Till Zimmermann, September 2019

ESTIMATING NMVOC-EMISSIONS ON COUNTRY LEVEL: PRODUCT- AND SOLVENT-BASED APPROACHES – THE CASE OF GERMANY

Selected results from project „Update of the German inventory for NMVOC emissions resulting from the use of solvents and solvent-containing products for the reporting years 2019 and 2020 (Project No. 110387)" commissioned by the German Environment Agency

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Summary

Organic solvents are used as components and additives in a variety of chemical products used by private end users, industrial users, and in industrial manufacturing processes. Depending on their chemical structure, many of these solvents have the property that they evaporate resulting in the formation of non-methane volatile organic compounds (NMVOC) contributing to the increase in tropospheric ozone concentrations and/or being toxic to human health and the environment. Reducing NMVOC emissions is a nationally and internationally recognized environmental goal, especially against the background of the ozone problem. The legal framework in place, requires an annual reporting of NMVOC of all EU member states to the EEA. To determine NMVOC emissions different approaches exist. In this article, the product-based and the solvent-based approach are described using the German NMVOC inventory and the ESIG model as examples. The German NMVOC inventory is maintained by the Federal Environment Agency and used for its annual reporting to the EEA. Both inventories differ significantly with regard to the calculated emissions. In this article, a closer look at these differences and potential causes is taken.

1 Background

Organic solvents are used as components and additives in a variety of chemical products used by private end users, industrial users, and in industrial manufacturing processes. Product examples range from personal care products to household or commercial (surface) cleaning agents to solvents used in industrial extraction processes, etc. Depending on their chemical structure, many of these solvents have the property that they evaporate (completely) at room temperature or under operation conditions due to their vapour pressure, resulting in the formation of non-methane volatile organic compounds (NMVOC). The Decopaint Directive (Directive 2004/42 / EC) defines organic compounds as compounds containing at least the element carbon and one or more of the elements hydrogen, oxygen, sulfur, phosphorus, silicon, nitrogen or a halogen, excluding carbon oxides and inorganic compounds Carbonates and bicarbonates and volatile organic compounds as an organic compound with an initial boiling point of not more than 250 ° C at a standard pressure of 101.3 kPa.

NMVOC include a large number of different compounds of which a large share is photochemically active, contributes to the increase in tropospheric ozone concentrations and/or is toxic to human health and the environment (Laurent und Hauschild 2014; Weichenthal et al. 2012; Amann et al. 2008). Just recently, an apparent growth of anthropogenic NMVOC emissions in China has been reported (Li et al. 2019).

Reducing NMVOC emissions is a nationally and internationally recognized environmental goal, especially against the background of the ozone problem. In the long term, a permanent ground-level ozone concentration of <120 µg/m³, below which no harmful health effects are expected (WHO convention), must be achieved. For this purpose, several international agreements have been made, which, among other aspects, contain requirements for the reduction of NMVOC emissions from the application of organic solvents, such as

- the UNECE Multicomponent Protocol (Gothenburg Protocol),
- the EC NEC Directive (directive 2001/81/EC) and the new NEC Directive (2016/2284/EC)
- the EC Decopaint Directive 2004/42/EC,
- the EC Solvent Directive 1999/13/EC (now superseded by the Directive on Industrial Emissions (Directive 2010/75/EU))

However, the NMVOC emissions from the use of organic solvents largely evade direct emission measurements. Most solvents are used outside of plants with targeted exhaust air treatment and emit diffusely into the ambient air. Even where solvents are used in production plants with exhaust air treatment, often processes are non-continuous (batch operations; e.g. systems for cleaning of parts, manual solvent uses as e.g. in spray booths). Here, occurring solvent emissions are typically only partially detected and caught by the exhaust air treatment system. In addition, the amount of exhaust air as well as the NMVOC load in the exhaust air fluctuate significantly over time. A direct measurement of the emission quantity is only possible with elaborate measurement campaigns and uncertainties regarding the transferability of measurement results to longer periods of time remain. Even in the few large plants with a comparatively constant use of solvents, such as band coating systems or similar, continuous measurements of NMVOC concentrations in the exhaust air are an exception.

