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SPATIAL VARIATION IN THE AIR POLLUTION EMISSION-TO-EXPOSURE RELATIONSHIPS

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ABSTRACT
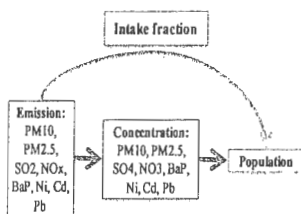
Cost-effective mitigation of the health damage caused by urban air pollution requires knowledge about the relative location, types, and magnitude of emission sources with respect to population exposure. In this study, we estimated the emission-to-exposure relationship using intake fraction (iF) for particulate matter (PM), nitrogen oxides (NO_x), sulfur dioxide (SO_2), benzo [a] pyrene (BaP), nickel (Ni), cadmium (Cd), and lead (Pb), in Warsaw, Poland. Air pollution concentrations were estimated for 2005 using the CALPUFF dispersion model. The resulting air pollution concentration fields were combined with population data to predict the contribution of each individual source to population exposure. The mean iFs between pollutants and source categories varied from 0.01 per million for SO_2, originating from high stack point sources, to 51 per million for PM_{10} derived from road dust[m1]. For primary $PM_{2.5}$, the difference between minimum and maximum intake fraction was 25 fold for the mobile sources. These results indicate that spatially targeted emission reduction policies inside an urban area could improve population health more than a general reduction of emissions from all the sources.

KEYWORDS

Dispersion model; particulate matter; intake fraction; exposure.

1. Introduction

Air pollution causes adverse health effects worldwide. In middle and high income countries, urban air pollution is among the 10 leading risk factors for mortality [1]. Specifically, fine particulate matter ($PM_{2.5}$, particulate matter with aerodynamic diameter less than 2.5 micrometer) has been estimated to cause hundreds of thousands of premature deaths worldwide annually[2, 3]. Other air pollutants that have been associated with adverse health effects include e.g. ozone[4], nitrogen dioxide[4], and heavy metals[5].

Cost-effective mitigation of the adverse health effects of air pollution requires understanding the types and locations of sources and their relative impact on the exposed population. The most common approach to estimate this emission-to-exposure relationship is to predict the movement of the air pollutant from the source with an atmospheric dispersion model and then combine the air pollution concentration fields with population data. Many studies use source-receptor matrixes to summarize emission-to-concentration data in cost-effectiveness models[6, 7].

Intake fraction is defined as an *"integrated incremental intake of a pollutant released from a source category and summed over all exposed individuals"* [8]. For air pollutants, an iF of one per million means that from every kilogram of pollutant emitted to the air, one milligram is inhaled. This concept allows one to summarize and easily compare the population exposure potential of different source emissions.

Several exposure studies have used the iF concept to compare and illustrate exposure differences between emission sources, height, location, and other parameters[9-19]. In an analysis of the iF literature, Humbert et al. derived a set of differing iFs for primary and secondary particulate matter (PM) in urban, rural and remote settings, which illustrate the exposure potential variability between pollutant types and location of emissions[20].

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Factors affecting emission-to-exposure relationships, such as population density near the source, also vary greatly inside urban areas and this variability could translate into significant iF variability within a city. For example, in Boston, Massachusetts, USA, Greco et al. found that minimum and maximum iF for mobile sources primary $PM_{2.5}$ emissions were 0.8 and 53 per million, respectively[10]. Another study from Canada reported that the iF for wood burning $PM_{2.5}$ emissions ranged from 6.6. to 24 per million, respectively for the city of Vancouver, Canada[14].

The high iF variability inside an urban area indicates that cost-effective mitigation of air pollution could benefit from more detailed exposure assessment, where the iF differences between individual sources and areas are taken into account. However, only four previous studies have examined and reported intra-urban iF variability[10, 12, 14, 18]. All of these studies estimated iF variability for primary $PM_{2.5}$ and assessed only three different source categories (mobile, domestic combustion and power plants). The magnitude of the intra-urban variability of iF for other pollutants and source categories is unknown.

The main focus of this study was on (i) estimation of iF variability between emission source categories and (ii) estimation of iF variability within the emission source category (so called intra-urban variability of iF) in Warsaw, Poland. All the calculations were done for the following air pollutants: primary and secondary PM_{10} and $PM_{2.5}$, nitrogen oxides (NOx), sulfur dioxide (SO2), benzo [a] pyrene (BaP), nickel (Ni), cadmium (Cd), and lead (Pb).

2. Material and Methods

The dispersion of air pollutants over the study area was predicted with the CALPUFF atmospheric dispersion model (http://www.src.com/calpuff/calpuff1.htm). Intake fractions were calculated for different sources and source categories by comparing the population-weighted concentration with the emission and population location data.

Emission data

The air pollutants considered in the study (primary and secondary) are shown in Table 1 and the emission rates in Table S1 (supporting material). Area and mobile sources are represented by 1km x 1km elements of spatial discretization of the domain. Emission fields were modeled for a 40 km x 50 km area around the city (Figure S1, supporting material).

