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Supplement of

Emission ensemble approach to improve the development of multi-scale emission inventories

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Supplementary material

S1. EU-wide comparisons of CAMS and EMEP vs the ensemble

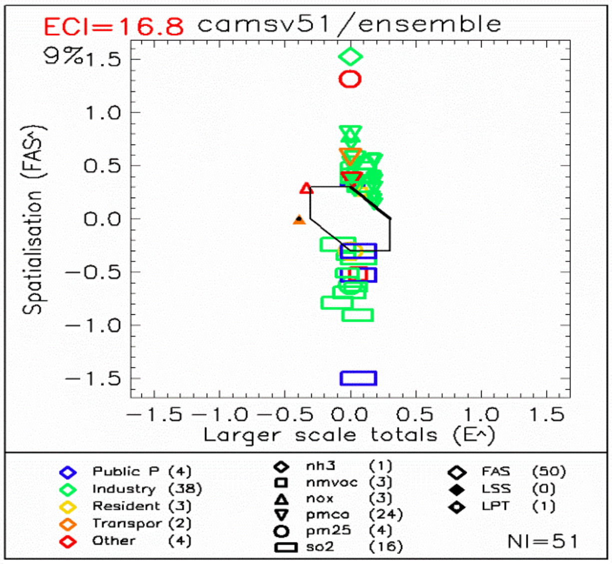

Figure S1: Diamond diagram that isolates the bilateral comparison between CAMS-REG and the ensemble. Symbols and colours are as specified in the legend. Only inconsistencies are displayed. For visualization purposes, we limit the axis to a factor 2 in terms of magnitude (from -2 to 2) and bound the ECI to 100 (e.g. values of ECI larger than 100 are plotted with a value of 2). Numbers within bracket in the bottom legend are the total number of inconsistencies for a given pollutant, sector or type.

For CAMS-REG, the ECI (=16.8) indicates that the largest inconsistency is around a factor of 15 larger than the estimated level of uncertainty. About 9% of the relevant emission points show an inconsistency larger than a factor 2. As indicated by the overview table, these 9% amount to 51 inconsistencies that are almost all related to urban share issues (50), mostly for PM_{co} and SO_2 from the industry sector.

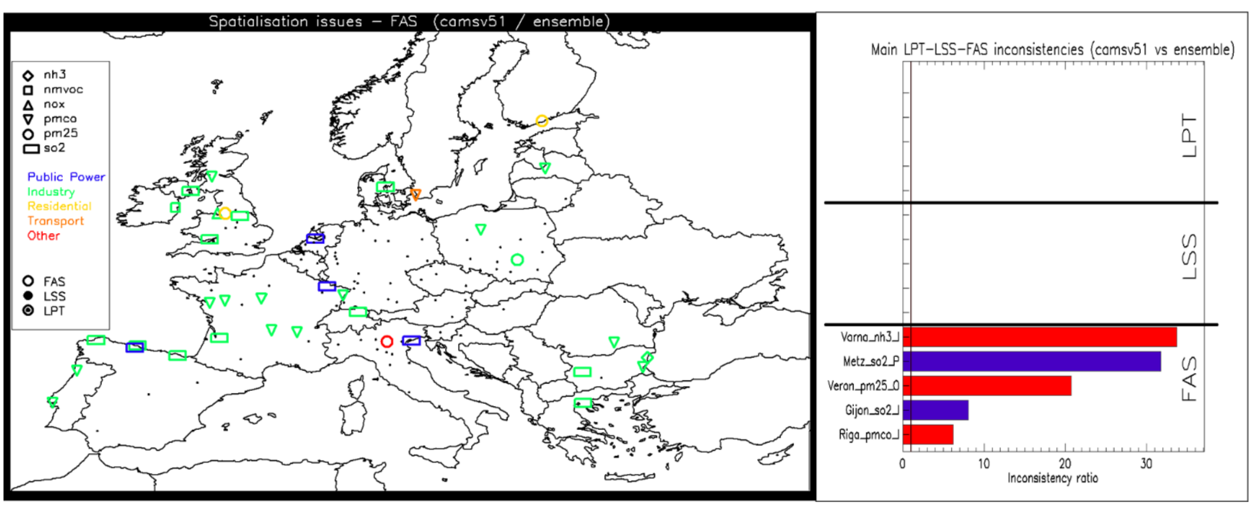

Figure S2: Left: Main inconsistencies spotted at urban scale for CAMS-REG when compared to the ensemble. Only the main spatial inconsistency (FAS) for each city is plotted. See explanation of symbols on the top left of the figure. Right: Major LPT (top 5), LSS (middle 5) and FAS (lower 5) inconsistencies. The two first letters indicate the country code for LSS and LPT whereas the

4 first city letters are given for FAS. Red shading indicates an overestimation and blue shading an underestimation for the EDGAR inventory.

Figure S2 points to the following main issues:

- All major inconsistencies are related to the choice of spatial distribution of the emissions (FAS) and occur in particular in Varna for industrial NH_3, in Metz for SO_2 from power plant and in Verona for PM2.5 from the other sector. These three inconsistencies exceed a factor 20. Note also that these inconsistencies are either over- or under-estimations (red and blue color bars, respectively). In Bulgaria, the largest industrial point source in E-PRTR (68% of the country total) is located near Varna, hence the high emissions there. The large differences among inventories occur due to the proportion of these emissions being placed within the city area (see Figure S8). The same explains the differences for SO_2 in Metz for the power plant sector or for PM2.5 in Verona for the other sector.

- Although of lower importance, inconsistencies are also spotted for industrial PMco emissions in France and are systematic in several cities across the country. The same occur for industrial SO_2 emissions in the UK and in Spain. The diamond plot shows that while PMco has larger estimates in the CAMS-REG inventory, the opposite is true for SO_2. A likely explanation for the differences in SO_2 emissions is that their attribution to point sources is done only for those included in point source reporting (E-PRTR). Smaller sources which are below the threshold for E-PRTR reporting are distributed as diffuse sources to industrial zones (land cover class). This may lead to over-allocation in some urban areas.

