	-	Naples	NA	Shen and Gao (2019)
Subway	-	0.79	-	Singapore	NA	Shen and Gao (2019)
Buildings	0.37	0.57	0.7	Helsinki	annual	Soares et al. (2014)
Residential	0.35	0.56	0.86	London	annual	Smith et al. (2016)
Residential	0.42	0.59	0.76	Europe	annual	Hänninen et al. (2004)
Work	0.23	0.47	0.71	Europe	annual	Hänninen et al. (2004)
Residential	-	0.7	-	Athens. Greece	Winter	Hänninen et al. 2011
Residential	-	0.63	-	Basle. Switzerland	Winter	Hänninen et al. 2011
Residential	-	0.59	-	Helsinki. Finland	Winter	Hänninen et al. 2011
Residential	-	0.61	-	Prague. Czech	Summer	Hänninen et al. 2011
Residential	-	0.53	-	Florence	Winter	Hänninen et al. 2011
Residential	-	0.7	-	Riverside. USA	-	Ozkaynak et al. (1993)*
Residential	-	0.56	-	Riverside. USA	-	Ozkaynak et al. (1993)*
Residential	-	0.62	-	Chongju. Korea	-	Lee et al. (1997)*
Residential	-	0.66	-	Birmingham. USA	-	Lachenmyer and Hidy (2000)*
Residential	-	0.35	-	Baltimore. USA	-	Landis et al. (2001)*
Residential	-	0.7	-	Boston, USA	-	Long et al. (2001)*

Residential	-	0.48	-	Seven cities. USA	-	Wallace et al. (2003)*
Residential	-	0.45	-	North Carolina	-	Williams et al. (2003)*
Residential	-	0.51	-	Three cities. USA	-	Reff et al. (2005)*
Residential	-	0.55	-	North Carolina. USA	-	Wallace and Williams (2005)*
Residential	-	0.48	-	L.A. USA	-	Sarnat et al. (2006)*
Residential	-	0.51	-	Houston, USA	-	Meng et al. (2007)*
Residential	-	0.66	-	L.A. County, USA	-	Meng et al. (2007)*
Residential	-	0.65	-	Elizabeth, USA	-	Meng et al. (2007)*
Residential	-	0.51	-	Helsinki, Finland	-	Hoek et al. (2008)*
Residential	-	0.3	-	Athens, Greece	-	Hoek et al. (2008)*
Residential	-	0.38	-	Amsterdam, NL	-	Hoek et al. (2008)*
Residential	-	0.37	-	Birmingham, UK	-	Hoek et al. (2008)*
Residential	-	0.63	-	Three Cities, USA	-	Meng et al. (2009)*
Residential	-	0.72	-	Three Cities, USA	-	Meng et al. (2009)*

* Adapted from Chen and Zhao (20.11)

Table S4-2: Infiltration ratios for $PM_{2.5}$ in different microenvironments from literature

Microenvironment	Min	Mean	Max	Location	Season	Reference
Car	0.91	0.99	1	Los Angeles	spring	Fujita et al. (2014)
Building	0.38	-	0.45	Los Angeles	winter	Hazlehurst et al. (2018)
Building	0.27	-	0.53	Los Angeles	summer	Hazlehurst et al. (2018)
Building	-	0.76	-	Europe	annual	Kousa et al. (2002)
Building	0.11	0.31	0.59	London	annual	Smith et al. (2016)
Building	0.3	0.545	0.66	Switzerland	annual	Meier et al. (2015)
Building	0.88	-	1	France	annual	Blondeau et al. (2005)
School	-	0.9	-	average	annual	Salonen et al. (2019)
Office	-	0.9	-	average	annual	Salonen et al. (2019)
Buildings	-	0.8	-	Seoul	winter	Bae et al. (2004)
Buildings	0.73	0.8	0.9	UK	annual	Challoner and Gill (2014)

Supplement S5—Evaluation of modeled NO_2 and $PM_{2.5}$ concentrations.

