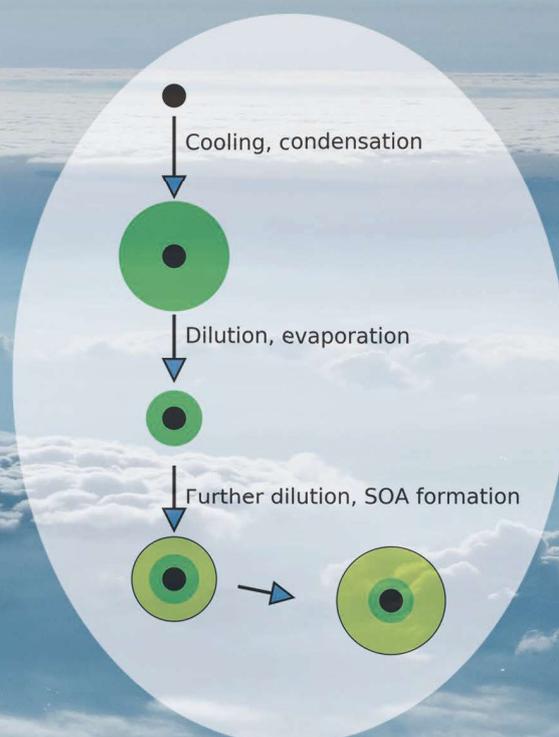


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Report of the expert workshop on condensable organics
organised by MSC-W, Gothenburg 17-19th March 2020

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Preface: Expert workshop on condensable organic compounds in PM emission inventories

In March 2020 MSC-W hosted an expert workshop on condensable organic compounds (funded by the Nordic Council of Ministers, NMR, see below), which brought together experts in emissions, measurements, emission inventories, atmospheric chemistry, air quality models and policy from Europe North America, and to create a much better understanding of the issues and possible approaches for dealing with this important class of compounds.

More than 30 experts took part in the workshop (Table A1), which was hastily re-arranged as a zoom meeting, including EMEP Chairs, inventory developers, measurement experts, industry (Concawe), the US EPA and the European Commission. The workshop discussed a number of approaches for dealing with this important class of compounds. This report presents more detail on the background to the workshop, the issues surrounding condensable organics, and presents suggestions for improving their treatment in the EMEP system.

Acknowledgements

This workshop and subsequent report were predominantly funded by the NMR's Nordic Working Group for Climate and Air (NKL). Additional contributions were made through EMEP under UN-ECE, and through the contributions of many co-authors and workshop participants. Thanks are especially due to Benjamin N. Murphy (US EPA) for valuable contributions at the workshop and during prior and subsequent discussions. Thanks also to Robert Pinder (US EPA) and the European Commission's Joint Research Centre for valuable comments on this report.

Table 1: Emissions-related acronyms

BC, EC	Black, elemental carbon
C_i^*	Effective saturation concentration at 298K, in $\mu\text{g m}^{-3}$. A measure of volatility of organic compounds.
C_{OA}	Concentration of particulate organic compounds ($\mu\text{g m}^{-3}$), e.g. in exhaust stack or ambient air
DPF	Diesel particle filter
DR	Dilution ratio (for dilution tunnel measurements)
DT	Dilution tunnel
EF	Emission factor
EI Guidebook	Emission inventory Guidebook (EMEP/EEA 2019)
ELV	Emission limit value
IIR	Informative inventory reports - part of emissions reporting to EMEP
NVOC, LVOC, SVOC, IVOC	Non-, low, semi- and intermediate- volatile organic compounds. See Sect. 2
SVOC _p , IVOC _p	particle phase of SVOC, IVOC
NEC, NECD	National Emissions Ceilings Directive
NMHC	non-methane hydrocarbons
NMOG	non-methane organic gas (NMHC plus oxygenated compounds)
NMVOC	non-methane VOC
NS	Norwegian standards (for EF measurements)
OA, OM	Organic aerosol, organic matter. Used interchangeably here for any condensed organic matter.
OC	Organic carbon, usually refers only to the condensed phase
CPM, FPM	Condensed and filterable particulate matter. These can include non-organic compounds (e.g. sulphates, ash).
POA, POM	Primary emissions of particulate OA, OM. In this report POA = FPOA + CPOA
CPOA	Condensable POA (particle phase)
FPOA	non-volatile (filterable) POA
PPM	Primary particulate matter emissions (including POM, EC, etc)
RWC	Residential wood combustion
SOA	Secondary OA, formed from oxidation of NMVOC
SP	Solid particles (non-volatile)
THC	Total hydrocarbons
VOC	Volatile organic compound

Table 2: Institute-related acronyms

<i>EMEP-related</i>	
CLRTAP	Convention on the Long Range Transboundary Air Pollution (www.unece.org/env/lrtap), also known as the Air Convention
EMEP	European Monitoring and Evaluation Programme (www.emep.int)
MSC-W	Meteorological Synthesizing Centre-West (of EMEP)
CEIP	Centre for Emission Inventories and Projections (of EMEP)
CIAM	Centre for integrated assessment Modelling (of EMEP)
GP	UN-ECE 'Gothenburg' Protocol
GAINS	IIASA's integrated assessment model: Greenhouse gas – Air pollution Interactions and Synergies
TFIAM	UN-ECE Task Force on Integrated Assessment Modelling
TFEIP	UN-ECE Task Force on Emissions Inventories and Projections
TFTEI	UN-ECE Task Force on Techno-Economic Issues
TFMM	UN-ECE Task Force on Measurements and Modelling
<i>Other</i>	
ACES	Dept. Environ. Sciences, Stockholm Univ.
CITEPA	Centre interprofessionnel technique d'étude de la pollution atmosphérique
Concawe	A division of the European Petroleum Refiner's Association
COPERT	Computer programme to calculate emissions from road transport (https://www.emisia.com)
EC	European Commission (or elemental carbon, see context)
EPA	Environmental Protection Agency
IIASA	Institute for International Applied Systems Analysis
INERIS	French National Inst. Industrial Envir. & Risks
SINTEF	Stiftelsen for industriell og teknisk forskning, Norway
TNO	Netherlands Organisation for Applied Scientific Research
UBA	German Environment Agency