Dispersion modeling

CALPUFF computations were performed with year 2005 emission and meteorological datasets in 1 hour time intervals[21]. Annual mean concentrations of different air pollutants were predicted for 563 receptor points located over a 1 km x 1 km grid (Figure S2, supporting material). The results from the CALPUFF calculations were recorded in a database so that the annual average concentration of each pollutant due to each of 6366 sources could be calculated[21].

The comparison of predicted annual averaged concentrations of PM_{10}, NO_x, SO_2, Pb, Ni and Cd with the measured concentrations of these pollutants in different monitoring stations around the city are shown in Table S2 and Figure S3 (supporting material). Other pollutants considered in this article are not measured regularly in Warsaw. Locations of the main monitoring stations are shown in Figure S2 (supplementary material).

For PM_{10}, NO_x and SO_2, the modeled annual average concentrations were within two orders of magnitude from the measured concentrations (Figure S3, supplementary material). For Pb and Cd the modeled concentrations were 89% and 62% higher than the measured one and 62% lower than measured one in one of the measurement stations (Bernardyńska, see Table S2 in supporting material). For Ni and Cd, the modeled concentrations were similar than the measured one in remaning two measurement stations (within -7% to +22%). For Pb the modeled concentration were under predicted 28% and 61% in other three measurement stations.

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Population

Population data were obtained from the European Environment Agency (EEA)[22]. The spatial resolution of the data was 100 m x 100 m and it covers all EU27 countries. The population of the study area was calculated from the EEA population data by taking 1 km buffers around each of the 563 receptor points and then joining the population grids to receptor points. Each 100 m x 100 m population grid was joined only for one receptor point to avoid double counting. Figure S1 (supporting material) shows population density variability for the study area. The population of Warsaw over all the receptor points was 1 790 872. These calculations were done with the ESRI ArcMap version 9.3.

Intake fraction (iF)

Intake fraction represents the fraction of the emissions that is inhaled by the study population. The iF was calculated using the equation:

$$iF_{j,k} = \sum_i (C_{i,j,k} \times Pop_i \times BR)/Q_{j,k} \qquad (2)$$

where iF is the intake fraction; $C_{i,j,k}$ is the predicted concentration increase of air pollutant (g/m^3) in a receptor point i, for pollutant j and for source or sources area k; Pop_i is the population number in receptor point i; BR is the average breathing rate; and Q is the emission strength (g/s). A constant BR of 20 m^3/day/person (~0.0002 m^3/s/person) was used in calculations. The same BR value has been used in a number of previous iF studies[10, 17].

The iFs were estimated separately for (i) four different emission source categories (e.g. mobile source), (ii) for different air pollutants (e.g. $PPM_{2.5}$), and for (iii) each individual emission sources (e.g. individual power plant). For PM_{SO4} and PM_{NO3}, the emission rates were multiplied with the factors of 0.67 and 0.48, respectively, to take into account the differences in chemical composition of inhaled PM versus emitted gases (see Table 1). Factors were based on the relative atomic mass differences between emission and

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concentration. The intake fraction calculations were done with Analytica version 4.4 (http://www.lumina.com/).

3. Results

The intake fractions (iF) for different air pollutants and source categories are presented in Table 2 and iF variability between individual sources in Table S3 (supporting material). The emission-weighted average iF for different pollutants varied between 51 per million (PPM_{10R}) and 0.02 per million (PM_{SO4}). Emission-weighted iF represents the average iF for the pollutant and it takes into account emission strength differences between different emission source categories.

Between the emission sources categories, the highest average iFs were predicted for primary PM and Pb emissions from mobile sources and lowest for secondary aerosols (PM_{SO4} and PM_{NO3} resulting from SO_2 and NO_x emissions, respectively) from high point sources (Table 2). Between the emission source categories, iFs were highest for mobile sources and area sources, and lowest for high point sources. For example, the difference in average iF between mobile sources and high point sources was 53 fold for $PPM_{2.5}$. This means that, on average, $PPM_{2.5}$ emission from mobile sources is 53 times more harmful for the study population than a similar emission from the high point sources, based on an assumption that the exposure-response function for all $PM_{2.5}$ is the same and linear.

For most pollutants, the iFs for mobile source emissions were higher than iFs for emissions from other source categories (Table 2). The exceptions were BaP, Cd, and Ni, for which the iFs were highest for the area sources and only second highest for mobile sources. Overall, the iFs for SO_2, NO_x, BaP, Cd, Ni and Pb were lower than respective iFs for primary PM, except for high point sources for which the iFs for BaP and Cd were higher than respective iFs for primary PM (Table 2).

The iF variability within individual emission sources was substantial (Figures 1, Table S3, supporting material). For NOx emissions from the high point sources, smallest iF for one source was 0.04 per million and highest 3.0 per million. This makes 75 times differences in iF between these two individual emissions sources. For most of the pollutants and source categories the difference between smallest and highest iF was 15 to 50 fold showing the significance of emission location. For the $PPM_{2.5}$, the individual iFs between sources formed a log normal distribution where the mode was between 10 and 20 per million while maximum iFs are around 100 per million (Figure 1). The mapping of these individual iF values over the study area shows that all the source areas with highest iFs are clustered in relatively small area in the center of the city (Figure 2).

