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European solvent VOC emission inventories based on industry-wide information

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ABSTRACT

The European Solvents Industry Group (ESIG) has established volatile organic compound emission inventories for solvents for individual countries or groups of countries within the European Union for 2008, 2009, 2013 and 2015. A top-down approach has been adopted starting with EU-wide solvents industry sales data as supplied by solvent manufacturers, to which atmospheric release factors were applied for each solvent use sector. ESIG total solvent emissions for the EU ranged from 2159 thousand tonnes year^{-1} in 2008 to 1981 thousand tonnes year^{-1} in 2015. Comparison with internationally-agreed solvent emissions suggests that official estimates of total solvent emissions across the EU are over 30% too high by comparison with ESIG estimates and showed too high a trend with time. Using official statistics on chemical exports and imports for each country, ESIG has been able to compile solvent emissions for each European country or groups of countries. The over-estimation by the internationally-agreed solvent emissions compared with the ESIG estimates for Europe as a whole was mirrored in the individual country estimates for 2008, 2009, 2013 and 2015. No simple, uniform relationship between national solvent emissions and per capita population for either total or domestic sector solvent emissions was found. Assumption of uniform solvents emission factors on a per-head of population basis, has led to the introduction of inaccuracies in internationally-agreed solvent emission inventories. More detailed work with Member States has shown that non-solvents are also being included in the official solvent VOC inventories-indeed these VOC inventories are for 'solvents and other product use'.

1. Introduction

Ground-level ozone (O_3) is a major policy concern because it is an important atmospheric pollutant that at elevated levels damages human health and vegetation (Monks et al., 2015). Policy actions within Europe to reduce O_3 levels began with the Nitrogen Oxides (NO_x) and Volatile Organic Compounds (VOC) Protocols to the United Nations Economic Commission for Europe (UN ECE) Convention on Long-Range Trans-boundary Air Pollution (CLRTAP) during the late 1980s (UN ECE, 2006). During the 1990s, the UN ECE added the Gothenburg Protocol to abate acidification, eutrophication and ground-level O_3. The European Union (EU), (formerly the Commission of the European Communities, CEC), has implemented directives controlling emissions for motor vehicles (CEC, 1991), large combustion plant, solvents (1999) and petrol evaporation (CEC, 1994). The two main bodies, the EU and the UN ECE, have set targets for emissions, including NO_x, NMVOC (non-methane VOC), sulphur dioxide (SO_2), ammonia (NH_3) and particulate matter less than 2.5 μm ($PM_{2.5}$). These take the form of ceilings on each of these emissions for all countries in the EU for 2020 and 2030 (EU, 2016).

The reductions in O_3 precursor emissions required in the early CLRTAP Protocols have been largely secured through mandatory motor vehicle emission directives (CEC, 1991). The evidence from urban VOC monitoring is that these directives have dramatically reduced VOC emissions, and urban levels in the United Kingdom, for example, have declined by close to an order of magnitude (Derwent et al., 2014). Similar declines have been reported in road transport emission inventories, demonstrating how the emission inventories have apparently captured the salient features influencing real-world VOC emissions from road transport. On this basis then, it is likely that European policies to control motor vehicle VOCs and NO_x have brought about the observed decline in episodic peak O_3 levels in North West Europe since 1990 (Derwent et al., 2010).

Although careful attention has been given by policy-makers to characterise and quantify VOC emissions from the road transport sector over the last thirty years or so, this has not been the case for other important VOC emission sectors, particularly biogenic and solvent emissions. Solvent emissions in Europe have not been well characterised and quantified and hence are not well represented in air quality policy models. As road transport emissions have declined, the

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potential role of biogenic and solvent emissions in ground-level ozone formation across Europe has grown. Whereas biogenic emissions from trees and plants cannot be subject to legislation, these VOC emissions dominate those from man-made sources. This study addresses solvent VOC emissions in the European Union but there comes a moment when industrial VOC emissions will have declined to a level where further reductions are no longer cost effective.