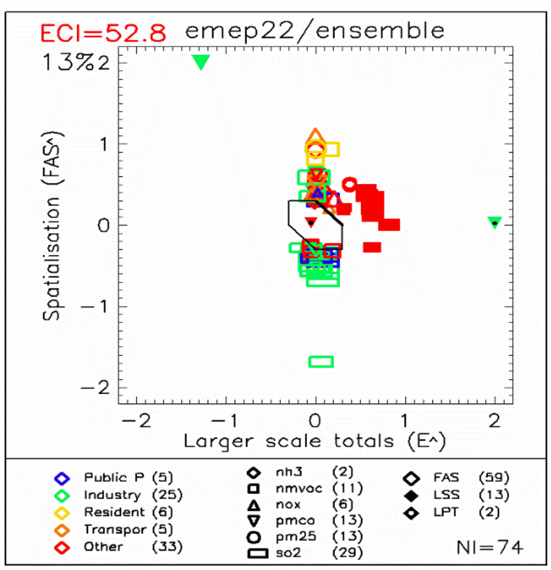

Figure S3: Same as Figure S1 but for EMEP

For EMEP, the ECI (52.8) indicates that the maximum inconsistency is about a factor 50 larger than the estimated level of uncertainty. About 13% of the relevant emission points show an inconsistency. As indicated by the overview table, these 13% amount to 74 inconsistencies that are mostly related to the spatial distribution of the emissions (FAS=59), mostly for SO_2 (29), and in a lesser extent to $PM_{2.5}$ (13), PM_{co} (13) and NMVOC (11) originating from the other sector (33), but also from the industry (25) sectors.

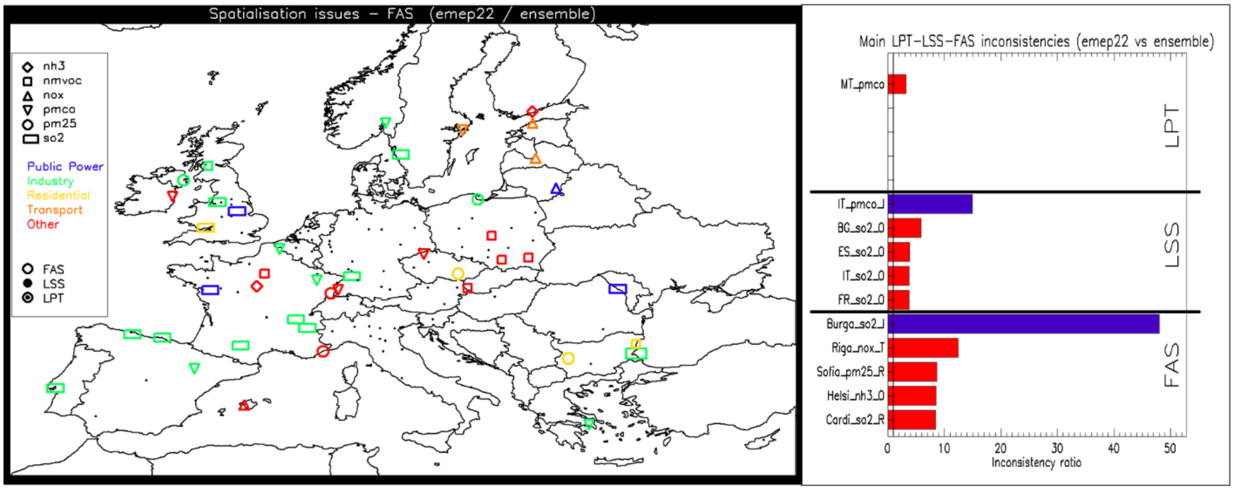

Figure S4: same as **Error! Reference source not found.** but for EMEP

Figure S4 points to the following main issues:

- One inconsistency only is spotted at country total level (LPT) for the PMco industrial emissions in Malta (factor 3). Similarly to what is reported for EDGAR for Malta, the low-emission filter is not efficient to remove these small (not relevant) emissions, given the small size of the country
- A series of inconsistencies are associated with the sectorial share at country level (LSS). The largest is observed for PMco industrial emissions in Malta (factor >30) and add up to the inconsistency at country total level previously highlighted. The same inconsistency, although as underestimation (blue shaded bar in figure S4), occurs in Italy with a factor 15. LSS inconsistencies also occur for SO_2 emissions from the other sector in countries like Bulgaria, Spain and Italy (between a factor 3 and 6)
- Regarding inconsistences related to the spatial distribution of the emissions, one large one (factor >50) is flagged in Burgas for SO_2 emissions from the industry sector (see Figure S9). This type of inconsistencies also occur in a lesser measure in other cities and similarly to CAMS-REG, are likely explained by the precision of their attribution as point sources.

Overall, inconsistencies associated with EMEP and CAMS-REG mostly appear for the other and industry sectors, mainly pointing to issues related to spatial distribution, i.e. to urban activity shares.

