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Supplement of

Speciation of anthropogenic emissions of non-methane volatile organic compounds: a global gridded data set for 1970–2012

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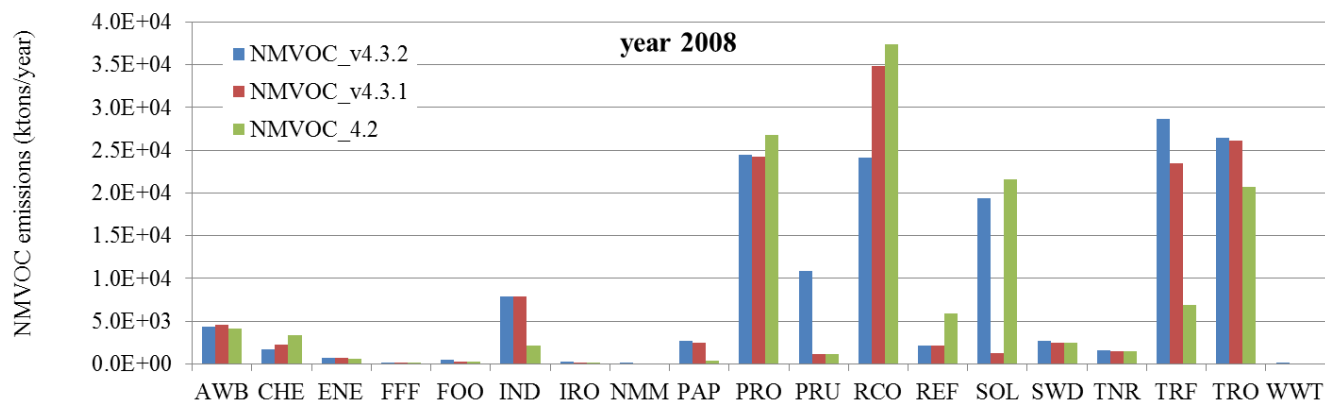
Supplementary material

S1 – NMVOC emissions comparison using EDGAR versions, HTAP_v2 and EEA inventories

Figure S1 shows the comparison of global NMVOC emissions by sector for different EDGAR versions v4.2 (refer to <http://edgar.jrc.ec.europa.eu/overview.php?v=42>), v4.3.1 (refer to <http://edgar.jrc.ec.europa.eu/overview.php?v=431>) and v4.3.2 (<http://edgar.jrc.ec.europa.eu/overview.php?v=432>) for the most recent year (2008) available for all datasets. Total emissions are slightly higher (ca 17%) in the current version of EDGAR compared to v4.3.1 mainly due to changes in the activity data and emission factors. At sector level, rather good agreement is observed between EDGARv4.3.2 and EDGARv4.3.1, although major differences are found for the application of solvents showing 15.6 times higher emissions for EDGARv4.3.2 due to revised activity data (to account for household products and other solvents use) and emission factors (especially for paints and pesticides), the residential and transformation industry sectors having ca 30% and 22% lower emissions. Finally, in EDGARv4.3.2 waste water treatment and glass production (from the year 1990) have been introduced. Figures S2 and S3 show the comparison of NMVOC emissions of EDGARv4.3.2 and the best estimates provided by the HTAP_v2.2 inventory for the year 2010 by HTAP sector and country (refer to Janssens-Maenhout et al. (2015) and http://edgar.jrc.ec.europa.eu/htap_v2/index.php). Very good agreement for all sectors is found between EDGARv4.3.2 and HTAP_v2.2 for Asian countries and North America (refer to Fig. S2), as well as for Europe (refer to Fig. S3). Lower NMVOC emissions are reported by EDGARv4.3.2 for India and Indonesia for the residential and transport sectors compared to the HTAPv2 data (although the reported HTAP_v2.2 emissions appear to be very high compared for example with the Chinese ones). On the other hand, EDGARv4.3.2 provides larger NMVOC emissions for Germany for the residential sector, although the HTAP_v2.2 data appear to be too low compared for example with France residential emissions. In general, larger differences between the two inventories are observed for the power generation due to the low NMVOC emissions associated with this sector.