There is no question that the solvent sector is an important source of ambient VOCs. Source apportionment studies point to solvent evaporation sources of VOCs in China (Cai et al., 2010), Hong Kong (Ou et al., 2015), Aliaga, Turkey (Dumanoglu et al., 2014), San Diego, USA and Tijuana, Mexico (Zheng et al., 2013), Athens, Greece (Moschonas and Glavas, 1996), Kaohsiung, Taiwan (Lai et al., 2005), Helsinki, Finland, (Hellen et al., 2003), Lille, France (Borbon et al., 2003), Wuppertal, Germany (Niedojadlo et al., 2007) and Houston, Texas (Leuchner and Rappengluck, 2010). In reviewing many of the early source apportionment studies, Watson et al. (2001) concluded that the solvent contributions found in these observational studies carried out in more than 20 urban areas mostly in the United States of America were often much lower than those reported in emission inventories. This has been shown subsequently to be the case in other studies, including, for example, in Wuppertal, Germany (Niedojadlo et al., 2007). On this basis then, it is certainly plausible that current solvent emission inventories may not be a reliable guide to actual real-world solvent emissions. However, it must be said that these two studies were performed 15–20 years ago and provide only limited coverage on the European scale.

In view of the disagreement between the European source apportionment studies and the national solvent emission inventories detailed above, the European Solvent Industry Group (ESIG) set about bringing their expertise to bear on solvent emission inventories across Europe. A number of different approaches were discussed within the industry and it was decided to build a top-down emission inventory based on solvents sales data to be provided by the European solvent industries themselves. These solvents include hydrocarbon, oxygenated and chlorinated solvents but currently exclude ethanol and methanol which are outside the scope of ESIG. It should be noted that chlorinated solvents exhibit low reactivity and so could, realistically, be excluded from inventories used to describe ozone formation. This study describes how industry-wide statistical information has been employed to compile top-down solvent emission inventories for Europe for 2008, 2009, 2013 and 2015.

2. Methods

The European Solvents Downstream Users Group supported by the ESIG set about compiling a top-down solvents emission inventory for Europe. For this initiative to produce an accurate contribution to the understanding of the role of solvents in air quality, a number of issues had to be agreed and resolved at the outset within the European solvent industries themselves. Details of the procedures adopted are given in the Supplementary Information attached to this paper.

It was important that the solvent sales data remained confidential and that there were no breaches of confidentiality in the handling of small countries and their suppliers. A definition of a solvent that was unambiguous and rigorous and would ensure consistent statistical information across the industry was agreed as follows: a solvent is an organic compound or mixture of organic compounds that is a liquid with a vapour pressure greater than 133 Pa at standard temperature and pressure. Additional definitions were unified for solvent speciation, solvent atmospheric release factors and end-uses (see the Supplementary Information attached to this paper). Solvent sales information was to be collected on a by-mass basis. The statistics were reported by end-use sector in the same market sectors as reported in the Generic Exposure Scenario (GES) of the Registration, Evaluation and Authorisation of Chemicals Regulation (REACH) (ECHA, 2013) (see Supplementary Information) for both hydrocarbon and oxygenated solvents.

Table 1
Total European solvent emissions based on industry-wide solvent sales statistics compared with internationally-agreed solvent emissions for the European Union.

Year	ESIG total solvent emissions, thousand tonnes year^{-1}	Internationally-agreed total solvent emissions, thousand tonnes year^{-1}
2008	2159	3335
2009	1917	3050
2013	1978	2928
2015	1981	2694

Essentially each solvent supplier was asked to provide their sales volumes to each EU country for each REACH sector. Details of companies surveyed are given in the ESIG website (www.esig.org). The confidential questionnaire responses were collected by the statistical section of the European Chemical Industry Council or Cefic (from its former French name: Conseil Europeen des Federations de l'industrie Chimique) for the calendar years 2008, 2009, 2013 and 2015. The sales data were then aggregated up into groups of countries by the Cefic statisticians so that a minimum of three companies (five companies in 2015) were supplying data to each country or block of countries. These aggregated data were then released to ESIG for the preparation of their top-down solvents emission inventories. A further level of aggregation was then applied to give total European solvent emissions for the EU-27 (EU-28 in 2015) for each year across all the reporting companies. These total European solvent emissions are presented in Table 1.