S2. Comparisons of EDGAR, CAMS and EMEP vs the ensemble over Poland

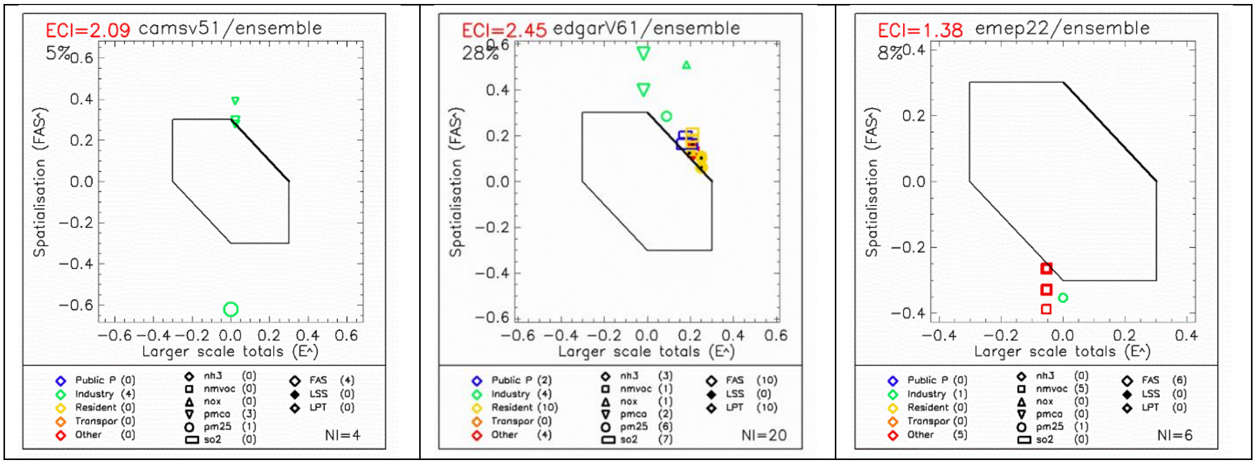

Figure S5: Overview diamonds. The diagrams show the comparison of the three ensemble components (CAMS-REG, EDGAR, EMEP) with the ensemble inventory over Poland. Symbols and colours are as specified in the legend. In all diagrams, only inconsistencies are displayed

For EMEP (right in Figures S5 and S6), inconsistencies are all related to the spatial distribution of the emissions (FAS) with factors slightly larger than 2 for the other sector NMVOC emissions in the cities of Krakow (Figure S5), Lodz and Rzeszow and for the PM2.5 industry emissions in Gdansk. Here again, the location of the main emission sources is similar with EDGAR and CAMS-REG, the EMEP estimates are significantly lower. In the case of NMVOC emissions, EMEP has higher values for all sectors, with the exception of residential combustion (GNFR C). The issue therefore originates from the sectorial share at country level.

For EDGAR (middle in Figures S5 and S6), we find a comparable share between country and urban scale inconsistencies. These country inconsistencies appear because the sum of LPT and LSS is larger than the threshold of 2 while their individual values remain below this threshold. This is why no country scale issues appearsFigure S6. The largest (factor 3) urban scale issues (FAS) are identified for the industrial sector for PMco in Kielce and Czestochowa and for NOx in Krakow. Gridded data for PMco/Kielce (Figure S16) indicate that industrial locations are quite similar with those of EMEP and CAMS-REG but the emitted amounts are much larger. EDGAR also shows different values in the residential sector for PM2.5 at country level. Explanations for such differences are linked with the fact that no emissions are allocated to biomass technologies in EDGAR, and that emission factors for some fuels are very different. For example, the EDGAR emission factor for other bituminous fuel allocated to small boilers is nearly the double of the default values. On the other hand, the values reported for Poland (2020) for both coal and biomass emission factors are well below default values, increasing the difference with the EDGAR estimation. Note that these emission factors have been significantly revised in the Poland 2022 submission, which will be reflected in future EMEP and CAMS-REG inventories.

Similar to EMEP, all inconsistencies in CAMS-REG (left in Figures S5 and S6) are related with the spatial distribution of emissions. The largest inconsistencies occur for industrial emissions of PM2.5 in Kielce (Figure S12) and of PMco in Bydgoszcz. In both cases, CAMS-REG distributes its emissions over more locations with a higher intensity.

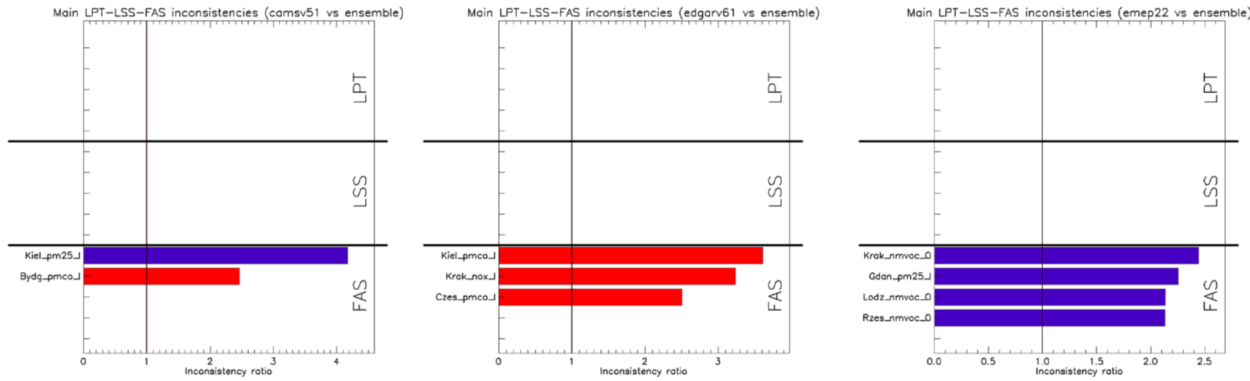

Figure S6: Major inconsistencies (up to 5 per category) for LPT, LSS and FAS for CAMS-REG (left), EDGAR (middle) and EMEP (right). Red and blue shadings indicate an overestimation or underestimation of the individual inventory with respect to the ensemble, respectively.

Many possible reasons for differences between local and Europe-wide inventories exist. In the case of Poland, a possible source of inconsistencies is a consequence of how the Polish NB operates and under what rules. Any given “user of the environment” is obliged to report emissions caused by a specific industrial/chemical process for which his/hers “permit to use the environment” is issued. The pollutants and GHG list that must be reported to NB differs among chemical/industrial processes altering “users of environment” obligations. Emission from NB data is not taken into account by the Polish National Statistical Office directly and the primary source of Europe wide inventories activity data relies on national statistics. Furthermore, while the Polish EMEP reports are partially based on NB and partially on original methodology (additional emission values) causing disagreements with NB, CED directly adopts emission values reported to NB without additional changes. This issue will be further investigated among CED and Polish EMEP compilers.