Focusing on European countries (see Fig. S4), detailed comparison by sector and country (defined with ISO codes) is also performed with officially reported EEA NMVOC emission inventories for the year 2010 (<http://www.eea.europa.eu/data-and-maps/data/national-emissions-reported-to-the-convention-on-long-range-transboundary-air-pollution-lrtap-convention-10>). Total NMVOC emissions at European scale are 15% higher for EDGAR compared to EEA and HTAP_v2.2. However, insights on the origin of such differences can be retrieved looking at sectorial emissions. The power generation sector in EU represents less than 2% of total NMVOC emissions although it shows quite some discrepancies among inventories. As shown in Fig. S3 and Fig. S4, industrial, residential and ground transport NMVOC emissions are characterized by better agreement among the three inventories, with the exception of few countries. EDGAR estimates 30-50% lower emissions for ground transport emissions for France, Poland and Czech Republic compared to HTAP and EEA, while it generally overestimates residential emissions (e.g. in particular for Germany, France and UK, possibly due to an underestimation of

the combustion of biomass in the household sector as reported by van der Gon et al. (2015)). Differences in the NMVOC emissions of the industrial sector among the inventories might be due to the underestimation by 50% of the EDGAR gas distribution subsector for Europe and by 15% at the global scale.



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Figure S1. Comparison of 2008 EDGAR emissions by sector for different versions.



Figure S2. Comparison of 2010 NMVOC sectorial emissions estimated by EDGARv4.3.2 and HTAP_v2 for Asian countries and North America.