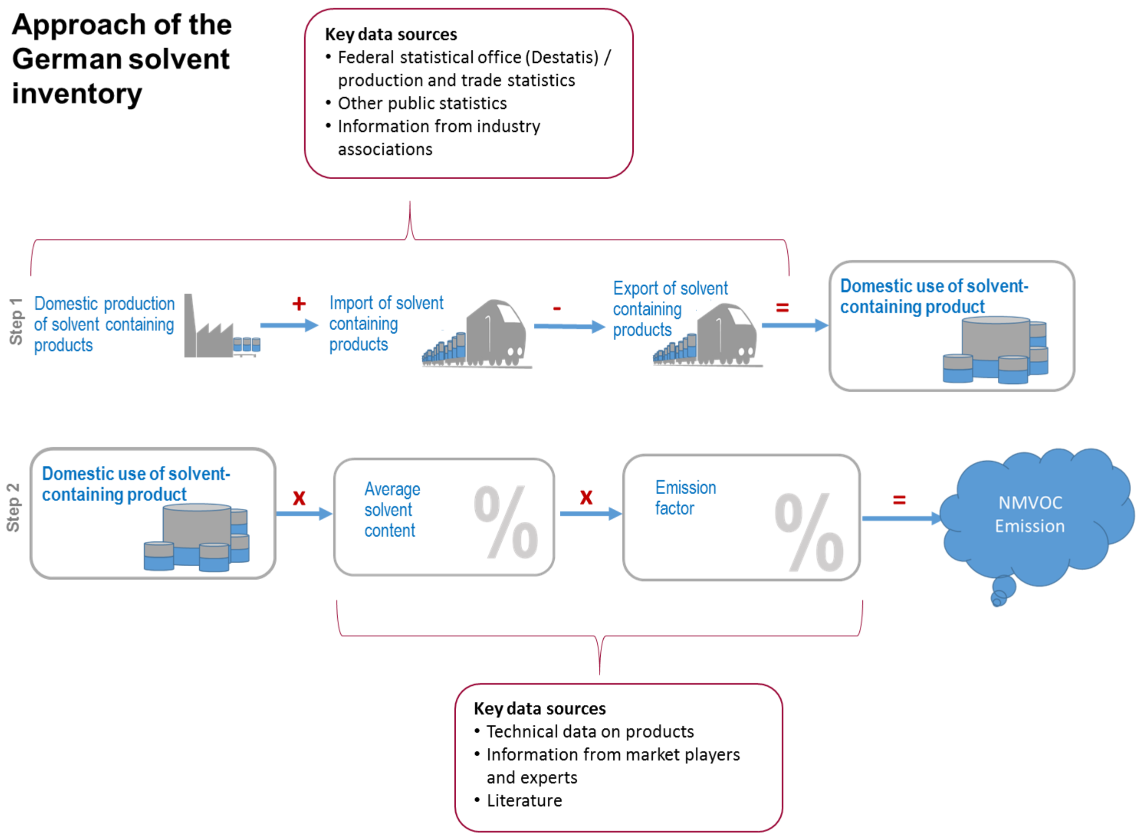
Since direct emissions data as a basis for extrapolation of the emission is not available, the determination of NMVOC emissions from solvent use, which is required on EU and member state level with regard to the established legal framework, is conducted using different methodological approaches, which can quite generally be differentiated into product-based (bottom-up) approaches and solvent-based (top-down) approaches. In this article, both approaches are described and relevant aspects of both are highlighted using the German NMVOC inventory and the ESIG model as examples. The German NMVOC inventory is maintained by the Federal Environment Agency and used for annual reporting to the EEA.