Executive Summary and Key Messages

Condensable primary organic aerosol emissions are a class of organic compounds that are vapour phase at stack conditions, but which undergo both condensation and evaporation processes as the stack air is cooled and diluted upon discharge into ambient air. Emission factors measured in or close to the high-temperature high-concentration exhaust stack or pipe may misrepresent, and even miss, the amount of PM or gas that actually enters the atmosphere, depending on the filters, dilution and sampling conditions of the emission measurement. In the current emission reporting to EMEP/CLRTAP there is no clear definition of whether condensable organics are included or not, and, if included, to what extent.

In March 2020 MSC-W hosted an expert workshop on condensable organic aerosol emissions (funded by the Nordic Council of Ministers), which brought together experts in emissions, measurements, emission inventories, atmospheric chemistry, air quality models and policy from Europe and North America, and to create a much better understanding of the issues and possible approaches for dealing with this important class of compounds.

More than 30 experts took part in the (zoom) meeting, including EMEP Chairs (EMEP Steering Body, TFIAM, TFMM, TFEIP, TFTEI), EMEP Centres (MSC-W, CEIP, CIAM), inventory developers (TNO, CIAM, COPERT, and national experts from UBA - Germany, SINTEF - Norway, IVL, ACES, Swedish EPA - Sweden, CITEPA, INERIS - France, ECCC - Canada, Univ. Patras - Greece), measurement experts (PSI - Switzerland, INERIS - France, Univ. York - England, NC State University - USA), industry (Concawe), the US EPA and the European Commission. The workshop discussed a number of approaches for dealing with this important class of compounds. This executive summary presents some of the key messages from the workshop. Further background, addressing the technical matters in more detail can be found in the main body of the report.

The main idea of the workshop was to promote discussion among different communities and Task Forces that have different expertise and needs with regard to condensable organics and PM emission inventories. In order to aid these discussions, a number of important questions were identified:

1. For which source categories are condensable organics important?
2. What is included in official national and other emission inventories?
3. Do we expect emissions of condensable organics to be missing in these inventories?

4. Can we reliably predict the contribution of condensable vapours from major sources to ambient PM using data from a smaller number of representative cases?
5. Can we recommend a practical approach for inclusion (or exclusion) of condensables in (a) emission inventories, and (b) chemical transport models?

One of the ongoing major tasks for the Air Convention (CLRTAP) is the revision of the so-called ‘Gothenburg’ Protocol ¹, with a final report of the review to be completed during 2022. It is important to note that for this review process, the consideration of condensables faces several (competing) challenges:

- (a) The need for emission data as soon as possible that are consistent across countries in order to get a fair ‘optimised’ distribution of emission abatement efforts aimed at improving health and ecosystems protection targets;
- (b) The difficulties to change existing practices of some countries;
- (c) The wish of scientists to start multi-year work programs for the best possible way to define and quantify condensable emissions and/or secondary PM formation in the atmosphere.
- (d) The need to assess the available options for short and longer term actions in terms of e.g. time frame (feasibility), scientific credibility (or possible systematic bias) and costs for countries.

Key Messages

1. The current situation regarding reporting of PM emissions and condensables is untenable and unfair, in that the same activity (eg burning one unit of wood in a particular appliance type) is given very different PM emission factors in national reporting from different countries. Assumptions behind these national emission estimates are often not transparently documented, and methods can change from year to year.
2. The workshop participants agree that condensables should be included in future emission inventories and modelling. Residential Wood Combustion (RWC) emissions are a priority because of their known large contribution to PM emissions, but it is also important to take stock of other sources (e.g. road transport) that might prove to be important.
3. The issue is not just “are condensables included or not?”, but “how are they included?”. The issues are complex, with emission factors (EFs) for condensables depending on a large number of factors, including measurement methods, sampling temperatures and concentrations of total organic aerosol (C_{OA}), sampling fuels, usage, and even ambient conditions.

¹1999 UN-ECE Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, <https://unecce.org/gothenburg-protocol>