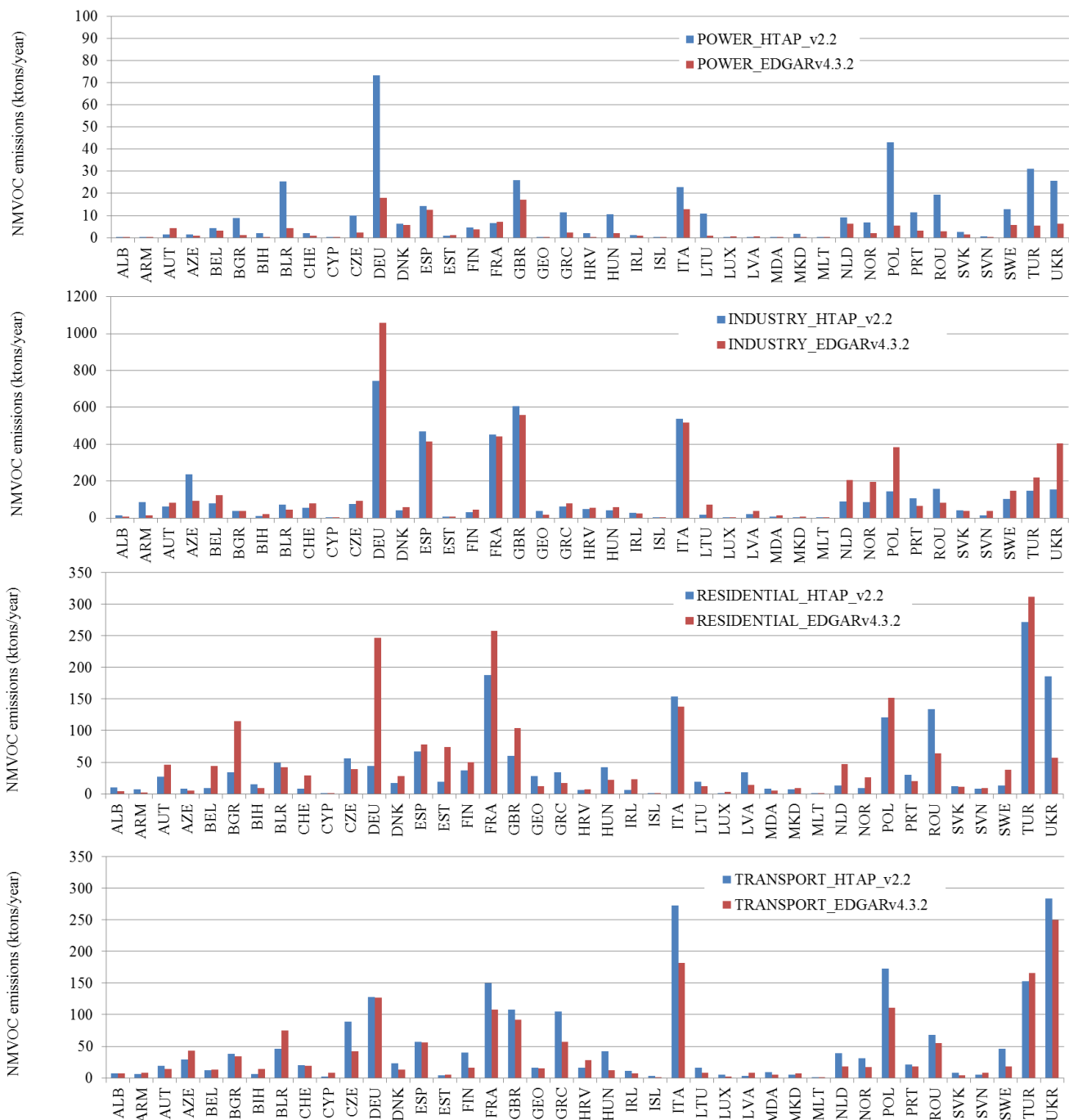


Figure S3. Comparison of 2010 NMVOC sectorial emissions estimated by EDGARv4.3.2 and HTAP_v2 for Europe.