4.   Discussion

We have predicted emission-to-exposure relationships for several air pollutants using intake fraction (iF). The iFs for primary PM were higher than the iFs for secondary aerosols, and the mobile sources and area sources had higher iFs than high point sources and other point sources. Several previous iF studies have shown similar results for PM air pollution[11]. The iF range within the emission source categories was over an order of magnitude for most pollutants. Only a few previous studies have examined iF variability within emission source categories before this study and those studies have examined primary $PM_{2.5}$ but not secondary.

Inter-pollutant and source category variability

For PM air pollution, the iF differences between primary and secondary PM, and between the emission source categories in this study were similar to those observed in previous studies[10, 15, 17, 20]. For example, a recent review of iF studies recommended an order of magnitude lower iFs for secondary aerosols than for primary PM^{20}. Also, iF relative iF differences between different emission source categories have been observed in previous studies. For example, Tainio et al. compared iFs between six primary $PM_{2.5}$ emission source

categories in Finland and the highest iFs were predicted for the mobile and area sources (specifically road traffic and domestic wood combustion, respectively)[17].

Non-PM air pollutants have not often been addressed in the iF literature. A search of the Intake Fraction Database (http://www.ktl.fi/expoplatform/) and the ISI Web of Knowledge (http://apps.webofknowledge.com/) revealed only three iF studies for other conservative pollutants [9, 11, 23].

Spadaro and Rabl[23] estimated iFs for Cd, Ni and Pb with a multimedia pathway model using average central-European parameterizations. The resulting iFs were 3.9, 3.9 and 7.1 per million, respectively, for the inhalation pathway while in the present study the average iFs were 12, 20 and 38 per million, respectively. According to Spadaro and Rabl, these iFs are for typical power plant emissions (high stack) and should be multiplied by 3 for typical industrial emissions (area) and by 20 for typical automotive emissions[23]. In the present study, the iF for high point sources was 1.4 for Ni, which is on a similar order of magnitude as the iF of 3.9 reported by Spadaro and Rabl. Taken together, it seems that iF for the inhalation exposure pathway is somewhere between one and a few tens per million for these metals in, what Spadaro and Rabl called typical European conditions. Spadaro and Rabl also estimated exposure through other exposure pathways and for all three metals the ingestion pathway was responsible for a greater proportion of total intake than the inhalation pathway.

For BaP, two previous iF studies with different methodologies have been published[9, 21]. Humbert et al.[11] estimated iF for BaP using the multimedia, multi-pathway model IMPACT North America (version 1.0). The resulting iF was 5.0 per million for urban settings and the maximum estimated iF was 30 per million, very similar to this study's mean estimate of 11 per million and maximum of 64 per million (Table S3, supporting material). Bennett et al. also considered BaP exposure through ingestion and estimated an iF value of 24 per million[9].

Intra-source variability

We estimated the distribution of iF from individual sources within a category as well as between source locations for different pollutants (Table S3, supporting material). For pollutants and sources that have large variability in iF values, for example $PPM_{2.5}$, PPM_{10}, SO_2, and NO_x, it may be more cost effective to target emissions reductions to those specific sources or sources areas that have high iF values. On the other hand, for pollutants (e.g. Cd, Ni, Pb) and source categories where the variability in the distribution of iFs is small, it may be more cost effective to try and reduce emissions across the urban area.

It is likely that population density differences near the emission sources are the main contributor to the variability of iF within a source category[20]. Meteorological conditions and emission height are similar inside the urban area so it is unlikely that these factors would explain the variability. For the power plants, the distance that captures half of the total exposure has been estimated to be several hundreds of kilometers[19] but for low emission height sources emitted in urban centers this distance can be kilometers, or less[10].

The comparison of our findings with the previous studies shows that the iF variation predicted in present study are in the same order of magnitude than the iF variation in previous studies[10, 12, 14, 18]. For mobile sources, Greco et al.[10] estimated iFs for 23 398 road segments inside the Boston, US. The dispersion of primary $PM_{2.5}$ was calculated within 5 km from the road segments. The iFs ranged from 0.8 and 53 per million with a mean of 12 per million. We found slightly higher, but similar values (mean 44 per million, with a range of 4-100 per million) for Warsaw. In our study the exposure was estimated for larger area than 5km x 5km area around the source and it could explain, together with other urban area differences, the slightly higher iF results.

Two previous studies have estimated iF variation for area sources. In Vancouver, Canada, the iF for wood burning was estimated to be 13 per million (geometric mean) with certainty

range of 6.6 to 24 per million (one geometric standard deviation)[14]. The iFs were based on measured concentrations and a land use regression model designed to estimate the spatial variability of wood burning related primary $PM_{2.5}$ in the study area. Lai et al.[12] estimated cumulative population inhalation transfer factors (PITF) for a hypothetical urban area. For outdoor sources they estimated PITF values between 4.4. and 44 per million, depending on the wind speed. The definition of PITF is identical with iF so the results are comparable. Both previously published results are close to the results predicted in present study (Table S3, supporting material).