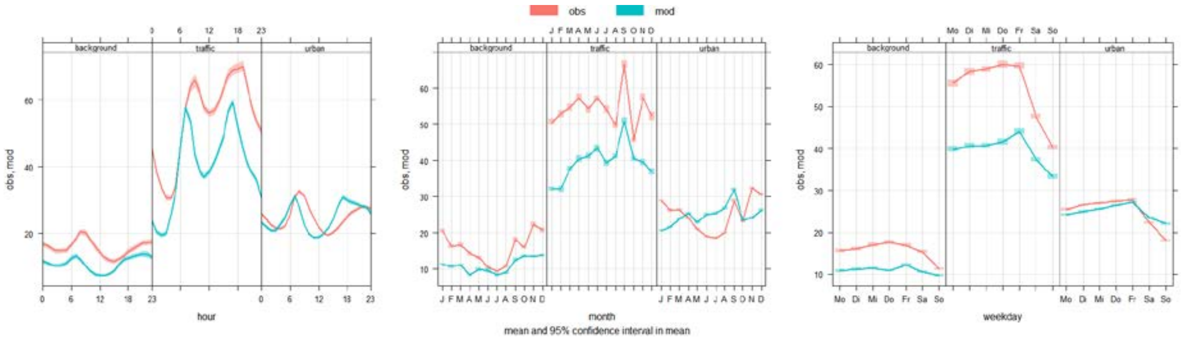


Figure S5-1: Hourly, monthly and daily variation of modelled versus measured hourly NO_2 concentrations averaged by background, traffic and urban stations.

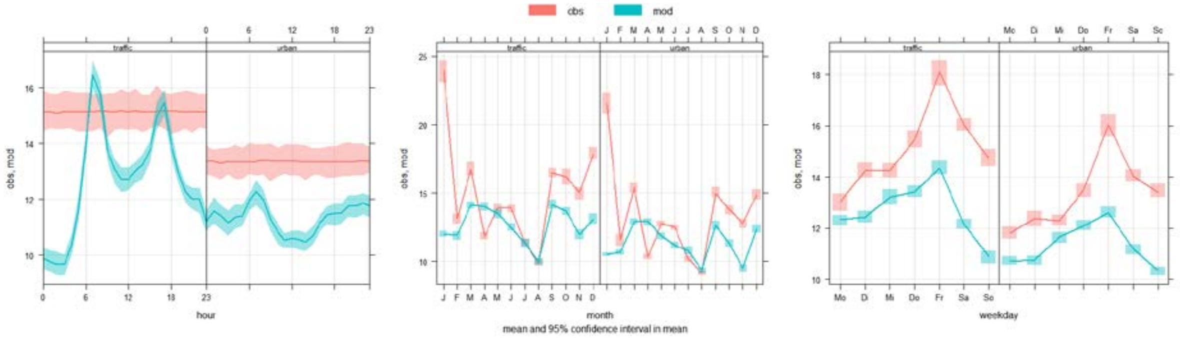
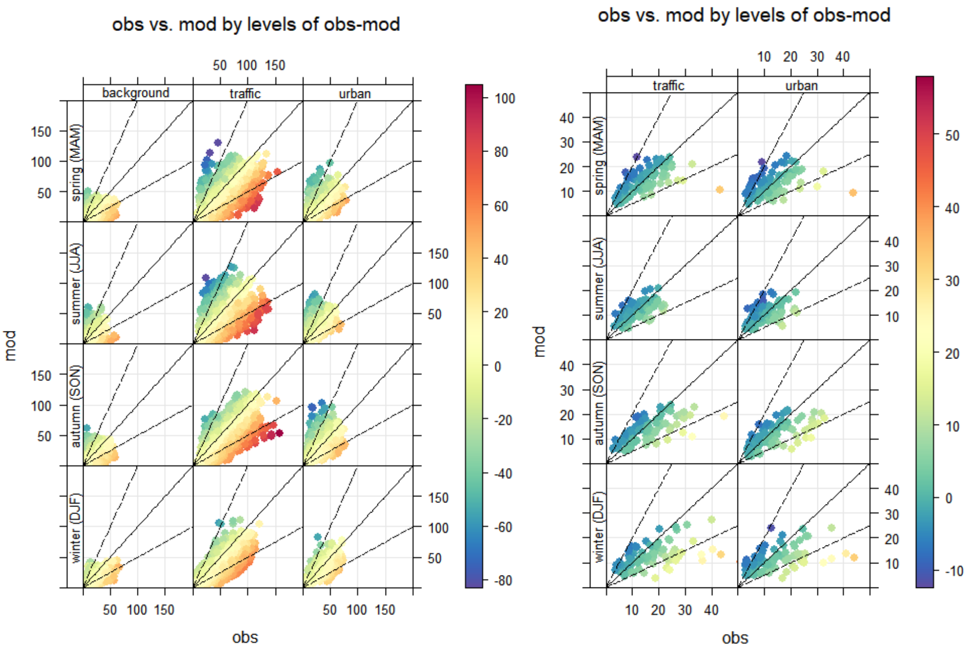