4. There is a clear need for clarification and standardisation of the methods used to define and report PM emissions. Ideally, EFs could be reported for a set of standard condition (e.g. temperature, OA concentration and/or dilution ratio), but for sources where that is not feasible conversion factors need to be developed which would make the PM emissions more comparable within the EMEP/CLRTAP system.
5. There is a clear need to increase the knowledge about activity statistics in national inventories and also for example in TNO and IIASA methodologies. For example, the appliance type (for RWC) has a major impact on the emission factor. A good overview of appliance types by country and the amount of solid fuel being burnt by appliance type would reduce uncertainties – or at least make them quantifiable.
6. Current emission limit values for residential heating (e.g. in the ecodesign directive) have not been designed with air pollution emissions in mind, and omit the condensable component. Options to better align these standards with air quality and health targets need to be investigated and defined (promising examples were presented in the workshop)
7. The current split of emissions reporting into either PM or NMVOC is problematic, in that some semi- and intermediate-volatility VOC (SVOC, IVOC) compounds could belong to either or both categories depending on ambient conditions. Further, some of these S/IVOC compounds may fall into an intermediate volatility range that is between the two conventional PM and NMVOC categories, and not be accounted for. Although probably small in mass emissions (especially compared to NMVOC), S/IVOC emissions may contribute substantially as precursors of secondary organic aerosol. Any new measurement and inventory framework should be made with this in mind.
8. Research-grade modelling is using the ‘Volatility Basis Set’ (VBS) framework to represent the full range of compounds, including S/IVOC. Ideally we would deal with organic emissions as a spectrum ranging from non-volatile PM components to gases under atmospheric conditions, with the inventory providing emissions for each VBS bin, and/or for explicit VOC compounds.
9. An interim solution whereby countries report the condensable fraction separately from the solids using consistent (or at least clearly specified) methods would aid transparency, and make it easier to compare and contrast country estimates. This might enable use of condensable EFs developed in some countries to gap-fill emissions in countries lacking in-country estimates of condensables.
10. The workshop agreed that the TNO Ref2 emissions provide a good no-regret step towards a harmonised emission methodology, but that these top-down estimates should be increasingly replaced by national estimates once procedures for quantifying condensables in a more harmonised way are agreed on and implemented.
11. Such improvements will need detailed discussion among the emission inventory communities (e.g. TFEIP, TFTEI, national experts) as well as with modellers who will have to account for the complex issues regarding volatility within the condensables and PM fractions.

12. Longer-term

- (a) Consider how to deal with organic emissions as a spectrum ranging from very low-volatility (always condensed in the atmosphere) components to volatile gases, with the inventory providing emissions for each volatility class (or explicit VOC compound).
 - (b) Explore use or extension of the US SPECIATE database which is very extensive (including both volatility classes and explicit VOC speciation for some sources).
 - (c) There is a need for a major effort through a scientific cooperation between several research groups to find the estimates for different sectors, appliances etc., in order to support a better science-based emission database. The issue should warrant for example a major EU project, open to many groups, in order to develop best possible emission estimates but also to identify and estimate uncertainties.
 - (d) Consider the use of an effective ambient PM emission factor, PMEA, which defines emissions at a standard temperature and ambient C_{OA} concentration and/or dilution ratio.
13. A road-map (see Fig. 5.1) was suggested to bridge the short and long-term scales, with a cyclic approach:
- (a) In year 1 the TNO Ref2 data is used in an initial estimate for residential combustion emissions, with modellers making educated choices about SVOC emissions and the VBS framework
 - (b) In subsequent years these top-down estimates should be increasingly replaced by national estimates once procedures for quantifying condensables in a more harmonised way are agreed on and implemented.
 - (c) Approach/updates should be tied to EMEP TFEIP meetings, with successive improvement of the reporting and transparency of the activity data and emission factors used.
 - (d) This process needs guidance and support! Voluntary contributions will lead to new mixtures of inconsistent assumptions.
14. The workshop recognises that the proposal to put larger focus on condensables might have policy implications and asks policy makers to consider possible implications with respect to potential adjustments of policy targets and base-year emissions.

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1. Introduction

1.1. Condensables - overview

As discussed in Robinson et al. (2010) primary particulate matter (PPM) is comprised of directly emitted particle mass plus any material that condenses into the particle phase without undergoing chemical reactions, though, as we will discuss in detail below, estimation of emission factors (EFs) for such emissions are also impacted by evaporation of some of the compounds. The organic component of PPM emissions is usually referred to as POM (primary organic matter) or, somewhat more commonly, primary organic aerosol (POA), which in turn consists of non-volatile (filterable) organic matter (FPOA), and the particle phase of ‘condensable’ organic aerosol (CPOA), i.e.:

$$\text{POA} = \text{FPOA} + \text{CPOA}$$

The CPOA are a class of compounds of low volatility that are vapour phase inside the flue stack (or exhaust), but which may partition between the gas and particle (condensed) phase upon cooling and dilution. Such compounds may or may not be included in current emission inventories for fine particulate matter ($\text{PM}_{2.5}$) and PM_{10} , as estimates of emission factors (EFs) depends on a wide range of factors, including sampling and analytical approaches, and assumptions about fuels and usage. As will be seen in Sect. 3 the treatment of such factors and resulting EFs varies from country to country and from one emissions source to another.

PPM can also be divided into so-called filterable (solid) PM, denoted FPM, and condensable compounds, denoted CPM. The FPM fraction includes soot/black carbon (BC), ash, FPOA, and other compounds. The CPM fraction includes inorganic compounds (mostly sulphates from sulphur present in fuels) and CPOA.

High sulphur fuel oil used in shipping or high sulphur coal burning in stoves or combustion plants could result in substantial condensable inorganic CPM, but with current low sulphur fuels (including wood) used in Europe the organic condensable fraction (ie CPOA) is dominant and the subject of this report.