ESIG total solvent emissions for the European Union ranged from 2159 thousand tonnes year^{-1} in 2008 to 1981 thousand tonnes year^{-1} in 2015, as seen in Table 1. This apparent decline amounted to about 8% which is comparable with or slightly greater than the estimated uncertainty in the ESIG statistical data (estimated as ± 7%). The decline partly reflects the recession period and partly the completion of the Solvents Emissions Directive (Commission of the European Communities, 1999) over the period from 1999 to 2009. A number of additional assumptions were made in the compilation of Table 1. It was assumed that the companies surveyed provided almost 100% coverage of solvent sales across the EU and that there was no import or export of solvents across the boundaries of the EU. Chemicals export data does show a net export of chemicals from the EU to the Russian Federation and the United States of America but ESIG does not have detailed information on net exports of solvents to these countries. Assuming no movement of solvents into or out of the EU ensures that solvent VOC emissions within the EU have not been underestimated.

Also shown in Table 1 are the internationally-agreed total solvents emissions for the EU provided by the EU member states to the LRTAP Convention and compiled by the European Environment Agency (EEA) (2016). The internationally-agreed total solvent emissions amounted to 3335 thousand tonnes year^{-1} in 2008 and 2694 thousand tonnes year^{-1} in 2015. The ESIG emission estimates are between 63% and 74% of the EEA estimates. It appears that not only do the EEA estimates overestimate solvent emissions by a large factor but they also overestimate the decline is solvent emissions since 2008, reporting a 19% decline rather than 8% from ESIG. On this basis then, it is certainly plausible that current internationally-agreed solvent emission inventories may not be a reliable guide to actual real-world solvent emissions.

3. Total solvent emissions from individual countries with Europe

Solvents emissions by country are presented in Tables 2–5 relating to the years 2008, 2009, 2013 and 2015 respectively. For statistical confidentiality reasons, emissions from countries with less than three representative solvent companies on the market, (extended to five in

Table 2
Comparison of ESIG total solvent emissions with the internationally-agreed solvent emissions for 2008 in thousand tonnes year^{-1}.

Country	European Solvents Industry Group					EEA
	HC + OXY	CHLOR	TOTAL before I/E	Import/(Export)	Grand TOTAL	TOTAL
Austria + Slovenia	37.5	2.9	40.4	45.7	86.1	101
Belgium + Luxemburg	143.1	11	154.1	(77.5)	76.6	57
Bulgaria + Romania	4.3	0.3	4.6	28.1	32.7	80
Cyprus + Greece	41.8	3.2	45.0	40.7	85.7	59
Czech Republic + Slovakia	17.6	1.4	19.0	41.4	60.4	124
Denmark	16.6	1.3	17.9	13.5	31.4	27
Baltic States + Finland	23.0	1.8	24.8	25.8	50.6	68
France	248.5	19.1	267.6	0.0	267.6	386
Germany	636.1	48.8	684.9	(405.8)	279.1	669
Hungary	6.2	0.5	6.7	17.1	23.8	23
Ireland	17.7	1.4	19.1	0.0	19.1	24
Italy	193.4	15	208.4	147.0	355.4	466
Netherlands	129.9	10	139.9	(83.0)	56.9	57
Poland	39.2	3	42.2	67.8	110.0	198
Portugal + Spain	206.5	15.9	222.4	106.0	328.4	531
Sweden	35.0	2.7	37.7	33.2	70.9	84
United Kingdom	208.3	16	224.3	0.0	224.3	379
Total	2005	154	2159	0	2159	3335

2015), were combined. Due to the free flow of goods in the European Union, there are no direct data for import and export of solvents within the EU and so an estimate has to be made. Cefic holds data for chemical exports and imports for each country (EUROSTAT, 2018) so that the net chemical transfer can be calculated for each Member State. By assuming this percentage of chemical transfer is directly proportional to percentage of solvent movements, an estimate of solvent import/export related to solvent downstream activities has been made for each country, or group of countries, in the EU. This assumption has been discussed with Member States in some detail, as highlighted in the Acknowledgements below. These calculations assume that there is no net import or export to or from the European Union.

These data confirm that Belgium, Germany and Netherlands are substantial exporters of solvents and downstream products (as such "exporting" VOC emissions) within the EU and are included in Tables 2–5. The UK and France are effectively in balance with Ireland an anomaly (due to the import and export of pharmaceutical products) and it is assumed that there is no net import or export of solvents for these three countries. The remaining EU Member States are net importers of solvents, and although they export and import to one another, their total net imports are assumed to equal the exports from Belgium, Germany and the Netherlands. The final column in Table 2–5 shows the total solvent VOC emissions taking account of the import/export adjustment.