Yet another issue is that in the case of specific installations registered in NB, reports might be based on direct stack measurements or actual condition of installations while the top-down approach accounts only for general resources/fuel consumption. The advantage of NB over top-down approaches is its sensitivity to temporal variability since reporting users are aware of any changes in fuel or other resources quality they consume, rapid changes in production volumes, new technologies used, newly mounted stack filters, etc. Those small changes might not be captured in full in bulk national statistics, commonly based on fuel sales. Finally, it must be commented that in the case of NB, the possible accidental “human factor” might be a source of additional errors since reports are done manually via the online system. Despite some automatic checking algorithms and manual expert evaluation, discrepancies are possible.

While EU-wide inventory compilers distribute country totals obtained from bulk national statistics, population density is often used as a spatial proxy. In this context, the resolution-free design of CED inventory might be a paradoxical limitation here since the exact geographical location of emission sources is prioritized, and some activities are very tough to allocate. For example, coating applications (2D3d) which are responsible for >63 Mg of NMVOC emissions (2018) in Poland, might be omitted in CED due to a lack of reliable spatial data in case they are not provided by NB users in full. Yet another issue is that this pollutant is not being monitored *in-situ* in Poland (and many other countries), which also hampers the interpretation of emission data.

S3. Comparison of gridded emission maps

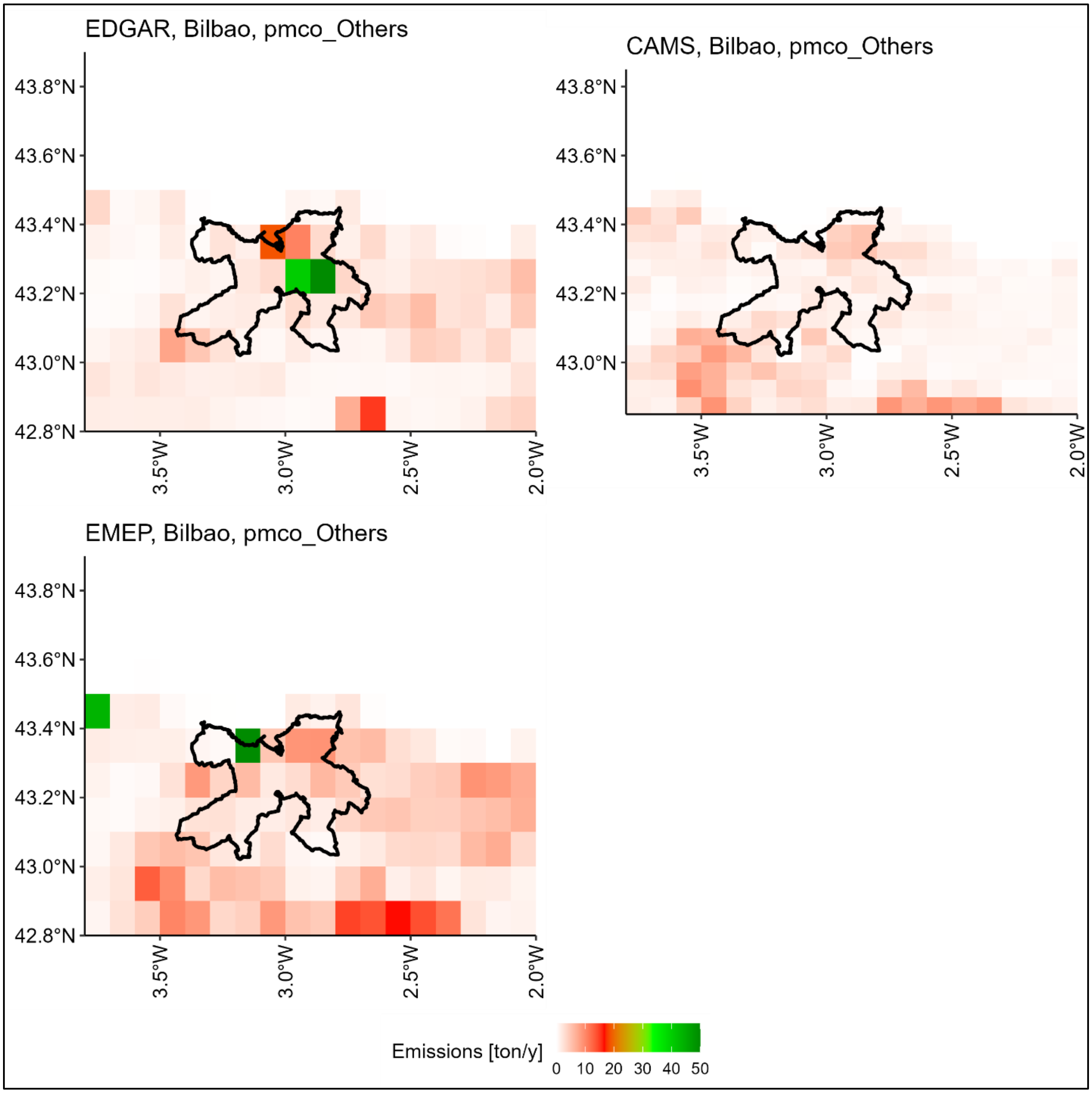

Figure S7: PMco emissions for the other sector in Bilbao. The dark line indicates the shape of the functional urban area.