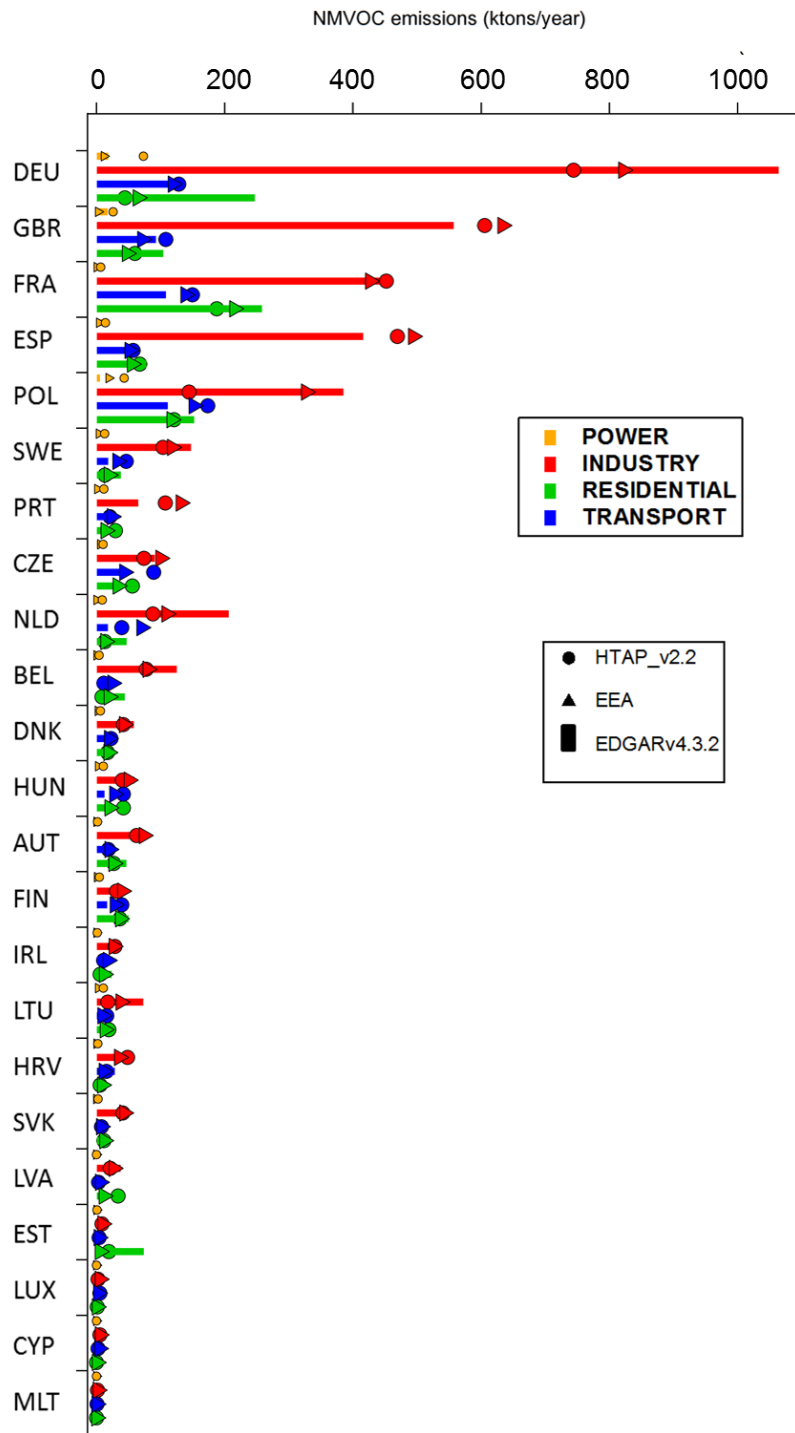


Figure S4. Comparison of 2010 NMVOC emissions from the power generation, industry, residential and combustion sectors of the HTAP_v2.2, EDGARv4.3.2 and EEA inventories.

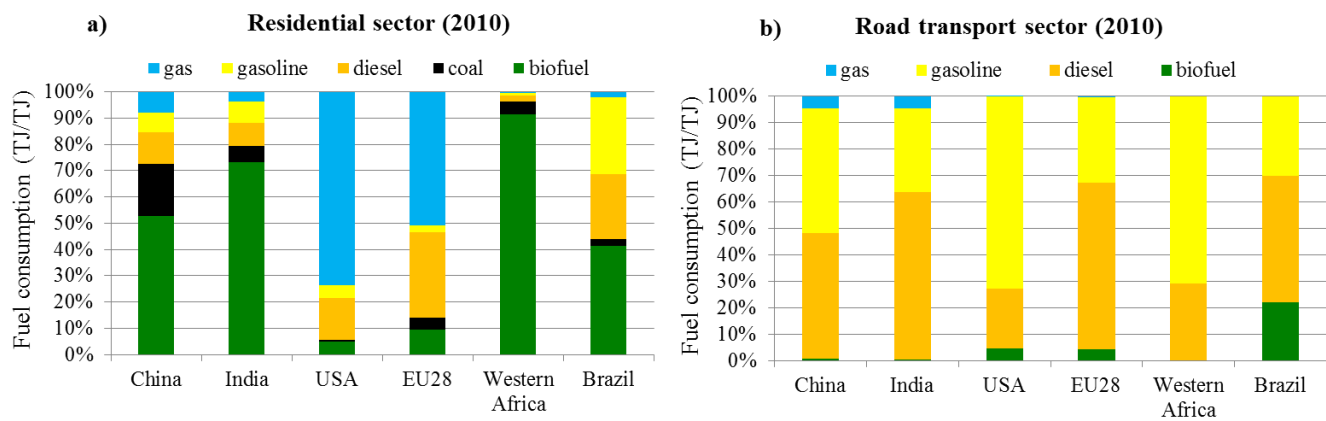


Figure S5. Share of different fuels consumed in the residential (a) and road transport (b) sectors in 2010 for major world regions.