As noted in Feng et al. (2018), the CPM emission issue was recognised by the EPA as early as 1983. In terms of the chemical transport modelling currently used for assessments of ambient $\text{PM}_{2.5}$, Donahue et al. (2006) and Robinson et al. (2007) highlighted the importance of addressing the volatility of POA emissions, and provided a framework for dealing with this

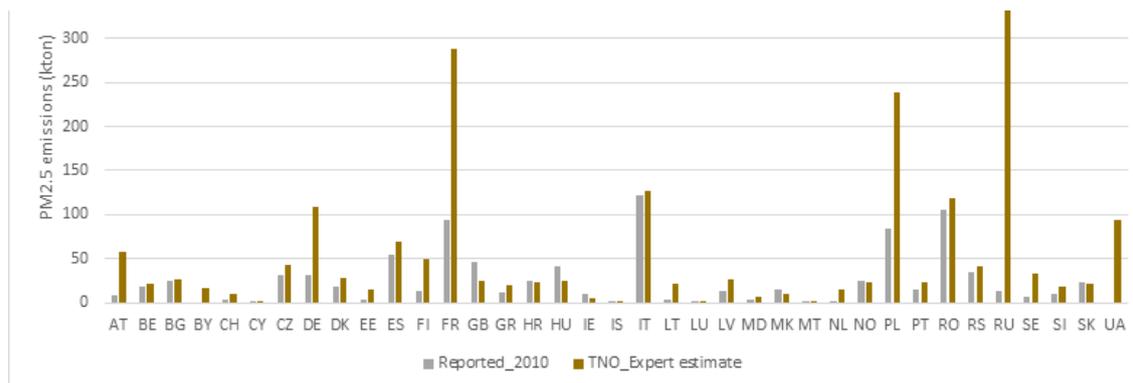


Figure 1.1: Reported $PM_{2.5}$ emissions from small-scale combustion for 2010 compared to TNO expert estimate where CPOA are included. Updated from TFEIP/TFMM (2018). (Note that differences in estimates are also affected by factors other than CPOA, see Sect. 4.3.)

in both experimental and modelling studies (the ‘VBS’, see Sect. 2). Thorough overviews of the underlying principles and relationships with emissions can be found in Robinson et al. (2010) and (for biomass-burning appliances) in Nussbaumer (2010).

The importance of CPOA for the European situation and EMEP was highlighted in Denier van der Gon et al. (2015), Simpson and Denier van der Gon (2015), Simpson et al. (2019), and Fagerli et al. (2020), who also found that condensables had significant implications for the modelling of organic aerosol and therefore PM levels in the European atmosphere. Among other problems, the different definitions of PM emissions result in inconsistent PM modelling (e.g. Bergström et al. 2012) and source-receptor relationships between countries (Simpson et al. 2019, Fagerli et al. 2020). Bergström et al. (2012), Couvidat et al. (2012) and Simpson et al. (2019) found that accounting for the gas-phase fraction of semi-volatile VOC (SVOC) significantly increases organic PM concentrations, particularly in winter, in better agreement with observations. In addition, these problems with the organic carbon (OC) fraction of European PM inventories are interlinked with those of elemental carbon (EC) since some countries apply OC/EC ratios to derive EC emissions from PM and the assumed carbonaceous fraction of PM, without knowing if CPOA are included or not in the PM EFs. So-called ‘intermediate volatility’ organic compounds (IVOC) can also contribute to CPOA, as well as being precursors to secondary organic aerosol (SOA) (Donahue et al. 2006, Jathar et al. 2014). The possible role of IVOCs have been discussed in the European context by Bergström et al. (2012), Jiang et al. (2019) and Simpson et al. (2019).

As an example, Figure 1.1 illustrates the differences between officially reported emissions of $PM_{2.5}$ from small-scale combustion for 2010, and expert estimates made by TNO which include CPOA in a consistent way across all countries. As can be seen, for some countries (e.g. Norway:NO, Italy:IT) the two estimates are comparable, but for others (e.g. Finland:FI, Sweden:SE) the expert estimate is far higher than the reported emissions. Although CPOA are not the only difference between the emission estimates for each country (Sect. 4.3), such inconsistencies pose grave problems for the modelling of $PM_{2.5}$ and for any analysis of emission control strategies or cost-benefit analysis. In the worst case these problems might lead to wrong priorities of measures.

The issues are, however, complex, with emission factors for CPOA being dependent on a large number of factors, including measurement methods, fuels, usage, and even ambient

conditions (Robinson et al. 2010). These complexities are further discussed in Sect. 2.

With regard to the inventories, it should be noted that the issues are not just “are condensables included or not?”, but “how are they included?”. As an example, in previous years Norway had much higher emission factors (EFs) than Sweden since Norway included and Sweden excluded CPOA. Since 2019, Sweden has included CPOA, but Norway still has higher EFs since its methodology allows for poorer combustion conditions, designed to better reflect real-world usage of residential combustion appliances.

These difficulties have obvious policy implications. The process of emission control strategy development within both UN-ECE and the EU relies heavily on so-called source-receptor (S-R) calculations (Amann et al. 2011, Simpson 2013). These S-R calculations give the change in pollutant concentrations or depositions resulting from changes in precursor emissions. Typically the S-R relationships are first calculated with the EMEP MSC-W chemical transport model, by reducing emissions from each precursor and country in turn by a certain percentage, and recording the resulting changes in air quality (e.g. $\Delta\text{PM}_{2.5}$) in each grid. Statistical fits to these EMEP S-R matrices are then implemented in the GAINS integrated assessment model (Amann et al. 2011), which also incorporates information on the costs of emission control and environmental targets. This system seeks to find the most cost-effective way of meeting environmental targets, and in doing so allocates different emission control requirements to each country and precursor emission.