Figure S5-2: Hourly, monthly and daily variation of hourly modelled versus daily measured $PM_{2.5}$ concentrations averaged by traffic and urban stations.



(a) (b)

Figure S5-3: Scatter plots of measured vs. modelled (**a**) hourly NO_2 and (**b**) daily $PM_{2.5}$ concentrations by type of station (urban background, traffic, urban) and season (from top to bottom: spring, summer, autumn, winter), showing the difference of measurement and model color coded.

Supplement S6 – Modeled NO2 and PM2.5 exposure

Table S6-1: Total calculated exposure to NO2 and PM2.5 of all approaches and in all microenvironments. Additionally the relative contribution of microenvironments in the *dynamic transport approach* is shown.

	Total Exposure			**relative contributions of *dynamic transport approach* environments to**	
	Static approach	***Dynamic approach***	***Dynamic transport approach***	**total exposure in *dynamic transport approach***	**transport exposure in *dynamic transport approach***
			NO_2		
Total	2.22E+11	2.51E+11	2.50E+11	100%	-
Home		1.52E+11	1.52E+11	61%	-
Work		4.39E+10	4.39E+10	17%	-
Other		2.96E+10	2.96E+10	12%	-
Transport		2.52E+10	2.42E+10	10%	100%
Walking			6.18E+09	2.5%	26%
Bicycles			3.67E+09	1.5%	15%
Motorcars			9.75E+09	3.9%	40%
Buses			2.14E+09	0.9%	9%
Subway trains			1.22E+09	0.5%	5%
Suburban trains			9.91E+08	0.4%	4%
Regional trains			2.88E+08	0.1%	1%
			$PM_{2.5}$		
Total	8.71E+10	1.05E+11	1.03E+11	100%	-
Home		5.96E+10	5.96E+10	58%	-
Work		1.66E+10	1.66E+10	16%	-
Other		1.69E+10	1.69E+10	16%	-
Transport		1.20E+10	1.04E+10	10%	100%
Walking			3.11E+09	3%	30%
Bicycles			1.77E+09	1.7%	17%
Motorcars			3.41E+09	3.3%	33%
Buses			9.04E+08	0.9%	9%
Subway Train			6.03E+08	0.6%	6%
Suburban train			4.60E+08	0.4%	4%
Regional train			9.97E+07	0.1%	1%

S6.1 Spatiotemporal variability of simulated total exposure

Besides spatial differences, there are differences in terms of diurnal variability in total exposure. Figure 10 shows the diurnal variation of hourly total exposure averaged over one year for the *dynamic transport* approach versus the *static* and *dynamic approaches* in selected areas. While the difference between the static and dynamic approaches is significant, the differences in total exposure between the dynamic approaches is very small. Nevertheless, in both cases the differences are highest during periods of high transport activity in the morning and evening rush hours. During the night, most people are at home and the emissions by traffic go down, leading to less concentrations and activity. Thus, the exposure calculation is strongly dependent on accurate time profiles for temporal distribution of emissions and in modeling of population dynamics.