For high point sources, Wang et al.[18] have estimated iFs for 49 different power plants from six different urban areas in China. The average iF for total suspended particulate matter was 3.0 per million and variability from 0.41 to 17.9. The representative values for in the present study were 0.83, 0.05 and 3.1 per million, respectively ($PPM_{2.5}$, high point sources). Thus, our iFs are approximately an order of magnitude lower than the iFs estimated in Wang et al.[18] study. The Wang et al. study took into account exposure within 50 km from the sources so the one magnitude difference in results is probably due to larger study domain and larger population density.

Uncertainties and limitations

There are a number of uncertainties and limitations related to this study. We will discuss more detailed possible underestimation of population, lack of time-activity data and indoor-outdoor penetration of pollutants.

The population of Warsaw was assumed to be 1.7 million, which approximates the city's official population count. However, Warsaw has a large non-official population that live in the city but are registered in other parts of the country. If the true population of Warsaw is significantly higher, it means that the iFs calculated in the present study underestimate the true exposure by underestimating the amount of people in the study area. The spatial

distribution of this unofficial population may also change the population density of various areas, thus potentially affecting the iF distributions, which have been shown to be strongly related to population density [10, 24, 25]. However, current data does not allow for the assessment of this area.

The other main uncertainty relates to the lack of consideration of time-activity and outdoor-indoor infiltration in the intake estimate. We assumed that the outdoor concentrations of pollutants at people's home addresses represent their exposure. The infiltration of pollutants from outdoors to indoors reduces the exposure to outdoor originated pollutants because only a fraction of the pollutants penetrate indoors. This would bias our iF estimates upward. People also spend time in other locations than their homes, including both more and less polluted microenvironments.

Some previous studies have incorporated time-activity and indoor-outdoor infiltration to their assessments [13, 14]. Loh et al. [13] used three different models to predict iFs for benzene emitted from traffic in Helsinki, Finland. The mean iFs for personal measurement model, spatial time activity model and simple box model were 39 per million, 10 per million and 7 per million, respectively. The highest iF was predicted with the model that took into account benzene concentration in different microenvironments (e.g. home, work, traffic). In the present study, we estimated exposure only based on outdoor concentration of pollutants. The results from Loh et al. [13] suggests that the iF estimates for mobile sources would have been higher also in Warsaw, if the exposure in different microenvironments, especially in traffic, would have been taken into account.

To our knowledge this is the first study that examined intake fractions for many pollutants and sources within a single urban area. Previous studies have reported iF variability only for $PPM_{2.5}$ emissions and for three different source categories (mobile sources, wood burning and power plants) [10, 12, 14, 18]. The iF variability noticed in these studies is in same order of

magnitude as the iF variability observed in present study. This proposes that iF variability could be similar in other urban environments and that generally urban areas would benefit from more spatially refined emissions mitigation plans.

ACKNOWLEDGMENT

SUPPORTING INFORMATION AVAILABLE

Three tables (emission volumes, comparison of modeled air pollution concentrations to measured concentrations, intake fraction variability) and three figures (population density, location of receptor areas, comparison of modeled PM_{10}, NO_x and SO_2 concentrations to measured concentrations).

TABLES and FIGURES

Table 1. Air pollutants considered in the present study. Emissions were inputs to the dispersion model and the concentrations were the outputs of the dispersion model.

Emissions	Concentrations
$PPM_{2.5}$ (primary particulate matter (PPM) with aerodynamic diam. ≤ 2.5 μm)	$PPM_{2.5}$ ($PM_{2.5}$ concentration caused by the $PPM_{2.5}$ emissions)
$PPM_{2.5R}$ ($PPM_{2.5}$ raised by road traffic – secondary emission)	$PPM_{2.5R}$
SO_2 (sulfur dioxide)	SO_2 PM_{SO4} (sulfate ($SO_4^=$) aerosol)
NO_x (nitrogen oxides)	NO_x PM_{NO3} (nitrate (NO_3^-) aerosol)
-	$PM_{2.5} = PPM_{2.5}+PPM_{2.5R}+ PM_{SO4}+ PM_{NO3}$
PPM_{10} (primary particulate matter with aerodynamic diameter ≤ 10 μm)	PPM_{10} (PM_{10} concentration caused by the PPM_{10} emissions)
PPM_{10R} (PPM_{10} raised by road traffic – secondary emission)	PPM_{10R}
-	$PM_{10} = PPM_{10}+PPM_{10R}+ PM_{SO4}+ PM_{NO3}$
BaP (benso [a] pyrene)	BaP
Ni (nickel)	Ni
Cd (cadmium)	Cd
Pb (lead)	Pb

Table 2. Mean intake fraction (iF) (standard deviation) for different pollutants and source categories.