1.2. Impact on source-receptor calculations: a simple example

To illustrate some of the important problems with the CPOA issue, consider two countries A and B, which have identical real-world emissions, which for simplicity we assume to be only from RWC, and with all primary particulate matter (PPM) consisting of organic compounds. Country A does not include CPOA, and reports emissions of X tonnes PPM to EMEP. Country B does include CPOA, and despite having identical real-world emissions to country A, this inclusion results in a reported emission of perhaps $3 \times X$. Now, many problems result from this discrepancy:

1. First, modelled concentrations of PPM will be estimated to be 3 times higher for country B than for country A, when they should be identical.
2. It will thus appear to be far more cost effective to reduce emissions from country B than country A, which would result in extra costs for country B compared to country A, and indeed in an overall increase in costs as country B would be asked to employ more dramatic and presumably expensive emission control measures than would be the case if both countries could tackle the emissions control with equal weighting.
3. Such inconsistencies will also give a false view of model performance, and make it more difficult to know if the model is performing well or not for $\text{PM}_{2.5}$ components.

For the example above we have assumed that PPM emissions are inert, which is of course a great simplification. However, any system which allows two countries with identical emissions to report them differently is inherently unfair and will lead to incorrect source-receptor relations. Good solutions for our simple example would be if either:

- a) both countries use the same emissions methodology, including CPOA in an agreed way, and hence report the same emissions, or
- b) both countries make their assumptions very explicit, such that expert groups might attempt to estimate how to account for these differences.

Of course, option (a) would be the ideal system, and widespread use of a common EF methodology (through the EMEP/EEA air pollution emission inventory Guidebook or COP-ERT for example, see Sects. 3.2.1, 4.2), would make application of this option to many countries feasible. However, many European countries are using country-specific EFs which are based on different measurement standards so option (b) might be the most pragmatic way forward. In both cases, increased transparency is essential if the goal is to generate fair and cost-efficient source-receptor matrices and emission control strategies.

2. Condensables and the volatility basis set

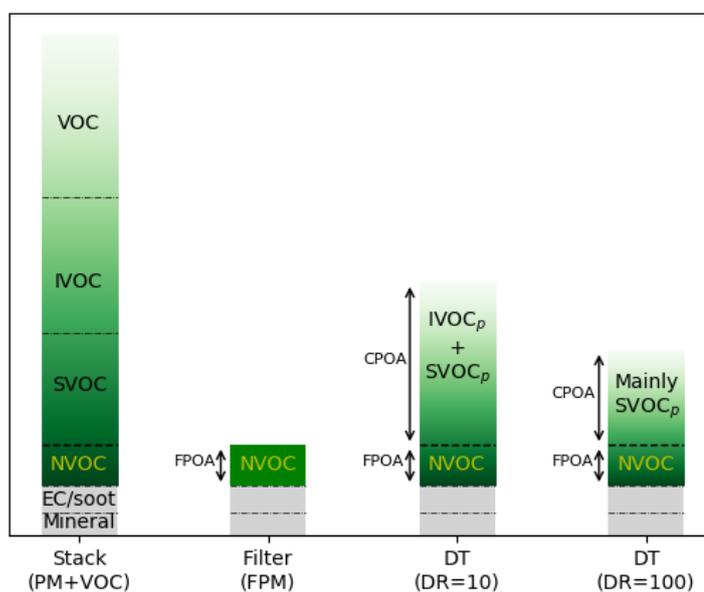


Figure 2.1: Illustration of PM emissions and FPOA, CPOA components, as measured with different sampling methods. The 'Stack' bar shows both particle and gas-phase concentrations of various components in the stack (based upon a sketch from Nussbaumer 2010), with non-, semi-, and intermediate volatility VOC marked as NVOC, SVOC and IVOC. The Filter bar represents the non-volatile components typically obtained using filter-based sampling. The two 'DT' bars illustrate PM emissions obtained using dilution tunnel sampling with dilution ratios (DR) of 10 or 100. The components sampled as FPOA and CPOA are indicated in each case. At DR=10, the CPOA consists of particulate phase SVOC and IVOC, but at DR=100 the particulate SVOC compounds are the main contributor. Note that this sketch is highly idealised, and ignores condensable inorganics, filter-adsorption of organics, that compounds labelled here as NVOC may indeed show volatility at sufficiently high temperatures, other impacts of temperature, RH, and many other factors.

2.1. Characteristics of condensable emission factors

The underlying problem associated with condensables is that a given compound might be gas or particle, and the partitioning between these two phases varies markedly between the point of fuel combustion and the ambient atmosphere. Emission factors measured in or close to the high-temperature high-concentration exhaust stack or pipe may misrepresent, and even miss, the amount of PM or gas that actually enters the atmosphere, depending on the filters, dilution and sampling conditions of the emission measurement. Filter-only measurements tend to provide just the solid fraction of PPM (plus adsorptive artefacts), which is often denoted SP or TSP, or, as in this report, FPM. FPOA is then the organic component of FPM. Dilution tunnels are normally used to capture the condensable components, which are denoted CPM for all compounds, or CPOA for the organic fraction. The amount of CPM or CPOA measured depends on many factors, including the dilution ratio (DR), sampling temperature, concentrations, residence times, and humidity. Figure 2.1 illustrates how organic gases of different volatility can be sampled as FPOA and CPOA under different dilution ratios.

Temperature is one of the key parameters, in that the higher the temperature the higher the vapour pressure (and hence C_i^* , see Box 1), and the more the partitioning of the condensable component is driven to the gas phase. Concentration is also a key parameter; the higher the mass concentration of existing organic aerosol (C_{OA} , see Box 1, Eqn. 2.1), the more any particular low volatility organic compounds will condense to the particle phase.