Tables 2–5 also show the nationally-reported solvent emissions tabulated by the EEA (2016) for each of the EU Member States. For some Member States in 2008 (see Table 2), total solvent emissions reported by the EEA are smaller than those reported by ESIG. Such Member States are Belgium + Luxembourg and Cyprus + Greece. However, for the majority of Member States the ESIG emissions are substantially lower than those reported by the EEA (2016). This is particularly the case for the Member States with high solvent emissions, namely Germany, Italy and Portugal + Spain. The overestimation by the internationally-agreed solvent emission compared with the ESIG estimates noted in Table 1 for Europe as a whole is mirrored in Table 2–5. Overestimation of solvent emissions appears to be a Europe-wide phenomenon and is not restricted to particular geographical regions or countries.

A similar pattern of overestimation by the EEA (2016) compared with ESIG is found in Table 3 for 2009 particularly for Germany, Italy and Portugal + Spain. The solvent emissions reported by the EEA (2016) for Italy fell strongly between 2009 and 2013 bringing them into

Table 3
Comparison of ESIG total solvent emissions with the internationally-agreed solvent emissions for 2009 in thousand tonnes year^{-1}.

Country	European Solvents Industry Group					EEA
	HC + OXY	CHLOR	TOTAL before I/E	Import/(Export)	Grand TOTAL	TOTAL
Austria + Slovenia	35.2	2.9	38.1	40.9	79.0	77
Belgium + Luxemburg	132.6	10.8	143.4	(68.8)	74.6	50
Bulgaria + Romania	5.8	0.5	6.3	39.8	46.1	56
Cyprus + Greece	34.3	2.8	37.1	38.5	75.6	59
Czech Republic + Slovakia	14.9	1.2	16.1	35.7	51.8	120
Denmark	14.5	1.2	15.7	1.7	17.4	27
Baltic States + Finland	19.9	1.6	21.5	17.6	39.1	59
France	207.9	16.9	224.8	0.0	224.8	349
Germany	541.6	44	585.6	(349.9)	235.7	596
Hungary	6.1	0.5	6.6	9.9	16.5	17
Ireland	14.8	1.2	16.0	0.0	16.0	23
Italy	181.8	14.8	196.6	137.4	334.0	436
Netherlands	133.3	10.8	144.1	(88.9)	55.2	54
Poland	42.8	3.5	46.3	64.5	110.8	201
Portugal + Spain	171.9	14	185.9	97.0	282.9	485
Sweden	27.5	2.2	29.7	24.4	54.1	87
United Kingdom	187.9	15.2	203.1	0.0	203.1	353
Total	1772	144	1916	0	1917	3050

Table 4
Comparison of ESIG total solvent emissions with the internationally-agreed solvent emissions for 2013 in thousand tonnes year^{-1}.

Country	European Solvents Industry Group					EEA
	HC + OXY	CHLOR	TOTAL before I/E	Import/(Export)	Grand TOTAL	TOTAL
Austria + Slovenia	42.5	3.2	45.7	50.6	96.3	83.0
Belgium + Luxemburg	113.6	8.6	122.2	(71.1)	51.1	41.2
Bulgaria + Romania	6.1	0.5	6.6	48.8	55.4	90.8
Cyprus + Greece	33.4	2.5	35.9	29.3	65.2	59.0[a]
Czech Republic + Slovakia	17.6	1.3	18.9	52.5	71.4	102.8
Denmark	16.3	1.2	17.5	2.3	19.8	29
Baltic States + Finland	24.9	1.9	26.8	19.1	45.9	98.5
France	229.3	17.4	246.7	0	246.7	318
Germany	535.1	40.5	575.6	(342.5)	233.1	637
Hungary	6.7	0.5	7.2	7.2	14.4	38
Ireland	18.0	1.4	19.4	0.0	19.4	20
Italy	194.8	14.7	209.5	139.2	348.7	363.9
Netherlands	157.0	11.9	168.9	(102.8)	66.1	56.2
Poland	49.7	3.8	53.5	71.2	124.7	208.8
Portugal + Spain	188.3	14.3	202.6	79.2	281.8	352.7
Sweden	38.7	2.9	41.6	17.0	58.6	86.2
United Kingdom	166.4	12.6	179.0	0.0	179.0	342.7
Total	1838.4	139.2	1977.6	0	1977.6	2927.7

Notes.
[a] Estimated EEA emissions based on 2009 data.

alignment with those from ESIG. However, the general overestimation by the EEA (2016) in 2013 compared with ESIG remained, viz: Table 4, and persisted into 2015, see Table 5.