2 Product-based approach: the case of the German Inventory

In Germany, the Federal Environment Agency (Umweltbundesamt, UBA) is responsible for the national solvent inventory, building on expert input and regularly conducted research projects. Using this inventory, NMVOC emissions are calculated annually and reported to the EEA.

The German solvent inventory follows the so-called "product-based approach" and is in line with the requirements of the EMEP guidebook (Tier-2 approach) (EEA 2016). To ensure an annual updatability of the inventory, the inventory has been based on data on production volumes and the respective trade balance (statistics maintained by the Federal Statistical Office) among other periodically updated statistics. The approach is summarized in the following figure.

Figure 1: Structure of the German NMVOC inventory



For all solvent-containing products/product-groups differentiated in the inventory, the VOC content and the use quantities are determined, i.e. the development of the activity rate is modelled based on the quantities of solvent-containing products put on the domestic (German) market. The quantities are then connected to information on the average solvent content and emission factors specific per application to calculate the resulting NMVOC emissions. Average solvent content and emission factors are based on technical product data as well as information from market players and literature.

In order to adjust the average "typical" solvent contents, the emission factors or the distribution of individual product groups to areas of different application conditions, detailed investigations are carried out periodically in different specific parts of the inventory (Tebert 2013; Jepsen et al. 2016a, 2016b; Zimmermann und Jepsen 2018b, 2018a; Theloke et al. 2000; Theloke 2005).

With 198 product groups, the German solvent inventory is significantly more differentiated than the 39 relevant SNAP codes of the EMEP system. The 198 product groups of the German inventory are allocated to the relevant CRF sectors described in the EMEP guidebook:

- 2D 3a: Domestic Solvent Use including Fungicides (coating applications excluded)
- 2D 3d: Coating Applications
- 2D 3e: Degreasing
- 2D 3f: Dry Cleaning
- 2D 3g: Chemical Products
- 2D 3h: Printing
- 2D 3i: Other Solvent Use

Below CRF codes, the structure follows SNAP codes with a further differentiation into sub-categories (product-groups) below the SNAP systematic. The diagram below schematically shows the structure of the inventory system.

CRF	2D 3d Coating Applications
SNAP	
	2D 3d - i Decorative Coating Application
60102	**Car Repairing**
	Car repairing
60103	**Construction and Buildings**
	6 sub-categories
60104	**Building DIY**
	6 sub-categories
60107	**Wood**
	2 sub-categories
	2D 3d - ii Industrial Coating Application
60101	**Manufacture of Automobiles**
	3 sub-categories
60102	**Car Repairing**
	Commercial and other vehicles
60105	**Coil Coating**
60106	**Boat Building**
60107	**Wood**
	Furniture
60108	**Other industrial paint application**
	12 sub-categories
	2D 3d - iii Other Coating Application
60109	**Other non-industrial paint application**
	3 sub-categories

SNAP	2D 3e Degreasing
60201	**Metal degreasing**
60203	**Electronic component manufacturing**
60204	**Other industrial cleaning (e.g. Precision mechanics, Optics, Manufacture of watches and clocks)**
	7 sub-categories

SNAP	2D 3f Dry Cleaning
60202	**Dry Cleaning**

SNAP	2D 3g Chemical Products
60301	**Polyester processing**
60302	**Polyvinychloride processing**
60303	**Polyurethane processing**
60304	**Polystyrene foam processing**
60305	**Rubber processing**
60306	**Pharmaceutical products manufacturing**
60307	**Manufacture of paints and lacquers**
60308	**Manufacture of inks**
60309	**Glues manufacturing**
60310	**Asphalt blowing**
60311	**Adhesive manufacturing, magnetic tapes manufacturing, photographs manufacturing**
60312	**Textile finishing**
60313	**Leather tanning**
60314	**Solvents manufacturing**
	7 sub-categories