Box 1: Simple gas-particle theory

The impacts of temperature and concentration on EFs for semi-volatile compounds can be explained by simple gas-particle absorption-partitioning theory (Pankow 1994, Donahue et al. 2006), which suggests that:

$$\frac{C_{i,p}}{C_{i,g}} = \frac{C_{OA}}{C_i^*} \quad (2.1)$$

where $C_{i,p}$, $C_{i,g}$ are the condensed particulate and gas phase concentrations of CPOA component i , C_{OA} is the total concentration of absorbing organic aerosol, and C_i^* is the so-called effective saturation concentration (related to vapour pressure) of species i (all in $\mu\text{g m}^{-3}$). As summarised in Donahue et al. (2009), some of the salient features of Eq. 2.1 are:

- i) When C_i^* is equal to C_{OA} , 50% of compound i is in the gas-phase, and 50% in the particle phase.
- ii) For $C_i^* < 0.1 C_{OA}$ nearly all (> 90%) of compound i is in the particle phase, and for $C_i^* > 10 C_{OA}$ nearly all of compound i is in the gas phase.
- iii) Thus, changing C_{OA} will change the partitioning of each compound i .

The VBS framework lumps organics into logarithmically spaced bins (base-10) of saturation concentrations (C^*) at 298K, as seen in Fig. 2.4. For example, a bin with $C^* = 10^2 \mu\text{g m}^{-3}$ would have $\log C^* = 2$, and include all compounds whose individual C_i^* values lay between $\log C_i^*$ of 1.5–2.5. This range corresponds to C_i^* of 32–320 $\mu\text{g m}^{-3}$ (approx., see also Box 2).

Considering a range of compounds, binned into n bins of similar volatility, this equations can be re-arranged to get the particle mass fraction:

$$X_p = \sum_{i=1}^{i=n} f_i \left(\frac{C_{OA}}{C_{OA} + C_i^*} \right) \quad (2.2)$$

where f_i is the mass fraction found in the C_i^* volatility bin (Grieshop et al. 2009b).

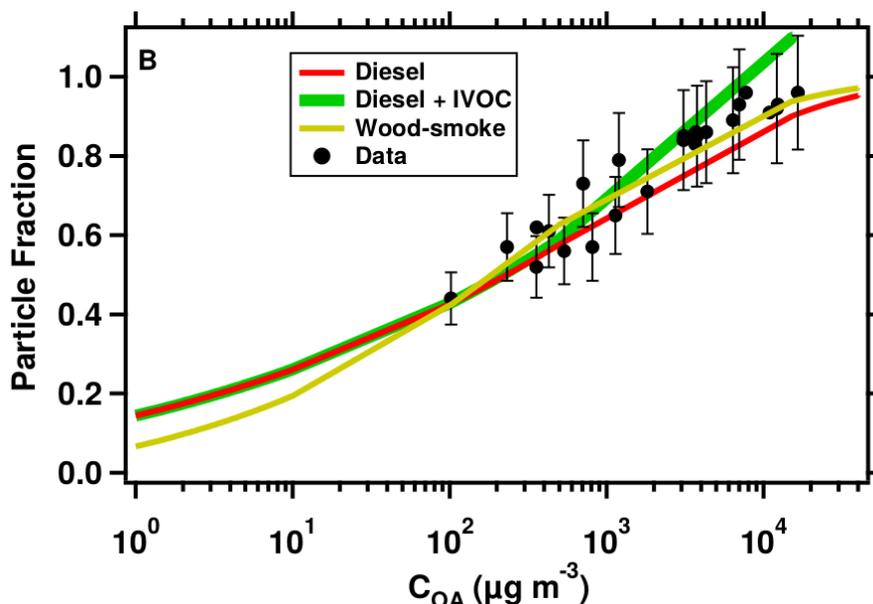


Figure 2.2: Measured gas-particle partitioning data (bullets with uncertainty bars) for wood-smoke expressed as the fraction of the semivolatile organics existing in the particle phase as a function of condensed-phase organic aerosol concentration (C_{OA}), showing how condensable compounds are found almost entirely in the particle phase at high C_{OA} levels, but mainly in the gas-phase as C_{OA} approaches ambient levels (ca. $1\text{--}10\ \mu\text{g m}^{-3}$). The curves show predicted partitioning based on volatility distributions for wood-smoke POA, diesel POA assuming no IVOC, and diesel POA with IVOC. Figure from Grieshop et al. (2009a).

Figure 2.2 shows how the particle fraction (X_p is the mass of particulate organic matter to the total mass of gas and particle-phase organics) is reduced from almost 1.0 for measurements made with C_{OA} of $10\ 000\ \mu\text{g m}^{-3}$ to under 0.2 for C_{OA} around $1\text{--}10\ \mu\text{g m}^{-3}$. This behaviour is important because PM emission factors made with dilution chambers are indeed made at widely varying and possibly very high C_{OA} levels, even over $10\ 000\ \mu\text{g m}^{-3}$ (e.g. Grieshop et al. 2009b). Applied without correction, this factor by itself will significantly overestimate the emission factor for POA and hence PM.

However, as noted above, although dilution processes encourage evaporation of organics, cooling processes encourage condensation. Figure 2.3, which extends an example presented by Robinson et al. (2010), illustrates how these opposing processes change the effective emissions factors (EF) in the plume of a vehicle exhaust. The Figure shows the change in temperature, and OC (particulate) emission factors as a function of the dilution ratio, ambient temperature (T_a) and ambient C_{OA} .