The present ESIG (top-down) study based on solvent sales in the EU indicates that internationally-reported EEA (bottom-up) inventories substantially overestimate EU solvent VOC emissions by 35% in 2008, 37% in 2009, 32% in 2013 and 26% in 2015. This confirms what had been concluded from the limited number of source-apportionment studies reported in the literature as discussed above. The availability of the solvents release factors from REACH in 2010 has helped ESIG improve the solvents industry statistical data. The ESIG calculations are based on data received from solvent suppliers and ESIG represents over 95% of all suppliers in the EU. It is estimated that the maximum error that could be made is a ± 5% error in total solvent volumes for a given country. A second inaccuracy could be in the atmospheric release factors used. Whilst these have been based on expert judgement, it is possible that an error of ± 5% could occur. Whilst every effort has been made not to underestimate ESIG solvent emissions (for example, 100% of chlorinated solvents are assumed to be released into the air), it is possible that the ESIG estimated emissions could be uncertain by ± 7%, from a consideration of the two possible sources of error outlined above and taken in quadrature.

Since, 2016, ESIG has been liaising with a number of Member States to check in more detail why the ESIG results differ from the internationally-agreed figures, as summarised in Tables 2–5. Member States were also invited to provide information on the import/export of solvents, if available. In these discussions it was clear that the bottom-up approach was more prone to total error than the top-down approach of ESIG because there are more processes in the calculations. Member States were obliged to take account of activity data, chemical use, percentage of solvents in chemical products and emission factors, often from different reference sources, see for example, for Denmark (Fauser and Illerup, 2008). Furthermore, it was clear that some Member State inventories included liquefied gases: butane and propane, which are

Table 5
Comparison of ESIG total solvent emissions with the internationally-agreed solvent emissions for 2015 in thousand tonnes year^{-1}.

Country	European Solvents Industry Group					EEA
	HC + OXY	CHLOR	TOTAL before I/E	Import/(Export)	Grand TOTAL	TOTAL
Austria + Slovenia + Croatia	40.5	3.2	43.7	34.4	78.1	90.0
Belgium + Luxemburg	149.0	8.6	157.6	(104.5)	53.1	42.0
Bulgaria + Romania	4.7	0.5	5.2	43.7	48.9	88.0
Cyprus + Greece[a]	27.5	2.5	30	19.3	49.3	2
Czech Republic + Slovakia	17.3	1.3	18.6	51.7	70.3	113.0
Denmark	11.3	1.2	12.5	7.5	20.0	27
Baltic States + Finland	20.9	1.9	22.8	31.5	54.3	49.0
France	200.9	17.4	218.3	0	218.3	289
Germany	568.5	40.5	609	(284)	325.0	541
Hungary	6.1	0.5	6.6	17.2	23.8	41
Ireland	16.4	1.4	17.8	0.0	17.8	19
Italy	190.5	14.7	205.2	136.3	341.5	353
Netherlands	169.4	11.9	181.3	(121)	60.3	56.0
Poland	38.2	3.8	42	69.5	111.5	219
Portugal + Spain[b]	195.7	14.3	210	82.1	292.1	360
Sweden	24.3	2.9	27.2	16.3	43.5	57
United Kingdom	160.6	12.6	173.2	0.0	173.2	350
Total	1840.8	139.2	1981	0	1981	2694

Notes.
[a] No data available from Greece.
[b] Spain and Portugal can be separated out to give totals of 272.3 and 19.8ktonnes respectively.

both propellants and solvents in aerosol dispensers, which explains some discrepancies with the ESIG estimations. Another concern is that C_{14+} compounds are too heavy to be classed solvents because of their intrinsically low vapour pressures but were included in some inventories. Many Member States are providing an inventory of solvent VOC Emissions combined with "other Product Use". Discussions continue with these Member States as to how to separate out "other product use".