SNAP	2D 3h Printing
60403	**Printing industry / printing**
	16 sub-categories

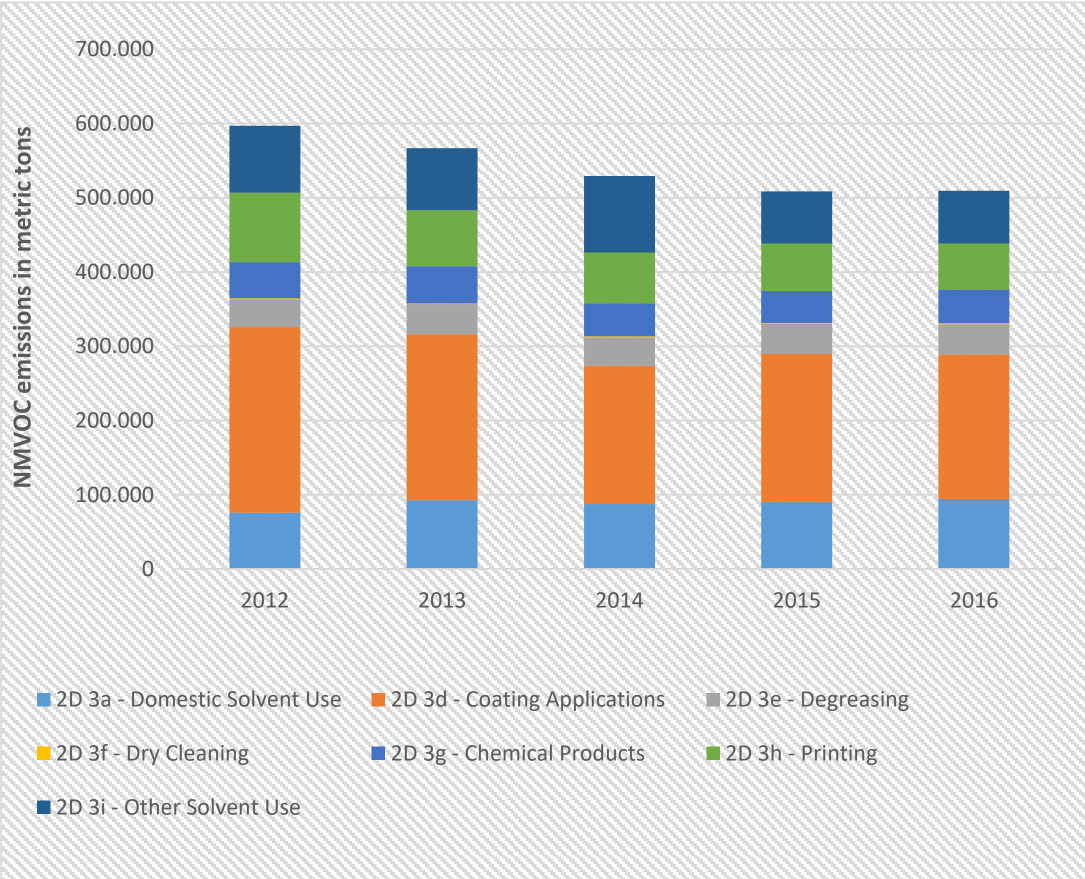
SNAP	2D 3i Other Solvent Use
60401/02	**Glass and mineral wool enduction**
60404	**Fat, eadible and non edible oil ectraction**
60405	**Application of glues and adhesives**
	22 sub-categories
60406	**Preservation of wood**
60407	**Underseal treatment and conservation of vehicles**
	2 sub-categories
60409	**Vehicles dewaxing**
	3 sub-categories
60412	**Other**
	14 sub-categories

Figure 2: Structure of the German NMVOC inventory

Based on the described inventory system, the NMVOC emissions are calculated annually and used with regard to the reporting obligations of the established legal framework.

The emissions over time with breakdown to CRF sectors are shown in the following chart.