To explain the base-case ($T_a=30^\circ\text{C}$, ambient $C_{OA}=10\ \mu\text{g m}^{-3}$) in more detail, the very high C_{tot} (ca. $100\ \text{mg m}^{-3}$, not shown) at the start of the simulation and the rapid cooling (from 400K) at the exhaust pipe outlet encourages condensation of CPOA in the first seconds. Further cooling happens very quickly, but so does dilution, which causes a maximum EF at a dilution ratio (DR) of about 5, before the EF starts to decline. At point 2 in this sketch (DR=30) the plume has reached ambient temperature, and further changes in EF are caused

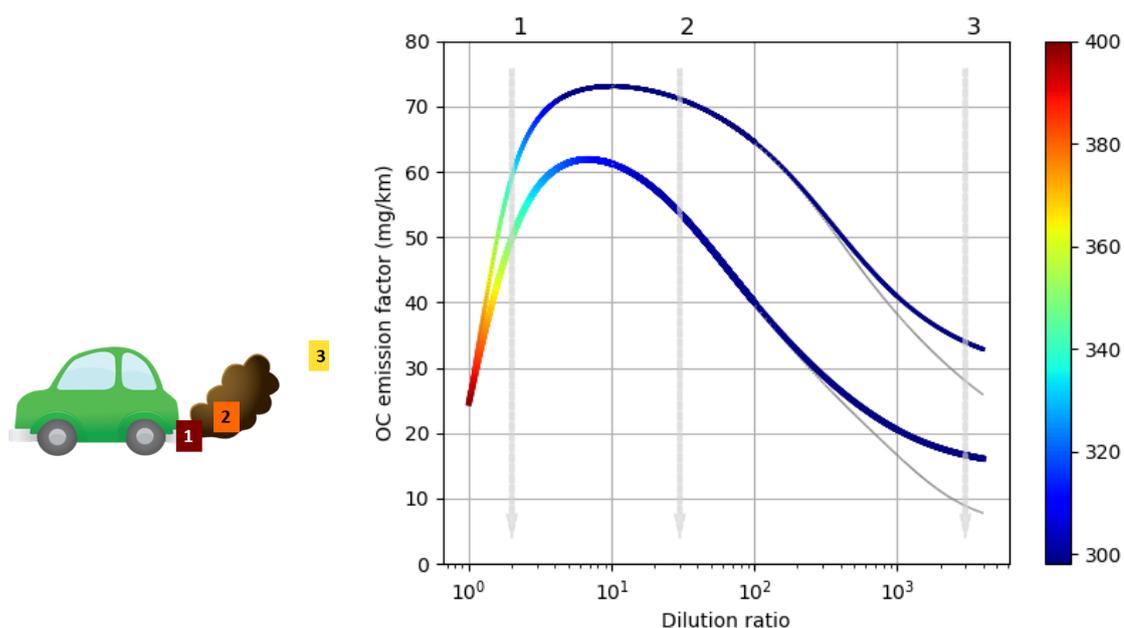


Figure 2.3: Predicted changes in OC emission factors (EFs) as a function of dilution ratio, derived from a diesel-engine setup similar to that used by Robinson et al. (2010). The coloured lines illustrate the development of temperature (colour, in degrees K) for the case where ambient C_{OA} is $10 \mu\text{g m}^{-3}$, with the thinner (grey) curve showing EFs when ambient C_{OA} is $1.0 \mu\text{g m}^{-3}$. The upper pair of curves are for ambient temperatures (T_a) of 0°C . The lower pair of curves is for T_a of 30°C . Note that these EFs are for an older US vehicle (from Schauer et al. 1999a), so only provided in order to illustrate the impacts of cooling and dilution on the EFs. The cartoon car is provided to help visualise the small spatial scales associated with the dilution ratios of 2, 30 and 3000, also indicated with labels and arrows 1, 2 and 3. For more details of the base-case setup ($C_{OA} = 10 \mu\text{g m}^{-3}$, $T_a = 30^\circ\text{C}$) see Robinson et al. (2010).

entirely by dilution (and in the real world increasingly by chemical effects, c.f. cover Fig.). Fig. 2.3, also illustrates that both T_a and ambient C_{OA} play a key role in deciding the effective EFs of this simple example, with colder temperature ($T_a = 0^\circ\text{C}$) leading to significantly higher EFs than the base-base ($T_a = 30^\circ\text{C}$), and lower C_{OA} results in lower EFs (due to increased evaporation of the CPOA).

This example was intended mainly as an illustration of the processes (although based upon real measurements), but the same effects are expected for other POA sources. For example, Nussbaumer et al. (2008b) provide a similar illustrative curve for RWC emissions, suggesting that peak emissions occur at a DR of about 20. Morino et al. (2018) also based their estimate of condensables in Japan upon an assumption that condensables could be approximated by emissions measured in a dilution tunnel for DR=20.

2.2. The volatility basis set (VBS)

These competing effects of dilution and temperature change can be explained to a reasonable degree by simple gas-particle absorption-partitioning theory (Pankow 1994, Donahue et al. 2006, also Box 1), and a good overview of the practical processes and implications can be found in Robinson et al. (2010). In this theory, the saturation vapour pressure, C_i^* , of each compound is of critical importance, since it determines the extent of partitioning between gas and particle phase for that compound. However, as noted in Robinson et al. (2007), less than 10% of the condensed and semi-volatile mass has been speciated, so complete classification of VOC or PM emissions by compound is impossible. Further, the vapour pressures of most compounds are extremely uncertain (Clegg et al. 2008, Bilde et al. 2015), so C_i^* values cannot be reliably assigned even for many known compounds. In order to overcome such limitations, the so-called volatility basis set (VBS) was introduced by Donahue et al. (2006) and Robinson et al. (2007), which effectively bins VOC compounds according to the logarithm (base-10) of their C_i^* values. Laboratory data (e.g. dilution and thermal desorption experiments) are used to estimate the amount of OM associated with each C_i^* bin rather than knowing which particular compounds are involved. The VBS system is typically also used to classify compounds as low-, semi- or intermediate-volatility VOCs (LVOC, SVOC, IVOC). Further classifications are often used (e.g. ELVOC or NVOC, Donahue et al. 2009, Murphy et al. 2014), to define compounds which are of even lower volatility than LVOC, but on the other hand SVOC is frequently used as a shorthand for all compounds which can partition to the particle phase (e.g. Donahue et al. 2006). Table 3.1 (in Box 2) illustrates a widely-used set of definitions associated with VBS volatility bins.