All these issues could lead to an overestimate of 30% or more in solvent VOC emission inventories by some Member States.

4. Discussion and observations

It is important to understand why solvent VOC emissions in many Member States have been overestimated. Emission inventory estimations for Germany, for example, would align with the ESIG results if there were no exports of solvents from Germany to other Member States. However, Niedojadlo et al. (2007) have shown that solvent VOC emissions in Wuppertal, Germany are overestimated by some 60%. In the absence of new measured data from Germany, it is assumed that the same overestimation applies to German solvent VOC inventories from 2008 to 2015. Removing this overestimation could bring the EEA estimates for Germany more into line with the ESIG estimations. Also, the reported UK solvent VOC emissions are too high by comparison with those from ESIG, especially given that the UK is assumed not to export solvents. The UK inventory contains liquefied gases used as both propellants and solvents in aerosol dispensers, so not covered by the ESIG estimations, as well as chemicals classified as "other product use".

It is clear that Member States do not use the same basis for calculating their solvent VOC emissions. Some still apply guesswork as they cannot obtain detailed information on solvents for their country. Outstanding questions remain as to whether it is appropriate for Member States to use some default total or domestic sector solvents emission factors per capita or whether there are important variations in per capita emissions across the EU? These matters are being addressed where ESIG has met and organised a webinar with Member States and held discussions with individual Member States (see Acknowledgements).

ESIG cannot endorse the average factors of kg/capita emissions proposed by EEA for the domestic sector as reflecting reality. Each country has a different Gross Domestic Product (GDP) and culture. Some countries cannot afford expensive cosmetics or cleaning agents, as examples. To illustrate these differences, ESIG has calculated total and domestic sector solvent emission factors on a per capita basis for the countries/group of countries in this study using the 2013 solvents emissions data. These results are shown in Table 6 below. Per capita emission factors vary considerably from country to country. They also vary annually as could be seen if the exercise were repeated for 2015. Also shown in Table 6 are the domestic sector solvents emissions on a per capita basis. Domestic solvents emissions factors vary from 0.5 to 2.4 with an average value of 1.1 kg per head per year for the EU. Each country or group of countries has its own individual factor and so the EU cannot be grouped into sets with similar factors.

Data on the populations of each country (capita) and their total solvent emissions from Table 6 were assembled and a linear regression of the form:

$$\text{Country solvent emissions} = a + b.\text{capita} \quad (1)$$

was constructed, where a and b were constants, taking the values 11.4 ± 18 thousand tonnes per year and 3.5 ± 0.5 thousand tonnes per year per million head of population, respectively. This simple linear regression accounted for about 80% of the variance in the country solvent emissions data. This provides some support for the commonly-assumed solvent emission factors based on widely available population statistics (see, for example, the global VOC emission inventory from Crippa et al., 2018). However, inspection of the residuals showed that for about half of the countries, this simple regression approach left many spatial features of the European solvent emissions unanswered. The residuals above showed a clear north-south divide, with the more northerly countries overestimated by the regression and the more southerly countries underestimated. Using the latitude (lat) data for the geographical centre of gravity for each country and an expression of the form:

$$\text{Country solvent emissions} = a + (c + d.\text{lat}).\text{capita} \quad (2)$$

a relationship was constructed where a, c and d were constants, using non-linear regression software. The values of the constants were found to be: a = 20.2 ± 12 thousand tonnes per year; c = 12.3 ± 2 thousand tonnes per year per million head of population and d = −0.19 ± 0.04 thousand tonnes per year per million head of population per degree latitude. All were highly statistically significantly different from zero. This new non-linear regression equation accounted for about 92% of the variance in the country solvent emissions data, much higher than the simple expression (1). Solvent emissions increased with decreasing latitude and decrease with increasing latitude because of the negative sign in the d.lat term for a given head of population.