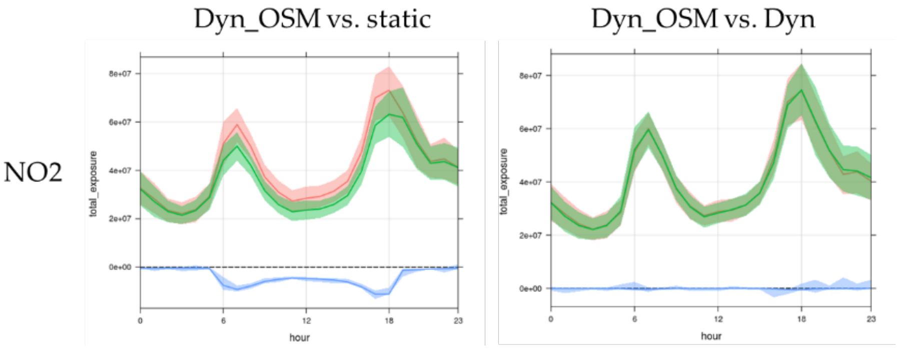


Figure S6-1: Diurnal variation of hourly total exposure to NO_2 averaged over all hourly values for 2016 in the Hamburg area. (**a**) Shows differences between the *dynamic transport approach* and the *static approach*, (**b**) shows differences between the *dynamic transport approach* and the *dynamic approach.*

Supplement S7—Sensitivity of modeled NO_2 and $PM_{2.5}$ exposure

To investigate on uncertainties due to simulated pollutant concentrations in this study, we compared simulated NO_2 and $PM_{2.5}$ concentrations with measurements (section 3.1). The results show highest underestimations at measurement stations close to roads with NMB of up to -35% for NO_2 and −20% for $PM_{2.5}$. To quantify the impact of this underestimation on uncertainty in exposure estimates, we compared the reference emission scenario as defined in section 3.1, with the original lower traffic emissions inventory (minimum scenario). Additionally we created a maximum emissions scenario by scaling the reference scenario with a factor derived from maximum calculated annual NMB. Thus, we applied a factor of 1.4 for NO_2 and a factor of 1.3 for $PM_{2.5}$ to the total road traffic emissions inventory to create a maximum emissions scenario. We applied the minimum and maximum scenarios in our modeling chain to identify minimum and maximum exposure compared to the reference exposure to NO_2 and $PM_{2.5}$ in all microenvironments. The results of the maximum and minimum emissions sensitivity runs (Figure S7-1), reveal relative changes in total exposure and PWE to NO_2 of −9% and +6% for all approaches, and of −1% and +1% for $PM_{2.5}$ respectively. In the transport environments of the *dynamic transport approach* there are uncertainties of −12% to +7% for approach in terms of NO_2 and −2% to +1% for $PM_{2.5}$. Thus, the calculated exposure values show in general a higher sensitivity to changing local NO_2 emissions from road transport, while $PM_{2.5}$ exposure is less sensitive. Nevertheless, for $PM_{2.5}$ the PWE in the *dynamic transport approach* environments Walking, and Buses might rise to values close to or above the WHO AQG limit value for annual $PM_{2.5}$ concentrations of 10 ug/m.

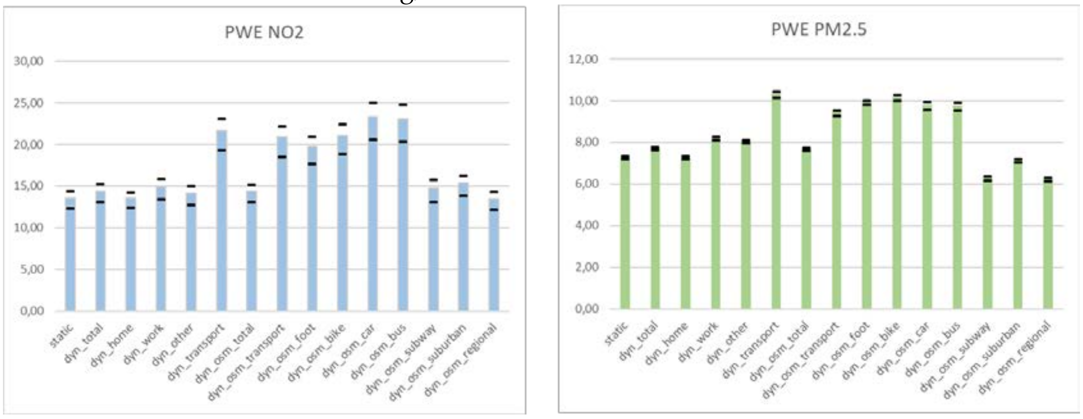


Figure S7-1: Calculated PWE to NO_2 and $PM_{2.5}$ as well as uncertainties in all environments and approaches due to a minimum and a maximum road traffic emission scenario.