Pollutant	Mobile sources	Area sources	High point sources	Other point sources	All sources
$PPM_{2.5}$	44 (18)	20 (12)	0.83 (1.0)	11 (10)	22
$PPM_{2.5R}$	50 (21)	-	-	-	50
PM_{SO4}	0.39 (0.14)	0.19 (0.080)	0.013 (0.015)	0.085 (0.067)	0.024
PM_{NO3}	0.87 (0.37)	0.60 (0.15)	0.029 (0.030)	0.21 (0.26)	0.36
PPM_{10}	45 (18)	21 (12)	0.72 (1.1)	9.9 (10)	19
PPM_{10R}	51 (21)	-	-	-	51
SO_2	32 (14)	18 (12)	0.70 (1.1)	9.5 (11)	1.8
NO_x	30 (14)	18 (11)	0.63 (1.1)	8.9 (10)	12
BaP	11	20	1.4	4.5	11
Cd	11	21	1.4	4.5	12
Ni	11 (7.0)	20 (12)	-	-	20
Pb	45 (18)	21 (12)	-	-	38

Figure 1. The distribution of individual iFs between primary $PM_{2.5}$ emission sources or source areas.
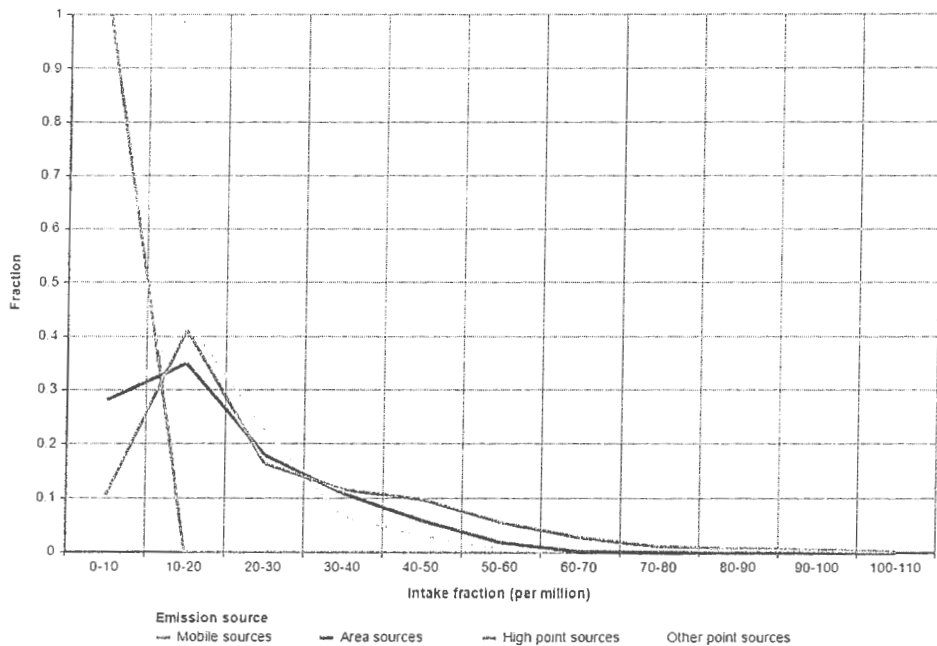
Figure 2. Intake fraction values for PPM$_{2.5}$ emissions due to mobile sources.
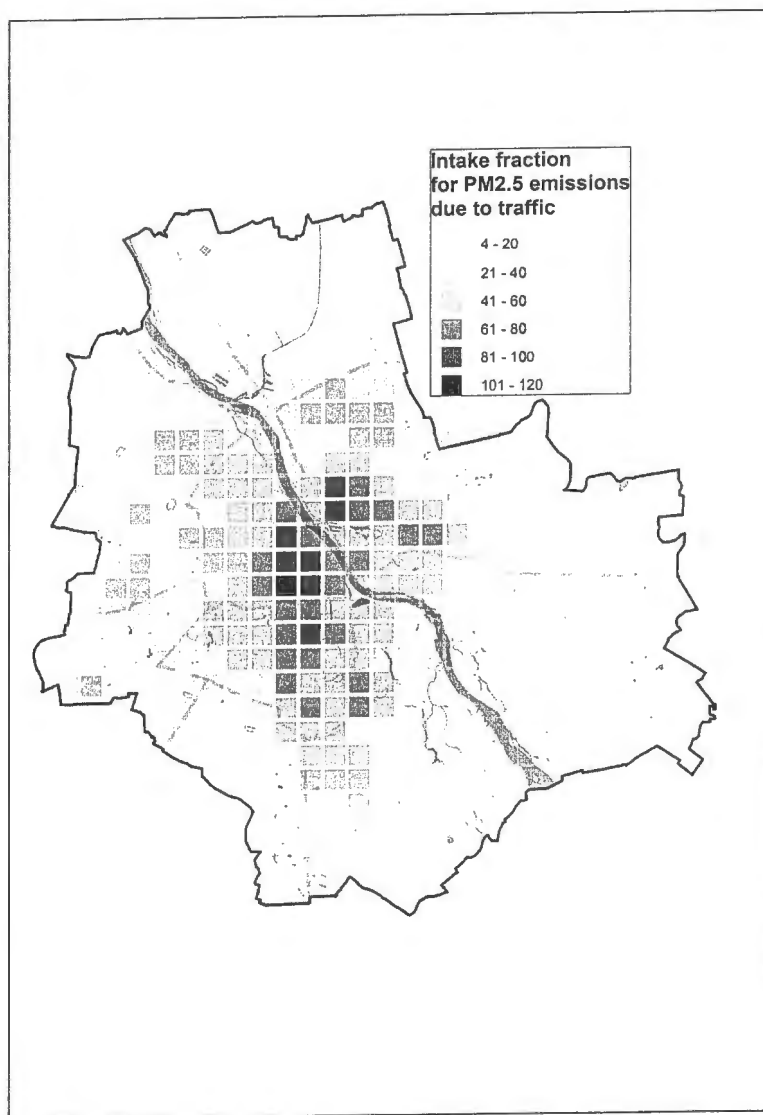
SUPPORTING MATERIALS

Table S1. Emission rates by source category.