Figure 3: Emissions over time - German NMVOC inventory



3 Approach of the ESIG inventory

The solvent-based approach used by ESIG is a methodological alternative to the product-based bottom-up approach currently used in the German solvent inventory. In the solvent-based approach, the domestically used quantities of solvents are determined top-down, distributed to different areas of use, and multiplied with emission factors.

Data availability is a central obstacle in the implementation of such an approach. Existing official statistics are neither sufficient with regard to their differentiation nor their completeness (e.g., lack of data in cased of less than three market players). Based on data from its member companies (production quantities), ESIG regularly provides emission estimates for the EU-27 and selected countries. For the country specific calculations, the domestically used quantities are calculated based on quantities reported by producers and an assumed export rate (currently 47 % are assumed for Germany).

In June 2015, key findings were published in the "Solvent VOC Emissions Inventories Position Paper" (ESIG 2015); in 2018 an updated version has been published (ESIG 2018) and in 2019 the ESIG inventory has been subject to a publication by (Pearson 2019).

Contrary to the German inventory which follows the EMEP system, the ESIG inventory is structured following 16 REACH end-use sectors, with specific emission factors for each end-use sector:

Table 1: ESIG Inventory: Reach end-use sectors and emissions factors (ESIG 2018)

#	End-use sector	Emission factors
1	Agrochemical Uses	100%
2	De-Icing	100%
3	Blowing Agents	100%
4	Binder and Release Agents	100%
5	Industrial Cleaning	70%
6	Professional Consumer Cleaning	50%
7	Industrial, Professional, and Consumer Coatings	75%
8	Functional Solvents	10%
9	Metal working/ Rolling Oils/ Lubricants Uses	0%
10	Oil field chemicals – Drilling-Mining-Extraction	0%
11	Polymers Processing (incl. rubber-tyre production)	10%
12	Road and Construction	95%
13	Use as Fuel/ Combustion	0,25%
14	Water Treatment	5%

15	Other Consumer Uses (household, aerosols, cosmetics)	90%
16	Pharmaceuticals Manufacturing	30%

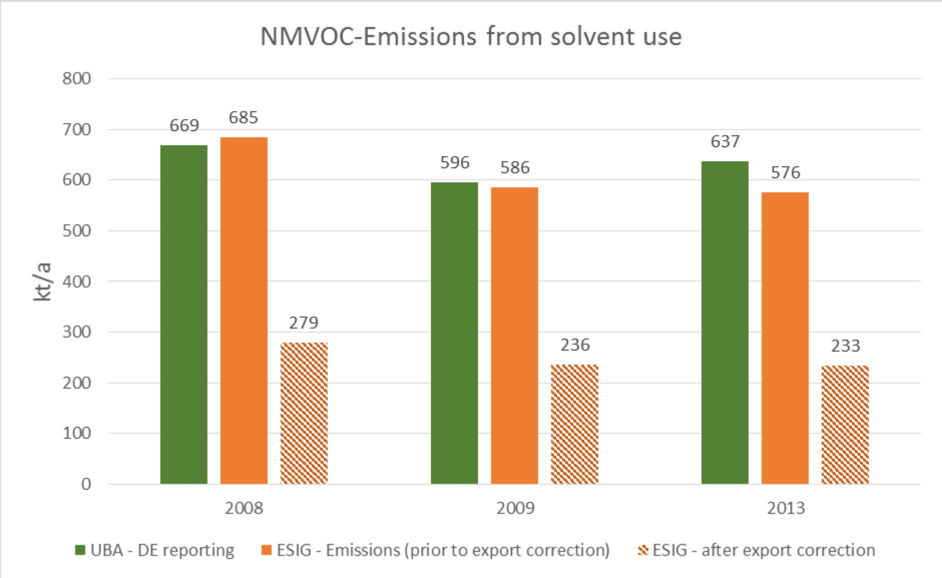
Based on the data provided by the member companies, the assumed exports and the emission factors, for Germany, VOC emissions of 325 metric kilo tons have been calculated for the year 2015 (Pearson 2019).