S2 – Mapping NMVOC profiles to EDGAR processes

Table S1. First step in mapping profiles to EDGAR process codes.

| Source code | Source description | Tech code | EOP code | Profile name | Mapping quality |
|-------------|-------------------------------------|-----------|----------|---|-----------------|
| CHE.BLK.CPS | CHa-Polystyrene (total) | NSF | NOC | Plastics Production - Polystyrene | 1 |
| CHE.BLK.CPT | CHa-Phthalic anhydride | NSF | NOC | Phthalic Anhydride - O-Xylene Oxidation - Main Process Stream | 1 |
| CHE.BLK.CPV | CHa-Poly Vinyl Chloride (PVC) | NSF | 020 | Plastics Production - Polyvinyl Chlorides and Copolymers | 1 |
| CHE.BLK.CPV | CHa-Poly Vinyl Chloride (PVC) | NSF | NOC | Plastics Production - Polyvinyl Chlorides and Copolymers | 1 |
| CHE.BLK.CRU | CHa-Rubber, total (SBR + synthetic) | NSF | NOC | Consumer Products: Rubber and Vinyl Protectants - Aerosols | 1 |
| CHE.BLK.CST | CHa-Styrene | NSF | NOC | Methyl Styrene | 1 |
| CHE.BLK.CVC | CHa-Vinyl chloride | NSF | NOC | Plastics Production - Polyvinyl Chlorides and Copolymers | 1 |
| CHE.BLK.CXY | CHa-Xylenes | NSF | NOC | m-Xylene | 1 |

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Table S2. Example of mapping profiles with a quality code of 2.

| Source code | Source description | Tech code | EOP code | Profile name | Mapping quality |
|-------------|---------------------------------------|-----------|----------|---|-----------------|
| ENE.CHP.OGS | Public cogeneration: Coke Oven Gas | BO0 | 223 | External Combustion Boiler - Coke Oven Gas | 2 |
| ENE.CHP.OGS | Public cogeneration: Coke Oven Gas | BO0 | 300 | External Combustion Boiler - Coke Oven Gas | 2 |
| ENE.CHP.OGS | Public cogeneration: Coke Oven Gas | BO0 | 423 | External Combustion Boiler - Coke Oven Gas | 2 |
| ENE.CHP.RGS | Public cogeneration: Refinery Gas | BO0 | 000 | External Combustion Boiler - Refinery Gas | 2 |
| ENE.CHP.OGS | Public cogeneration: Refinery Gas | BO0 | 002 | External Combustion Boiler - Refinery Gas | 2 |
| ENE.CHP.OGS | Public cogeneration: Refinery Gas | BO0 | 003 | External Combustion Boiler - Refinery Gas | 2 |

Notes: BO0 = combustion: boiler for gas/ liquid of any size

Table S3. Example of mapping profiles with a quality code of 3.

| Source code | Source description | Tech code | EOP code | Profile name | Mapping quality |
|-------------|--------------------------------|-----------|----------|--|-----------------|
| TRO.ROA.BDS | Biodiesel in Road transport | BS0 | NOC | Biodiesel Exhaust - Light Duty Truck operated at 0 °C; Cold Start | 3 |
| TRO.ROA.BDS | Biodiesel in Road transport | BS0 | PEU | Biodiesel Exhaust - Light Duty Truck operated at 0 °C; Cold Start | 3 |
| TRO.ROA.BDS | Biodiesel in Road transport | BS0 | EU1 | Biodiesel Exhaust - Light Duty Truck operated at 0 °C; Cold Start | 3 |
| TRO.ROA.BDS | Biodiesel in Road transport | HD0 | NOC | Biodiesel Exhaust - Light Duty Truck operated at 0 °C; Cold Start | 3 |
| TRO.ROA.BDS | Biodiesel in Road transport | HD0 | PEU | Biodiesel Exhaust - Light Duty Truck operated at 0 °C; Cold Start | 3 |
| TRO.ROA.BDS | Biodiesel in Road transport | HD0 | EU1 | Biodiesel Exhaust - Light Duty Truck operated at 0 °C; Cold Start | 3 |