Table S7-1: Applied infiltration factors for transport environments in the dynamic_OSM approach and their impacts on total exposure as well as PWE to NO_2 and $PM_{2.5}$ calculations.

		F_{inf}		Exposure change NO_2/$PM_{2.5}$	
NO_2	Min	Ref	Max	Min	Max
Walking	1	1	1	0%	0%
Bicycles	1	1	1	0%	0%
Motorcars	0.7	0.9	1	−22%	11%
Buses	0.7	0.9	1	−22%	11%
Subway trains	0.4	0.6	0.8	−33%	33%
Suburban trains	0.5	0.7	0.9	−29%	29%
Regional trains	0.4	0.6	0.8	−33%	33%
Transport	-	-	-	−14%	+8%
Total	-	-	-	−1%	+1%
$PM_{2.5}$	**Min**	**Ref**	**Max**	**Min**	**Max**
Walking	1	1	1	0%	0%
Bicycles	1	1	1	0%	0%
Motorcars	0.5/0.6	0.7/0.8	0.9/1	−27%	+27%
Buses	0.7	0.9	1	−22%	+11%
Subway trains	0.5	0.7	0.9	−29%	+29%

Suburban trains	0.5	0.7	0.8	−29%	+29%
Regional trains	0.4	0.6	0.8	−33%	+33%
Transport	-	-	-	−14%	+13%
Total	-	-	-	−1%	+1%

The calculated impacts of minimum and maximum Finf in different showed linear de- and increases in calculated exposure values. The impact on exposure averaged over all modes of transport is 14% to +8% for both, NO_2 and $PM_{2.5}$. In terms of impact on total exposure in the *dynamic transport approach* the impact is +/−1%. Thus, when analyzing each mode of transport separately, the impact of Finf is highly sensitive. Moreover, the maximum range of uncertainty in the *dynamic transport approach* environments Motorcars and Buses is exceeding the annual WHO AQG limit value of 10 ug/m^3 for $PM_{2.5}$. When analyzing the total exposure of all environments, the impact of Finf for different modes of transport is low due to the relative high contribution of other microenvironments to the total exposure. Nevertheless, in future studies it is desirable to apply Finf for different indoor and transport environments, which represent city-specific building infrastructure and different air-intake or ventilation techniques in buildings, car cabins or inside of buses and trains. This will increase the robustness and benefit of exposure estimates in general and in different transport environments.