4 Comparison of results and need for a closer look

The ESIG emission estimates clearly differ from the EU-27 environmental agencies' reports as well as the emissions reported for Germany by the UBA, which are calculated using the inventory system described above. I.e., for 2015 the German inventory provides an emission estimate of 508 kt compared to 325 kt in the ESIG reports.

A detailed comparison for the years 2008, 2009 and 2013 using data provided by ESIG is shown the following picture.

Figure 4: NMVOC-emissions from solvent use in the German inventory and the ESIG model



It is noteworthy, that the values from the UBA inventory and the values of ESIG correlate very well until the export correction in the ESIG inventory is done which results in significant differences for all considered years. As outlined above, in the German inventory an export-correction is done, as well

Based on an assessment of available information and an exchange with ESIG, the differences have been analysed in more detail.

4.1 Comparison of emission factors

A comparison of emission factors from both inventories shows some minor differences but in general does not provide an explanation for the differences in the emission calculations.

An exemplary comparison of selected emission factors from both inventories is presented in the following table. A one-to-one comparison is not possible as the German inventory is significantly more differentiated. However, emission factors appear to be in comparable ranges for the key sources of VOC emissions.

Table 2: Emission factors in German Inventory and ESIG model

End-use	ESIG	DE
Metal working/ lubricants	0%	3-10%
Use of pharmaceuticals	30%	50%
Dry Cleaning	50% (Professional consumer cleaning)	95%
Polymer processing (incl. Rubber)	10%	Polymer specific PVC: 0,5% PU: 7,8% PS foam: 50% Rubber: 90%
Coatings	70%	5-95%

4.2 Comparison of 2015 emissions for Germany

In Annex 1 to the chapter to 2D 3a of the EMEP Guidebook (EEA 2016) a mapping table is provided to link the REACH classification (as used in the ESIG inventory) to the CRF categories. It is emphasized in the Annex that the table presents a default link table and country-specific information should be used whenever available.

Table 3: Mapping table: REACH end use sectors to CRF categories (from EMEP Guidebook, Annex 1 to 2D 3a)

	2D3a	2D3b	2D3c	2D3d	2D3e	2D3f	2D3g	2D3h	2D3i
Agrochemical Uses	100%								
Blowing Agents									100%
De-Icing	50%								50%
Binder and Release Agents									100%
Industrial Cleaning					100%				
Professional Consumer Cleaning	100%								
Coatings- industrial and adhesives, inks				80%				15%	5%
Coatings – professional/ consumer and thinners, paint industry	30%			70%					
Functional Solvents							100%		
Metal working/ Rolling Oils/ Lubricants Uses									100%
Oil field chemicals – Drilling-Mining-Extraction									100%
Polymers Processing (incl. rubber-tyre production)							100%		
Road and Construction		100%							
Use as Fuel/ Combustion									100%
Water Treatment									100%
Other Consumer Uses (household, aerosols, cosmetics)	100%								
Pharmaceuticals Manufacturing							100%		
Others									100%
Chlorinated solvents							100%		

Using this mapping table, the results from both inventories for the year 2015 are compared: The ESIG results for the different REACH sectors are allocated to the CRF sectors using the respective factors. Then the emissions are compared to the respective emissions from the German inventory.

Table 4: Breakdown of emissions to CRF codes

CRF	Emission in 2015 according to German Inventory (UBA) [metric tons]	Emissions in 2015 according to ESIG using mapping table [metric tons]	Difference [metric tons]
2D 3a	91.502	50.424	41.078
2D 3b	0	3.316	-3.316
2D 3c	0	0	0
2D 3d	199.332	152.346	46.986
2D 3e	40.876	10.638	30.238
2D 3f	1.182	0	1.182
2D 3g	42.882	39.971	2.911
2D 3h	64.027	23.603	40.424
2D 3i	101.337	45.213	56.124

This comparisons shows that the UBA inventory provides significantly higher emissions in the categories of Domestic solvent use, Coating applications, Degreasing, Printing, and Other solvent use. At the same time, both inventories do not differ significantly with regard to the distribution of the emissions to the different CRF sectors. Coating applications have a somewhat higher relevance in the ESIG inventory (47% compared to 37%) while degreasing is more relevant in the UBA inventory (8% compared to 3%). All other categories differ less than 5%.