5 Notes: BS0 = busses, HD0 = heavy duty vehicles

Table S4. Example of matching profiles with a quality code of 4, 5 and 6.

| Source code | Source description | Tech code | EOP code | Profile name | Mapping quality |
|-------------|---|-----------|----------|---------------------------------|-----------------|
| ENE.AEL.BFG | Auto produced electricity: Blast Furnace Gas | BO0 | 000 | Coke Oven Blast Furnace Gas | 4 |
| ENE.AEL.BFG | Auto produced electricity: Blast Furnace Gas | BO0 | 002 | Coke Oven Blast Furnace Gas | 4 |
| ENE.AEL.BFG | Auto produced electricity: Blast Furnace Gas | BO0 | 003 | Coke Oven Blast Furnace Gas | 4 |
| ENE.AEL.CRU | Auto produced electricity: Crude Oil | BO0 | 000 | Other Electric Power Generation | 5 |
| ENE.AEL.CRU | Auto produced electricity: Crude Oil | GT0 | 000 | Other Electric Power Generation | 5 |
| ENE.AEL.CRU | Auto produced electricity: Crude Oil | IC0 | 000 | Other Electric Power Generation | 5 |
| TNR.SEA.HFO | Residual Fuel Oil in International marine bunkers | BSP | NOC | Residual Oil-Fired Power Plant | 6 |
| TNR.SEA.HFO | Residual Fuel Oil in International marine bunkers | BSS | NOC | Residual Oil-Fired Power Plant | 6 |
| TNR.SEA.HFO | Residual Fuel Oil in International marine bunkers | CSP | NOC | Residual Oil-Fired Power Plant | 6 |

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S3 – Aggregation of the 25 NMVOC species

Table S5. Aggregation of the 25 NMVOC species into 8 main groups.

In this work we have developed the NMVOC split into 25 species. However, in order to show and discuss the results, they have been grouped into 8 major categories as reported in Table S5.

| 25 NMVOC species codes | 25 NMVOC species | 8 aggregated NMVOC species |
|-------------------------------|---------------------------------------|-----------------------------------|
| voc1 | Alkanols (alcohols) | Alkanols |
| voc2 | Ethane | Alkanes (C2 - C5) |
| voc3 | Propane | Alkanes (C2 - C5) |
| voc4 | Butanes | Alkanes (C2 - C5) |
| voc5 | Pentanes | Alkanes (C2 - C5) |
| voc6 | Hexanes and higher alkanes | Alkanes (C6+) |
| voc7 | Ethene (ethylene) | Alkenes |
| voc8 | Propene | Alkenes |
| voc9 | Ethyne (acetylene) | Alk(adi)enes/alkynes |
| voc10 | Isoprenes | Other |
| voc11 | Monoterpenes | Other |
| voc12 | Other alk(adi)enes/alkynes (olefines) | Alk(adi)enes/alkynes |
| voc13 | Benzene (benzol) | Aromatics |
| voc14 | Methylbenzene (toluene) | Aromatics |
| voc15 | Dimethylbenzenes (xylenes) | Aromatics |
| voc16 | Trimethylbenzenes | Aromatics |
| voc17 | Other aromatics | Aromatics |
| voc18 | Esters | Other |
| voc19 | Ethers (alkoxy alkanes) | Other |
| voc20 | Chlorinated hydrocarbons | Other |
| voc21 | Methanal (formaldehyde) | Alkanals |
| voc22 | Other alkanals (aldehydes) | Alkanals |
| voc23 | Alkanones (ketones) | Other |
| voc24 | Acids (alkanoic) | Other |
| voc25 | Other NMVOC (HCFCs, nitriles, etc.) | Other |