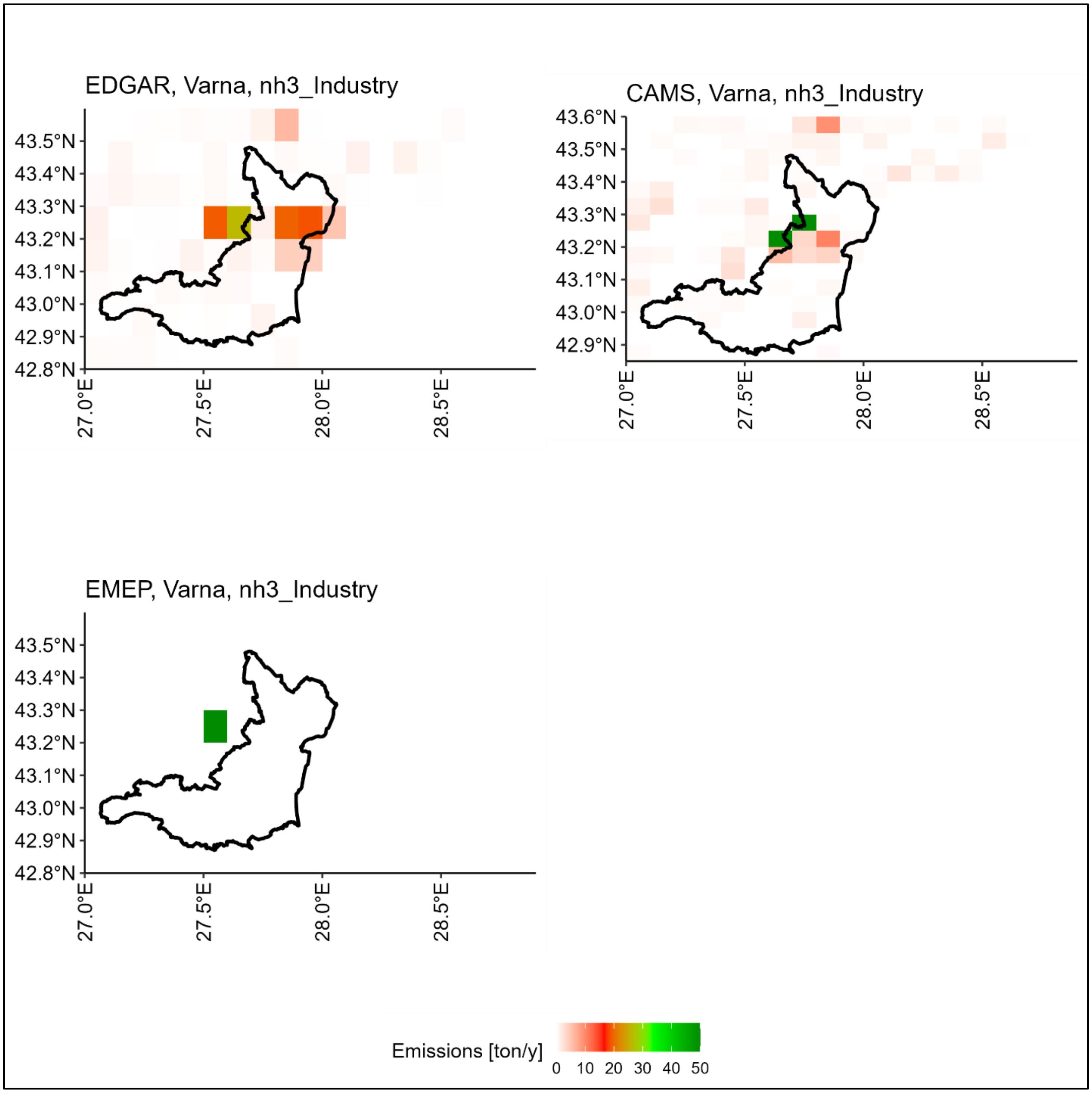

Figure S8: NH3 emissions for the industry for Varna. The dark line indicates the shape of the functional urban area.