On this basis, assuming total or domestic sector solvent emissions per head of population to be independent of country was not a good approximation because it made no allowance for the location of the country within the European continental area. Countries in the south of Europe with low latitude values have significantly higher solvent emissions per capita than those in the north with higher latitude values. This presumably reflects the higher temperatures and solar ultraviolet levels in more southerly latitudes which drive up solvent usage and solvent evaporation. The concept of solvent emissions being independent of country on a per capita basis cannot be addressed simply using an average figure for the EU and any new initiatives within the UN ECE and EU on solvent VOC emission inventories need to steer Parties and Member States towards more realistic estimates of solvent VOC emissions. In this respect, ESIG has been working with a number of Member States (see Acknowledgements) to identify reasons for the differences between official and ESIG estimates. This continued liaison is identifying how both Member States' inventories and those of ESIG

Table 6
Total and domestic sector solvent emissions per head of population in kg per capita per year based on the ESIG estimates for 2013 and on literature population data.

Country	Total solvent emissions, kg/capita/year	Domestic sector solvent emissions kg/capita/year
Austria + Slovenia	9.0	1.7
Belgium + Luxemburg	4.8	1.5
Bulgaria + Romania	1.9	0.5
Cyprus + Greece	5.9	2.4
Czech Republic + Slovakia	4.4	1.5
Denmark	3.5	0.8
Baltic States + Finland	3.9	0.7
France	3.8	1.5
Germany	2.9	0.7
Hungary	1.4	0.5
Ireland	4.1	2.1
Italy	5.7	1.4
Netherlands	3.9	1.1
Poland	3.2	0.7
Portugal + Spain	4.9	1.4
Sweden	6.0	1.3
United Kingdom	2.8	1.0
EU27 average	3.9	1.1

Notes.
a. Population data taken from the Economist (2014).

can be improved. One example is how emissions factors for paints are being revised downwards in the light of a higher proportion of water-based paints. Another example is the elimination of heavy hydrocarbons (C_{14+}) from the inventories of some Member States as these are not VOCs.

Future work will be aimed at better defining and understanding the trans-boundary transport of solvents between European Member States. An estimate has been made here using the chemicals import/export data available within Cefic for each Member State for each the years 2008, 2009, and 2013, with 2015 import/export calculations improved based on feedback from selected Member States and a modified import/export to 2013. It was clear that export of solvents from Germany had been overestimated in 2013 and those from the Netherlands and Belgium and Luxemburg had been underestimated. The import of solvents into Denmark was also modified based on official data from Denmark. The 2013 export figures for the three countries could be amended to ensure that ESIG estimates did not exceed the official solvent totals for the Netherlands and Belgium and Luxemburg. Taking the 2013 modifications into account, these data for chemical imports and exports for most of the EU-27 countries are assumed to be proportional to solvent imports and exports, and a final assessment of solvent emissions in each country has been made for 2015.

This study has excluded any import/export beyond the European Union, and, although the effect on the EU total emissions will be small, any external import may be more pronounced in specific Member States. Accurate import and export data of solvents between Member States remain the key improvement in future calculations by ESIG on solvent VOC emissions in the European Union.

5. Conclusions

ESIG has established a detailed set of solvents VOC inventories for individual countries or groups of countries within the EU for 2008, 2009, 2013 and 2015. The approach has been to start with solvents sales data supplied by each solvent manufacturer, and then apply atmospheric release factors for each of the REACH sectors (a top-down approach). Comparison with internationally published solvent emissions (a bottom-up approach) suggests that official estimates are some 30% too high by comparison with Industry estimates. More detailed work with Member States has shown that non-solvents are being included in the official solvent VOC inventories-indeed these VOC inventories are for 'solvents and other product use'. This means that when the Commission of the European Commission called for a reduction in solvents emissions (CEC, 1999), they had not taken into account that these published inventories by Member States are already substantially over-estimated.

Further work by the EEA and other bodies need to recognize that there are inherent difficulties with the concept of a single, universal average emission factor per capita for total solvent VOC emissions and for domestic sector solvent VOC emissions across the EU. Such solvent emission factors are country specific and lower than those published in the EEA guidebook on emission inventories (EEA, 2009). The factors reported here in Table 6 should replace those in the EEA guidebook.

Future work by ESIG will continue on establishing solvent VOC emissions inventories for recent years and will include better estimates of import/export of solvents between EU Member States as well as exploring the import and export of solvents into and out of the EU. The establishing of accurate solvent VOC inventories is a process of continuous improvement to which ESIG is committed.

Declaration of interest

The author declares that there are no conflicts of interest in the preparation of this manuscript.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.02.014.

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