S4- Details on the EDGAR v4.3.2 methodology

Total NMVOC emissions from a given sector *i* in a country *C* accumulated during a year *t* are estimated with the following formula in the EDGAR database:

$$EM_i(C,t) = \sum_{j,k} \left[AD_i(C,t) * TECH_{i,j}(C,t) * EOP_{i,j,k}(C,t) * EF_{i,j}(C,t) * (1 - RED_{i,j,k}(C,t)) \right]$$

- 5 EDGAR emission estimates are based on country-specific activity data (AD) for each anthropogenic emission sector *i*, on which a mix of *j* technologies (TECH) and a mix of *k* end-of-pipe measures (EOP) are installed; uncontrolled emission factors (EF) for each sector *i* and technology *j* with relative reduction (RED) by abatement measure *k* are also used in the calculation. The technology mix, (uncontrolled) emission factors and end-of-pipe measures are defined at country-specific, regional, country group (e.g. Annex I/ Non-Annex I), or global level. In particular, NMVOC emission factors are consistent with the EMEP/EEA 2013 Guidebook (EEA, 2013) for Europe and scientific literature has been taken into account to introduce country- and region- specific information, while abatement measures are implemented mainly for the road transport sector (consistent with the Euro standards), for the production of chemicals (CHa-formaldehyde (methanal), total polyethylene, CHa-propylene glycol, total polystyrene), for power generation (auto produced electricity and public electricity production from natural gas) and for landfills. Further details on the EDGAR methodology can be found in Section S4 of the Supplementary material of Crippa et al. (2016a).

Table S6 reports the Euro standards implementation over time as reported by regulations. Country- and region- specific time series with the penetration of the Euro standards are applied in the EDGAR database as reported in Crippa et al. (2016b).

- 20 **Table S6 - Euro standards implementation over time (1990-2012). Note that mopeds Pre-Euro standards are defined as PEU for Europe and are also assumed to take place from 1970 till 1992.**

| | 1990 | 1991 | 1992 | 1993 | 1994 | 1995 | 1996 | 1997 | 1998 | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | 2009 | 2010 | 2011 | 2012 |
|-----------------------------------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| Passenger car | PEU | PEU | EU1 | EU1 | EU1 | EU1 | EU2 | EU2 | EU2 | EU2 | EU3 | EU3 | EU3 | EU3 | EU3 | EU4 | EU4 | EU4 | EU4 | EU5 | EU5 | EU5 | EU5 |
| Light duty vehicle | PEU | PEU | EU1 | EU1 | EU1 | EU1 | EU2 | EU2 | EU2 | EU2 | EU3 | EU3 | EU3 | EU3 | EU3 | EU4 | EU4 | EU4 | EU4 | EU5 | EU5 | EU5 | EU5 |
| Heavy duty vehicle and bus | PEU | PEU | EU1 | EU1 | EU1 | EU1 | EU2 | EU2 | EU2 | EU3 | EU3 | EU3 | EU3 | EU3 | EU3 | EU4 | EU4 | EU4 | EU4 | EU5 | EU5 | EU5 | EU5 |
| Motorcycle/Moped | PEU | PEU | PEU | PEU | PEU | PEU | PEU | PEU | PEU | EU1 | EU1 | EU1 | EU2 | EU2 | EU2 | EU2 | EU3 | EU3 | EU3 | EU3 | EU3 | EU3 | EU3 |