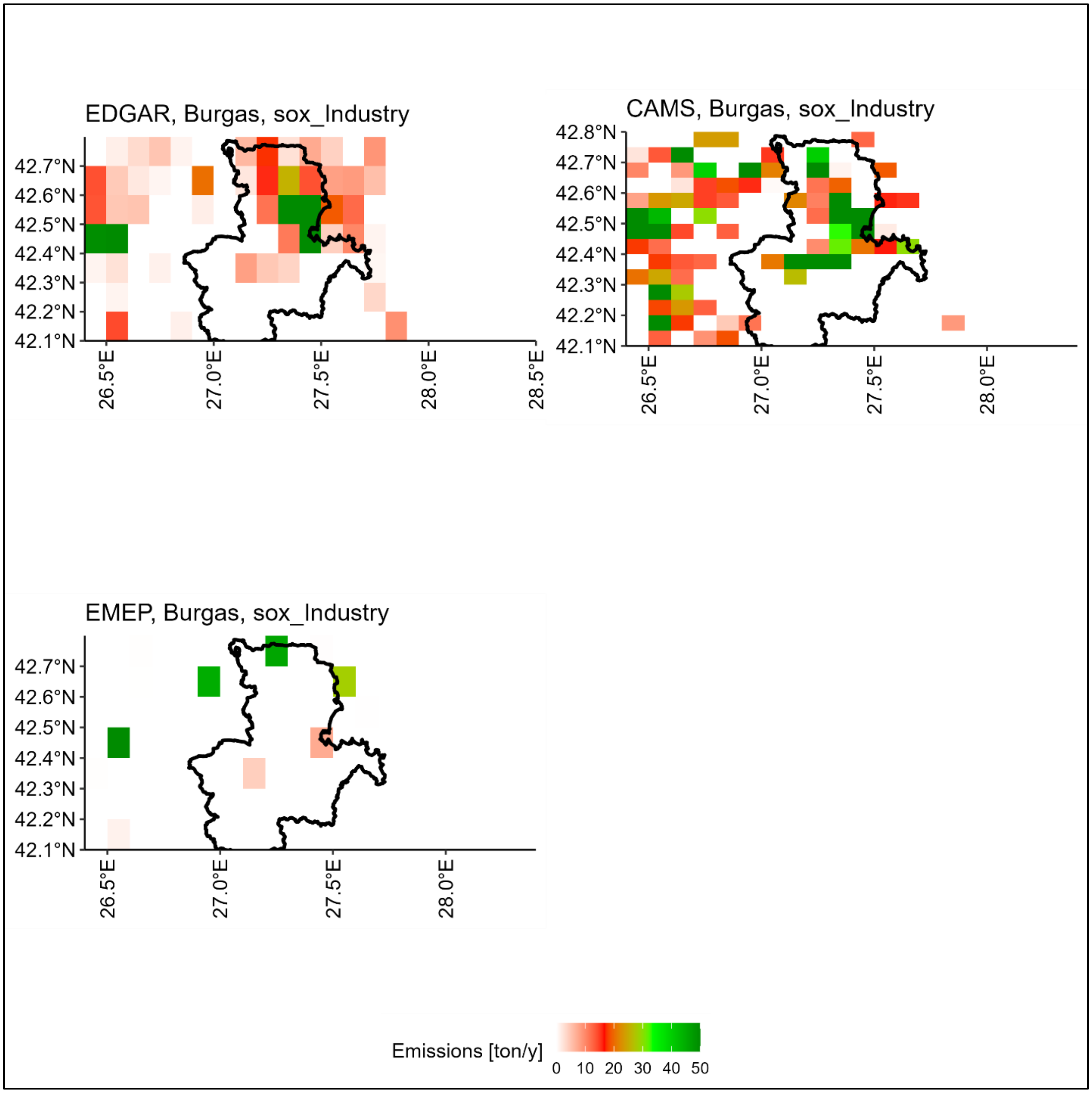

Figure S9: SO2 emissions for industry in Burgas (ES). The dark line indicates the shape of the functional urban area.

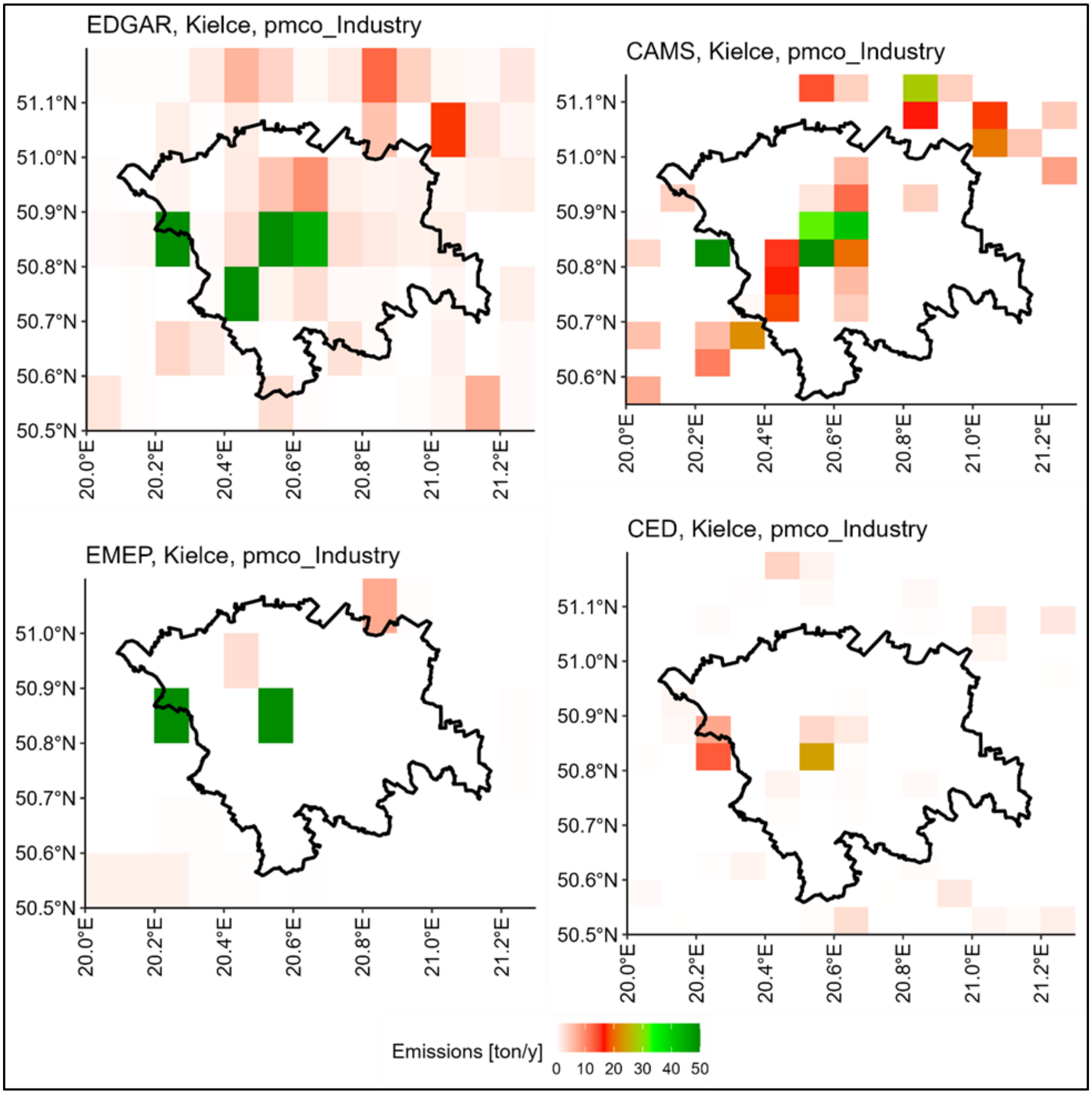

Figure S10: PMco emissions for the industry sector in Kielce. The dark line indicates the shape of the functional urban area.