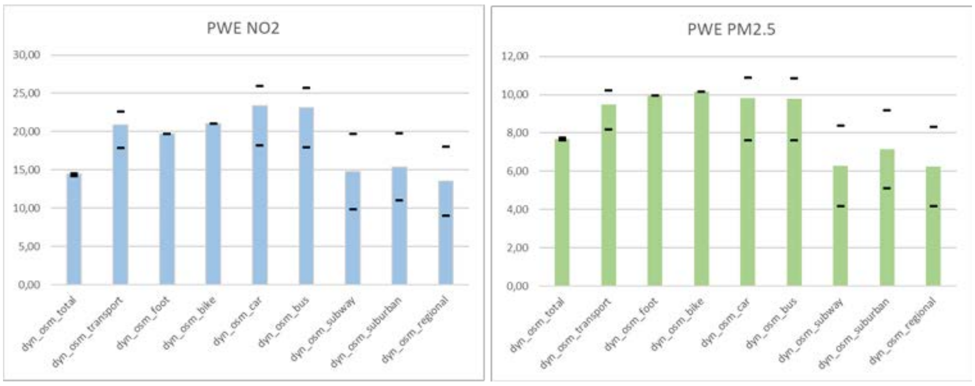
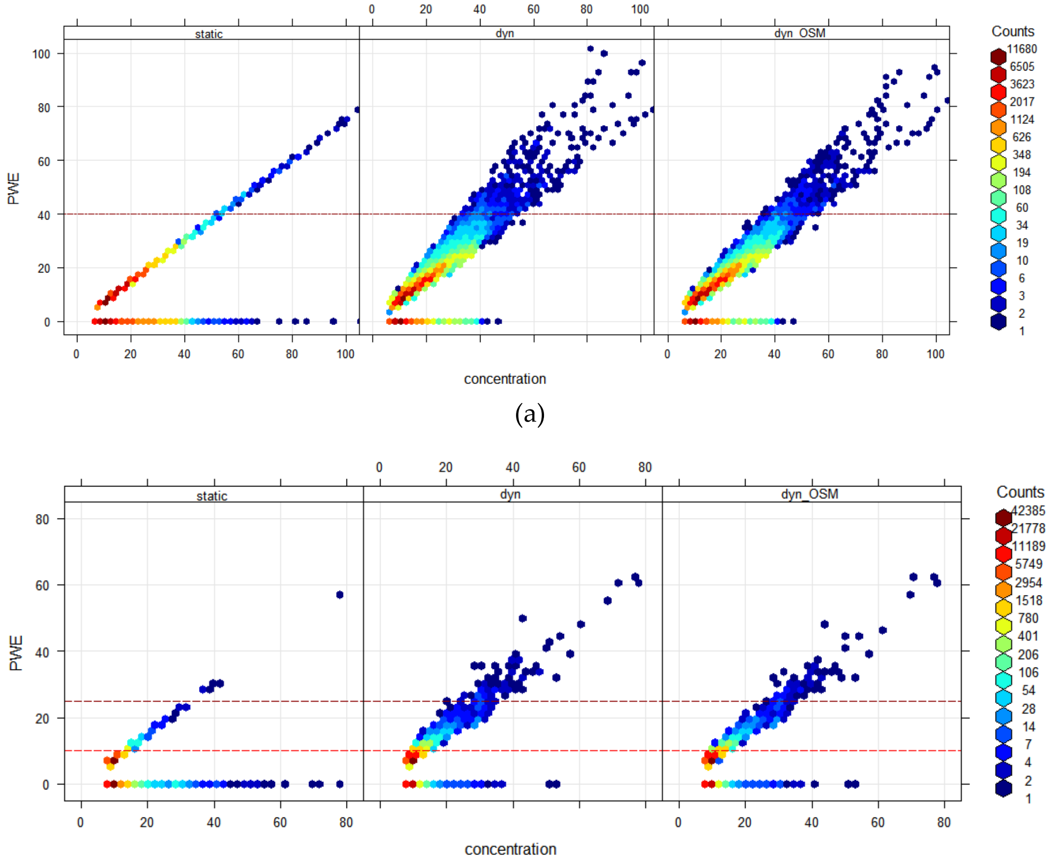


Figure S7-2: Calculated PWE to NO_2 and $PM_{2.5}$ as well as uncertainties in all environments and approaches due to a minimum and a maximum road traffic emission scenario.

S8 Population weighted exposures

Figure S8-1 shows scatter plots of annual total NO_2 and $PM_{2.5}$ exposure grid cell values plotted against the grid cell values of annually averaged NO_2 and $PM_{2.5}$ concentrations for all approaches. This allows for a combined analysis of PWE and concentrations, revealing values with high concentrations but low exposure. It becomes evident, that for both pollutants the PWE is less affected by high concentration values than the dynamic approaches. This leads to higher and more PWE concentrations and finally to more exceedances of the annual limit values. While in the *static approach* the limit value of 40 µg/m³ is exceeded in 190 grid cells, in the *dynamic approach* there 483 and in the *dynamic transport approach* there are 427 exceedances modelled. For PWE $PM_{2.5}$ there are 6 exceedances of the EU limit value of 25 µg/m³ for the static approach, 99 for the *dynamic approach* and 86 for the *dynamic transport approach*, respectively. Looking at the WHO annual limit value of 10 µg/m³ $PM_{2.5}$, we modelled 663 exceedances for the *static approach*, 3184 for the *dynamic approach* and 2631 for the *dynamic transport approach*. Thus, the severity of limit value exceedances is much more evident in both dynamic approaches.