	Unit	Mobile sources	Area sources	High point sources	Other point sources
Number of sources	#	1156	877	16	1017
$PPM_{2.5}$	g/s	16	84	7.6	15
$PPM_{2.5R}$	g/s	26	-	-	-
SO_2	g/s	23	69	1328	125
NOx	g/s	325	46	396	60
PPM_{10}	g/s	24	157	21	54
PPM_{10R}	g/s	151	-	-	-
BaP	μg/s	0.045	16	2.0	12
Cd	μg/s	0.20	17	2.0	12
Ni	μg/s	2.0	56	-	-
Pb	μg/s	106	109	-	-

Table S2: Comparison of modeled and measured air pollution concentrations. Measurements are from the year 2005.

No.	Monitoring station	PM_{10} [µg/m³]			NO_x [µg/m³]			SO_2 [µg/m³]		
		Modeled	Measured	Error [%]	Modeled	Measured	Error [%]	Modeled	Measured	Error [%]
1	Białobrzeska	33	29,5	11,9	-	-	-	-	-	-
2	Bednarska	42	34,1	23,2	46,7	56,8	-17,8	-	-	-
3	Komunikacyjna	40	51,7	-22,6	51,2	76,2	-32,8	13,9	9,8	41,8
4	Żelazna	34	32,9	3,3	45,6	36,3	25,6	15,6	9	73,3
5	Krucza	43,2	41,7	3,6	31,3	35,7	-12,3	9,8	9,3	5,4
6	Ursynów	32	32,8	-2,4	27,5	19,2	43,2	9,3	8,8	5,7
7	Nowoursynowska	33,5	42,2	-20,6	23,1	25,6	-9,8	9,5	10,5	-9,5
8	Tołstoja	22	37,2	-40,9	35,2	43,1	-18,3	12,1	11,8	2,5
9	Targówek	31,6	31,9	-0,9	-	-	-	-	-	-
10	Anieli Krzywoń	24	31,3	-23,3	-	-	-	-	-	-
11	Bernardyńska	33	21,2	55,7	-	-	-	10,4	8,7	19,5
12	Bora-	40,5	34,9	16,0	-	-	-	-	-	-
13	Żegańska	27	39,2	-31,1	-	-	-	-	-	-
14	Puszczy Solskiej	-	-	-	26,4	34,1	-22,6	10,6	12,9	-17,8
15	Porajów	-	-	-	19,6	24,1	-18,7	-	-	-
16	Lazurowa	-	-	-	-	-	-	10,1	11,2	- 9,8

Table S2 (cont). Comparison of modeled and measured air pollution concentrations. Measurements are from the year 2005.

No.	Monitoring station	Pb [ng/m³]			Ni [ng/m³]			Cd [ng/m³]		
		Modeled	Measured	Error [%]	Modeled	Measured	Error [%]	Modeled	Measured	Error [%]
1	Bernardyńska	22,7	12	89,2	2,3	6,1	-62,3	0,73	0,45	62,2
2	Żelazna	24,5	34	-27,9	1,8	1,5	20,0	0,65	0,7	-7,1
3	Żegańska	20,2	41	-50,7	3,2	2,8	14,3	1,10	0,9	22,2
4	Anieli Krzywoń	18,3	47	-61,1	-	-	-	-	-	-

Table S3. Intake fraction variation between individual emission sources. Results are shown for:

0, 0.25, 0.5, 0.75 and 1.0 fractiles.