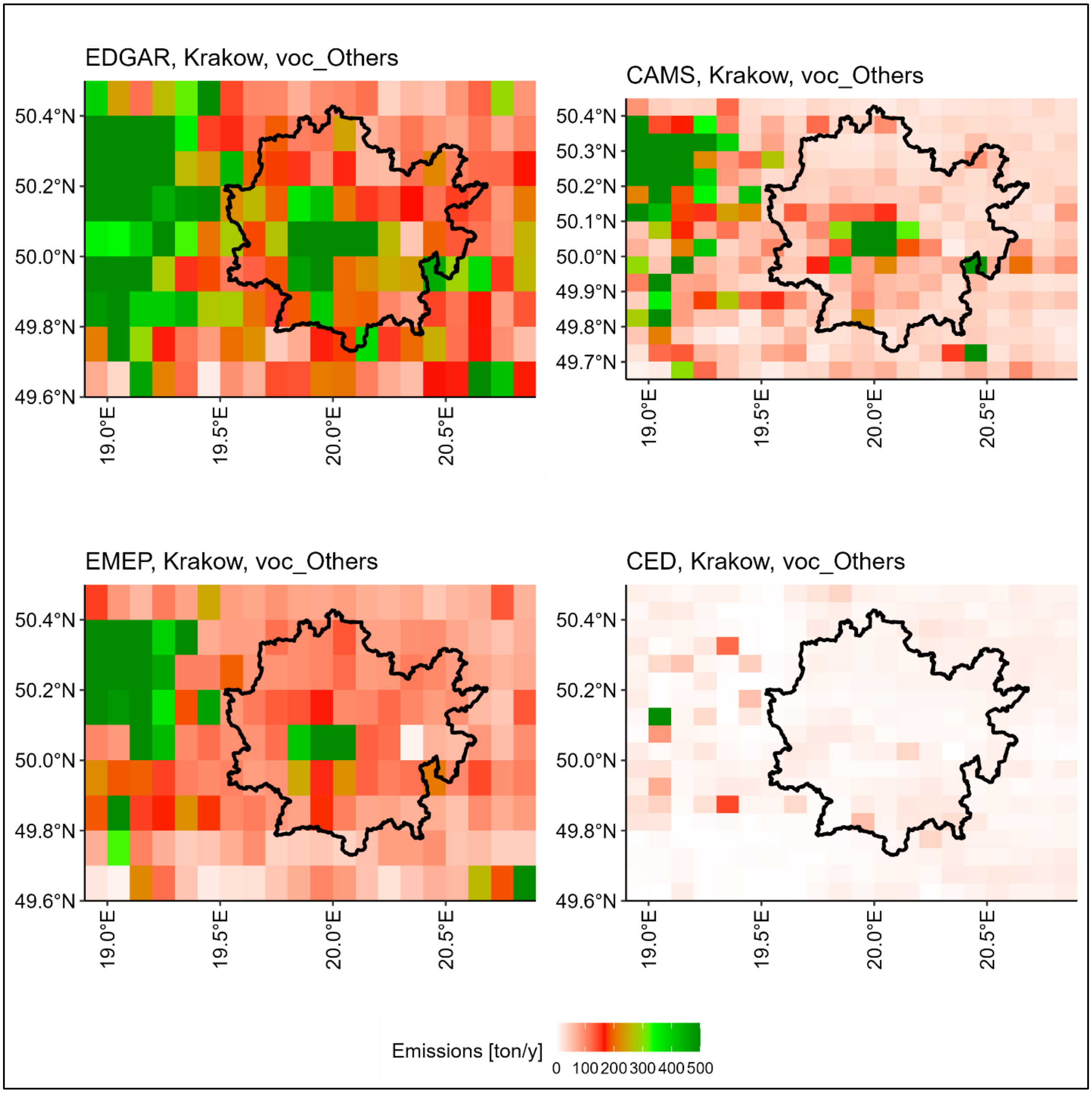

Figure S11: VOC emissions for the other sector in Krakow. The dark line indicates the shape of the functional urban area.