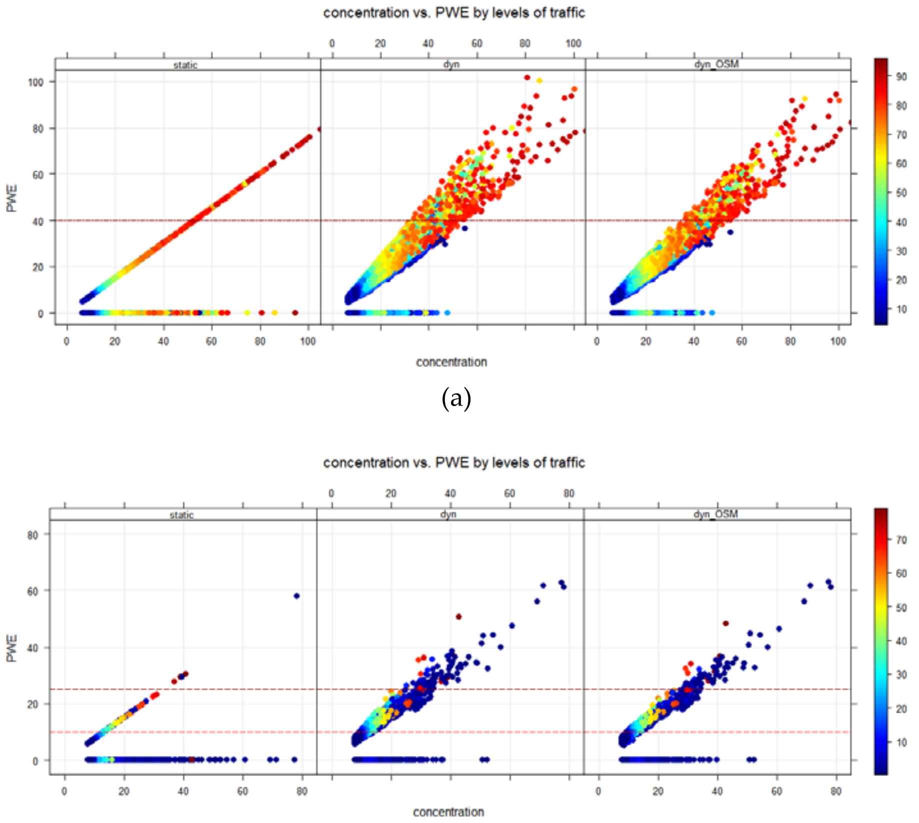


(a)

(b)

Figure S8-1: Simulated concentrations versus PWE grid cell values of NO_2 (a) and $PM_{2.5}$ (b). The color code shows counts for pairs of value ranges.

To analyze the impact of road transport to simulated PWE values, we show the levels of relative traffic contribution to each PWE grid cell value in Figure S8-2. For NO_2 higher concentrations and PWE are mostly going along with higher contributions of traffic, which can reach up to 96% and generally influence the exceedances of limit values by a minimum of 50%. For $PM_{2.5}$, the impact of road traffic related PWE can reach a maximum of 77% but is generally lower, with average contributions 8%. Nevertheless, exceedances of the WHO limit value can be influenced by 10–70% (30% in average) of road traffic contribution. Besides some discordant values, the EU limit value exceedances of $PM_{2.5}$ PWE are only affected by a maximum of 10% road traffic contribution. Highest contributions for $PM_{2.5}$ are mostly by industrial point sources at specific locations, which congregate with regional background concentrations.



(a)

(b)

Figure S8-2: Simulated concentrations and PWE grid cell values of NO_2 (a) and $PM_{2.5}$ (b). The color code shows the relative contribution of road traffic related PWE [%].

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