Pollutant	Fractile	Mobile sources	Area sources	High point sources	Other point sources
$PPM_{2.5}$	0.00	4.0	4.0	0.05	1.1
$PPM_{2.5}$	0.25	13	9.7	0.20	10
$PPM_{2.5}$	0.50	20	14	0.97	16
$PPM_{2.5}$	0.75	36	26	1.3	22
$PPM_{2.5}$	1.00	100	63	3.1	67
$PPM_{2.5R}$	0.00	4.6	-	-	-
$PPM_{2.5R}$	0.25	15	-	-	-
$PPM_{2.5R}$	0.50	23	-	-	-
$PPM_{2.5R}$	0.75	42	-	-	-
$PPM_{2.5R}$	1.00	114	-	-	-
PM_{SO4}	0.00	0.047	0.064	0.001	0.034
PM_{SO4}	0.25	0.14	0.11	0.006	0.10
PM_{SO4}	0.50	0.22	0.15	0.023	0.13
PM_{SO4}	0.75	0.37	0.22	0.031	0.18
PM_{SO4}	1.00	0.75	0.44	0.041	0.44
PM_{NO3}	0.00	0.16	0.30	0.002	0.048
PM_{NO3}	0.25	0.41	0.48	0.012	0.27
PM_{NO3}	0.50	0.58	0.63	0.054	0.41
PM_{NO3}	0.75	0.90	0.73	0.065	0.59
PM_{NO3}	1.00	1.6	0.91	0.084	1.6
PPM_{10}	0.00	4.1	4.0	0.051	1.1
PPM_{10}	0.25	13	9.5	0.21	11
PPM_{10}	0.50	20	14	0.99	17
PPM_{10}	0.75	36	26	1.4	22
PPM_{10}	1.00	102	64	3.1	68
PPM_{10R}	0.00	4.6	-	-	-
PPM_{10R}	0.25	15	-	-	-
PPM_{10R}	0.50	23	-	-	-
PPM_{10R}	0.75	42	-	-	-
PPM_{10R}	1.00	115	-	-	-

Table S3(cont). Intake fraction variation between individual emission sources. Results shown for 0, 0.25, 0.5, 0.75 and 1.0 fractiles.

Pollutant	Fractile	Mobile sources	Area sources	High point sources	Other point sources
SO_2	0.00	1.7	3.1	0.05	2.7
SO_2	0.25	6.4	7.3	0.20	10
SO_2	0.50	12	12	0.96	17
SO_2	0.75	27	22	1.4	23
SO_2	1.00	81	63	3.1	66
NO_x	0.00	1.6	2.9	0.042	0.93
NO_x	0.25	6.1	7.6	0.18	9.5
NO_x	0.50	12	13	0.82	15
NO_x	0.75	26	23	1.3	21
NO_x	1.00	79	61	3.0	65
BaP	0.00	4.1	4.0	1.4	2.8
BaP	0.25	4.5	9.2	1.4	3.8
BaP	0.50	7.2	13	1.4	7.4
BaP	0.75	12	24	1.4	12
BaP	1.00	25	64	1.4	12
Cd	0.00	4.1	4.0	1.4	2.8
Cd	0.25	4.5	9.3	1.4	3.8
Cd	0.50	7.4	13	1.4	7.4
Cd	0.75	12	24	1.4	12
Cd	1.00	25	64	1.4	12
Ni	0.00	4.1	4.0	-	-
Ni	0.25	4.5	9.3	-	-
Ni	0.50	7.5	13	-	-
Ni	0.75	12	24	-	-
Ni	1.00	25	64	-	-
Pb	0.00	4.1	4.0	-	-
Pb	0.25	14	9.3	-	-
Pb	0.50	20	13	-	-
Pb	0.75	37	24	-	-
Pb	1.00	102	64	-	-

Figure S1. Population density in the study area. The emission field was for the 40 km x 50 km area shown by the square line. Calculations of *iF* involved only emissions inside the city borders.

Population density
(100 m x 100 m)
0.0 - 0.6
0.7 - 1.4
1.5 - 49.3
49.4 - 92.8
92.9 - 243.6

0   3.75   7.5   15 Kilometers
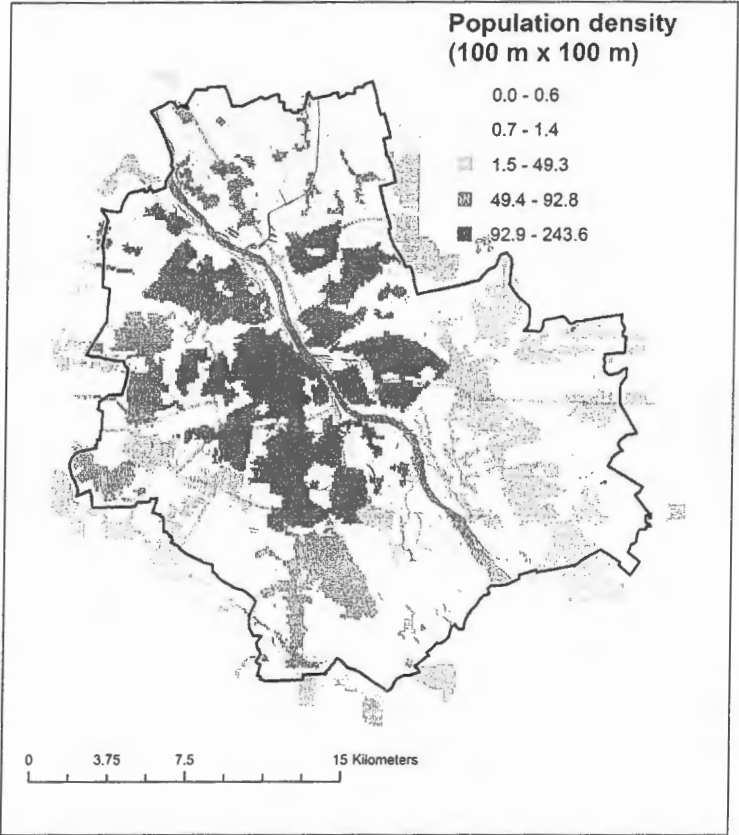
Figure S2. Location of measurement stations and the location of receptor areas.