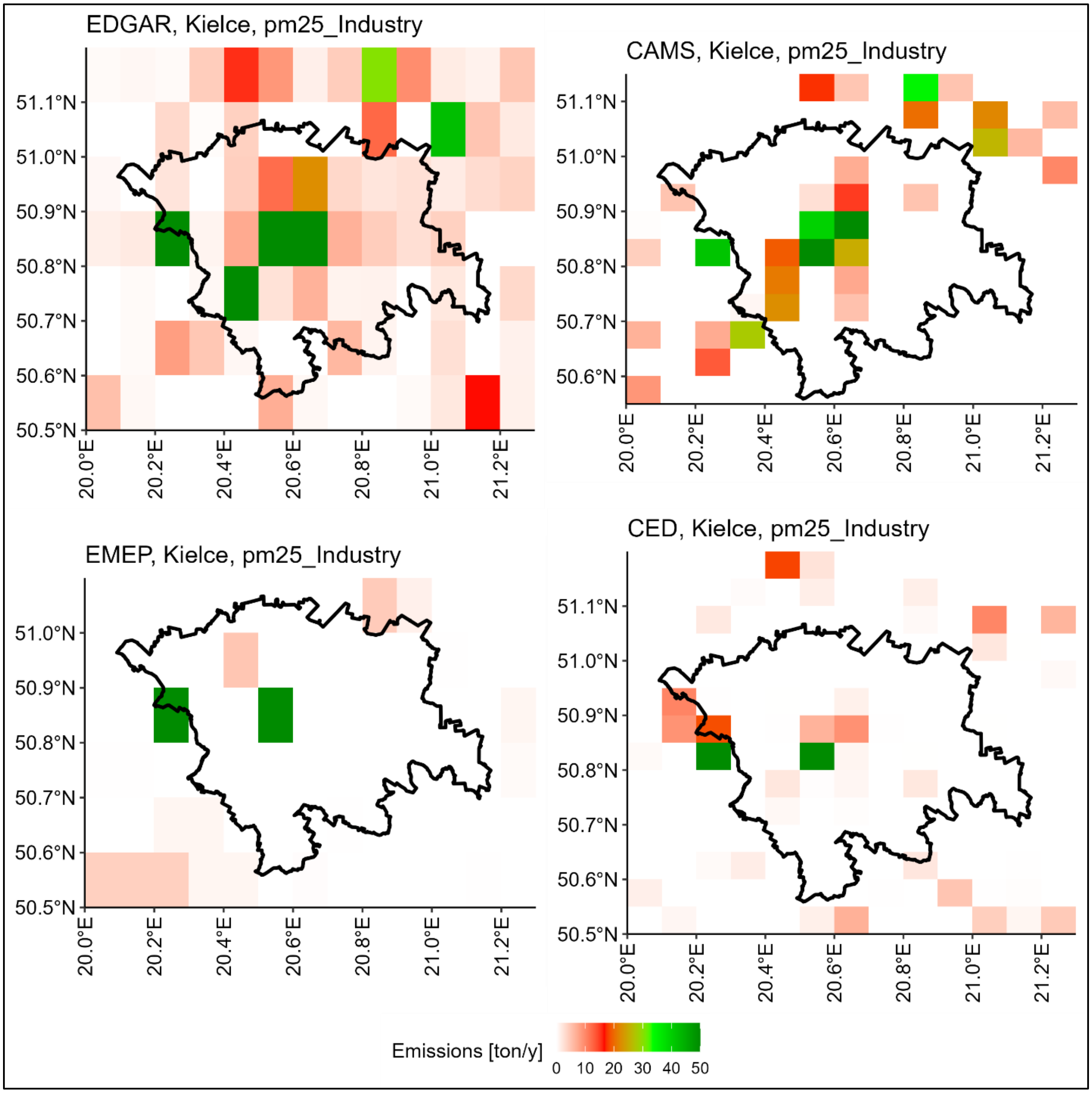

Figure S12: PM25 emissions from the industry sector in Kielce. The dark line indicates the shape of the functional urban area.

. GNFR	SNAP	CED
A_PublicPower	SNAP01	Emission from NB named as SNAP1
B_Industry	SNAP03 and SNAP04	Emission from NB named as SNAP3 and SNAP4
I_Offroad	Part of SNAP08	Emission from agricultural tractors
F_RoadTransport	SNAP07	Emission from road transport
C_OtherStationaryComb	SNAP02	Emission from residential heating
D_Fugitive	SNAP05	Emission from heap and excavation and from NB named as SNAP5
E_Solvents	SNAP06	Emission from NB named as SNAP6
K_AgriLivestock i L_AgriOther as one GNFR K+L	SNAP10	Emission from agriculture and lifestock
J_Waste	SNAP09	Emission from landfills and from NB named as SNAP5

Table S1: Translation from SNAP to GNFR applied in CED

	NMVOC		NH3		NOx		PM10		PM25		SO2	
	CED	**EMEP**	**CED**	**EMEP**	**CED**	**EMEP**	**CED**	**EMEP**	**CED**	**EMEP**	**CED**	**EMEP**
GNFR A	0.28[1)]	2.68[1)]	0.13	0.00	115.91	135.09	8.27	5.96	5.60	3.21	126.29[1)]	193.17[1)]
GNFR B	17.76[2)]	107.75[2)]	5.98	4.25	69.30	74.70	12.06[2)]	59.99[2)]	8.89	34.69	45.23[2)]	107.83[2)]
GNFR C	201.71	101.83	-	8.31	50.55	73.69	190.66[5)]	88.51[5)]	187.02[5)]	59.12[5)]	113.10	115.26
GNFR D	1.50[3)]	79.17[3)]	-	0.06	0.32[3)]	3.58[3)]	19.44	9.58	4.81	1.64	0.16[3)]	7.52[3)]
GNFR E	13.98[3,4)]	164.11[3,4)]	0.14	0.15	0.00	0.06	0.02[3)]	0.93[3)]	0.021[1)]	0.93[1)]	0.00	0.68
GNFR F	74.98	75.52	2.93	2.95	274.00	273.50	18.10	18.06	13.57	13.54	0.54	0.58
GNFR G	0.00	0.01	0.00	0.05	0.00	4.03	0.00	0.15	0.00	0.15	0.00	0.20
GNFR H	0.03	0.08	0.00	0.00	0.66	1.85	0.00	0.01	0.00	0.01	0.05	0.15
GNFR I	3.30	4.12	0.02	0.02	35.56	46.01	1.48	1.69	1.47	1.67	0.05	0.15
GNFR J	0.77[2,3)]	6.31[2,3)]	0.27	0.89	1.33	1.97	0.51[2,3)]	4.47[2,3)]	0.47[2,3)]	5.73[2,3)]	0.18	0.26
GNFR KL	105.97	105.96	303.43	300.14	69.91	69.22	29.87	29.48	3.32	3.29	0.00	0.01
SUM	420.27	647.53	312.91	316.84	617.54	683.74	280.41	218.83	225.17	123.97	285.59	425.82

Table S2: Country totals for CED and EMEP data (biggest differences are underlined and commented below).

1) NB users are not obligated to report all pollutants (NMVOC can be omitted) in this sector. For EMEP reporting, the missing emission values are estimated using official emission factors (EMEP/EEA air pollutant emission), which depend on basic activity data (Tier 1 – fuel consumption)

2) Reports provided to NB are based on user-specific permits which specify the list of pollutants to be reported. In EMEP reports, emissions are calculated using official EMEP/EEA emission factors.

3) In the case of NB, some reports might be based (or supplemented with) on individual emission measurements resulting from user-specific industrial processes. Such in-situ data does not always align with EMEP reporting methodology, nor does it cover the same set of pollutants.

4) For some processes categorized into GNFR E, which are not fully addressed in CED, EMEP emissions are based on population (like domestic solvent use, including fungicide and dry cleaning).

5) PM10 and PM2.5 emissions from stationary combustion are much lower in EMEP, because the data used in this work do not yet include condensable emissions whereas CED does.