4.3 Assessment of export factors

In the ESIG inventory, a general export factor is applied to the solvents produced in Germany. In the German inventory, the quantities placed on the market are determined using domestic statistical data. As the export of solvents (and hereby of emissions) presumably is a key aspects with regard to the differences between ESIG and UBA calculations, a closer assessment of the relevance of exports and of the export factors with regard to the German inventory is done in the following.

In a first step, for both inventories the emissions before the consideration of any (imports and) exports have been calculated. In the second step, the statistical data used in the German inventory has been analysed in detail and where possible, export factors specific per CRF codes have been calculated. Conducting this analyses was not possible for the entire inventory, but for the main categories.

Table 5: Main findings from CRF category analysis

CRF	Calculated export factor	Difference after export correction	Difference before export correction
2D 3a	18 % (products) 53 % (solvents)	41.000 t	22.000 t
2D 3d	25 %	50.000 t	20.000 t
2D 3e	11 %	30.000 t	26.000 t
2D 3f	-	No comparison possible, but low overall significance.	
2D 3g	For 4 SNAP codes available: 60301: -206 % 60302: 43 % 60303: 20 % 60304: 7,4 %	2.900 t	Export rates could not be determined for all SNAP codes considered in the German inventory. The export rate for SNAP 60301-4 however indicate the 47 % assumed in the ESIG

CRF	Calculated export factor	Difference after export correction	Difference before export correction
			model might result in an underestimation of emissions.
2D 3h	42 %	40.400 t	57.200 t
2D 3i	For 2 SNAP codes available: 60405: 30, 5 60406: 46,2 %	-	56.000 t

Regarding the analysis and its outcome it needs to be noted, that the mapping table from the EMEP Guidebook which has been used to allocate emissions from the REACH end use sectors to CRF categories, is a helpful tool but has certain limitation in its preciseness. One reason for this is methodologically: REACH end use sectors have a different purpose than CRF category and a precise 1:1 matching (or relative matching) is not possible. Another reason is that, the allocation depends on country specific aspects as also emphasized in the EMEP Guidebook which recommends to use country specific allocation factors when available. Such (country specific) factors, however, have not been available.

Considering this, still, the following observations can be made: The analysis indicates that the 47 % export rate of the ESIG model might be too high, leading to an underestimation of the emissions. A comparison of emissions before an export-correction leads in some cases to a significantly decreased difference between both, ESIG and German inventory. This is in particular the case for 2D 3a, d and e.

The results for these categories show, however, that even without consideration of any exports, the ESIG numbers are still somewhat lower than the numbers from the German inventory which might results from inaccuracy of the mapping table, differing emission factors and/or differences in the quantities reported by the ESIG member companies to ESIG to the quantities found in the German production and trade statistics.

5 Conclusion

Solvent or NMVOC inventories are required to fulfil the obligations set out by the existing legal framework. With regard to these obligations, the German solvent inventory has been structured in a way allowing it to be updated annually using statistical data. At the same time, the German inventory is rather detailed and differentiates between many application areas of solvents.

Compared to the German inventory with its product-based bottom-up approach, the ESIG inventory uses data provided by ESIG member companies on production amounts and distributes these to different areas of applications structured according to REACH-end use sectors. I.e., here a top-down approach is taken.

Both inventories differ significantly with regard to the calculated emissions. Analysing the potential reasons for those differences is challenging due to the different structure of both inventories. The analysis did however indicate, that the differences are to some extent caused by the export-correction done in the ESIG inventory which appears to overestimate exports at least for some sectors.

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