0   2   4 km
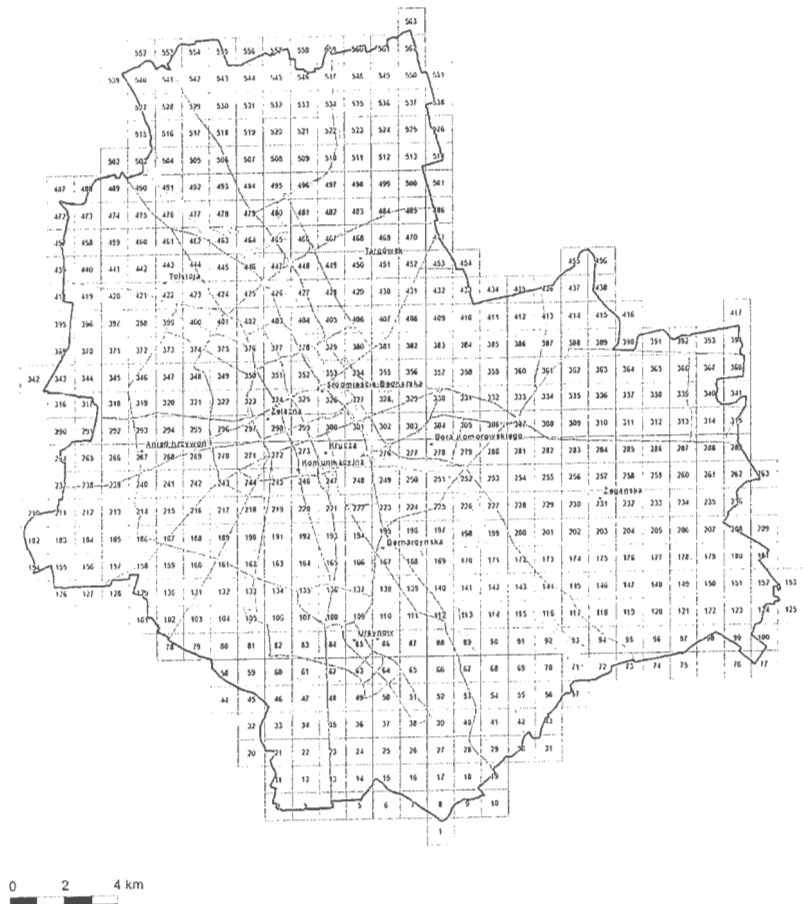
Figure S3. Measured vs. calculated annual average concentrations [µg/m3] in 2005:
a) PM_{10}, b) NO_x, c) SO_2

a)

b)

c)
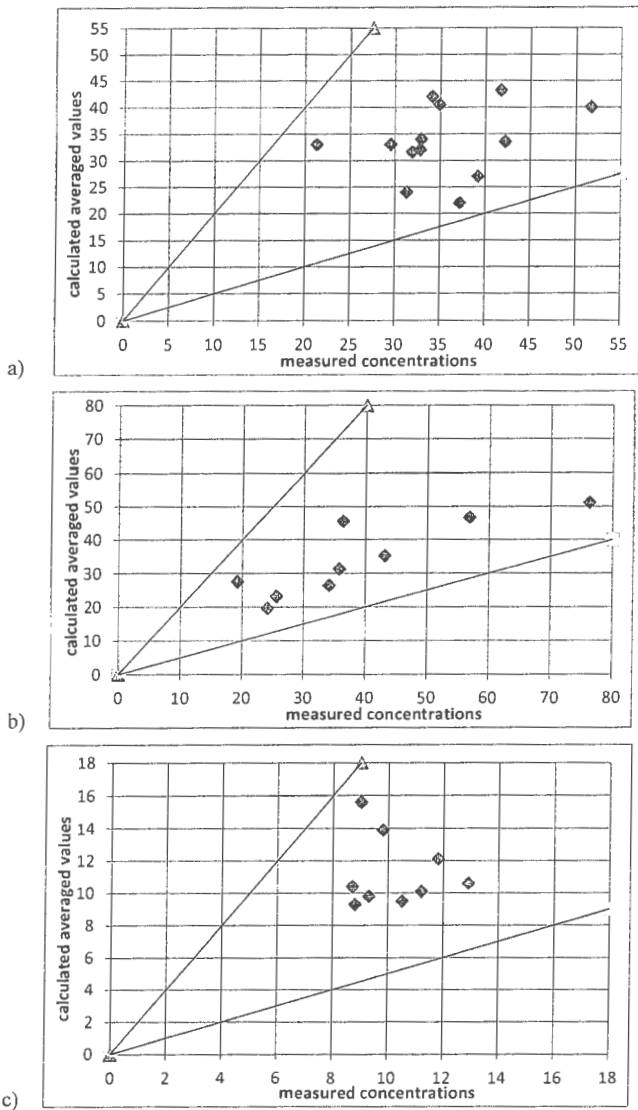
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