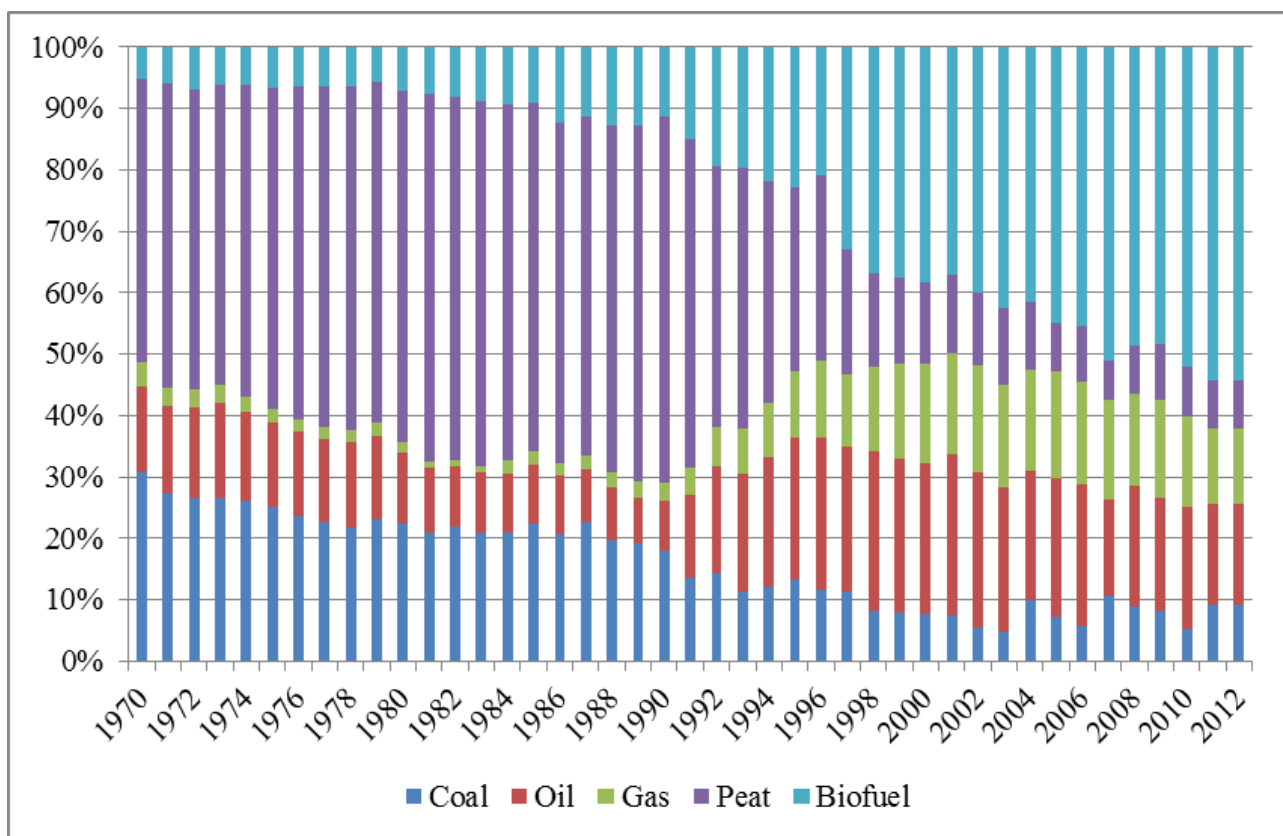


Figure S6. Relative share of different fuels to NMVOC emissions of residential sector in Germany during 1970-2012.

Table S7. Matching of RETRO sectors and EDGAR sources.

| RETRO sector | RETRO sector description | EDGAR source mapped |
|--------------|--|---------------------|
| Agr | Agriculture and Land use change | AWB |
| Exf | Extraction and distribution of fossil fuels | PRO, REF |
| Inc | Industrial combustion | IND, TRF |
| Pow | Power generation | ENE |
| Res | Residential, commercial and other Combustion | RCO |
| Sol | Solvent use | SOL |
| Tra | Road transport | TRO |
| Was | Waste treatment and disposal | SWD |

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