Box 2: What are SVOC, IVOC, CPOA?

The terms semi- and intermediate- volatility VOC (SVOC, IVOC) are widely used by experimentalists and chemical transport modellers dealing with atmospheric OA, but definitions vary, and sometimes widely. Donahue et al. (2012) noted that the definition of "semi-volatile" used in VBS systems includes much lower vapor pressures than those commonly viewed as semi-volatile, and the difference between common perception and this aerosol-specific definition of semi-volatile can be a source of confusion.

Robinson et al. (2007) defined compounds to be SVOC with C_i^* in the range $0.1 - 10^3 \mu\text{g m}^{-3}$ and IVOC from 10^3 to 10^6 . In terms of the VBS system this means that the bin centred on 10^3 contained both SVOC and IVOC. Later papers defined the bin-centres to lie at logarithmic intervals, e.g.:

	SVOC	IVOC
Murphy et al. (2014), Lu et al. (2018)	0, 1, 2	3, 4, 5, 6
Shrivastava et al. (2008)	0, 1, 2, 3	4, 5, 6

Table 2.1: $\log C^*$ values associated with SVOC and IVOC by different studies

Details of the boundaries between bins vary a little, e.g. the $\log C^* = 2$ bin has a lower boundary of $0.3 \mu\text{g m}^{-3}$ in Robinson et al. (2010), but 0.32 in Murphy et al. (2014), but these are simple approximations to the mid-point. In this case the mid-point would lie at $10^{1.5}$, which is 0.316 . The major difference though is whether the $\log C^* = 3$ bin is assigned to SVOC or IVOC.

Reference is sometimes made to carbon numbers (C#) with regard to volatility, though often what is meant is C# number 'equivalent' (see discussion in Lu et al. 2018). Robinson et al. (2007) regarded IVOC to be compounds less volatile than C12 n-alkanes or C9 carboxylic acids. Lu et al. (2018) (Fig. 2.4) suggested SVOC and lower volatility compounds are associated with C23 and more, that IVOC covers C12–C22, but this was for vehicle emissions. RWC emissions will probably have much more polar components that have lower C_i^* for a given C# (Hatch et al. 2015, Jen et al. 2019).

In the high C_{OA} conditions of stacks or engine exhausts even some of the IVOC compounds can partially partition to the particle-phase, so we cannot define condensables as simply SVOC or SVOC + IVOC. Hence term CPOA in this report, to capture essentially the sum of all SVOC and IVOC condensable compounds that are present in the particle under the appropriate experimental conditions: $CPOA = SVOC_p + IVOC_p$

Fig. 2.4 provides practical examples of the differing volatility classes for ‘typical US’ gasoline and diesel cars (from Lu et al. 2018). This figure contains a lot of useful information: C_i^* values are given on the lower x-axis; equivalent n-alkane carbon numbers on the upper x-axis; an estimate of the partitioning fraction X_p at $10 \mu\text{g m}^{-3}$ and 25°C indicated with the dashed red line; and amounts of organic compounds collected for each VBS bin with different measurement techniques. It can be noted that Lu et al. (2018) compiled data from many previous studies in an attempt to capture the full range of emitted organic compounds. As was made clear in e.g. Robinson et al. (2007), many of these compounds are not captured in the existing emission inventories. Simplifying a little, PM inventories tend to include those compounds caught on the quartz filters indicated in Fig. 2.4 (at least, if heated filters are used), and NMVOC inventories capture mainly the C2–C11 compounds indicated by the blue shaded area. Such NMVOC emissions were traditionally in focus for ozone formation, and the less plentiful I/SVOCs were generally ignored, but these latter compounds are of course now understood to be important precursors for SOA and hence PM.

A European example showing the limited carbon-number range of typical emission inventories is given in Fig. 2.5, which shows an estimate of NMVOC from recent (year 2012) UK inventories. The inventory covers only C2–C12 compounds, thus this inventory also omits the SVOC and IVOC compounds, and hence CPOA.

Although the VBS system has both challenges and limitations (e.g. concerning assumptions on enthalpies of vaporisation and other properties as discussed above, whether equilibrium between gas and particle phases can be assumed, or indeed the loss of chemical structure information), it does help to clearly expose and account for the full range of organic compound volatilities, and to highlight the existence of SVOC and IVOC emissions which are often not reported as either PM or VOC. These unaccounted-for emissions can be substantial sources of SOA in the atmosphere (e.g. Robinson et al. 2007, Bergström et al. 2012, Denier van der Gon et al. 2015, Ots et al. 2016b, Murphy et al. 2017, Jiang et al. 2019). Although derived from laboratory data, the VBS framework has also been shown to successfully predict partitioning in the ambient atmosphere. For example, Saha et al. (2018) showed that the VBS system for gasoline vehicle emissions (from May et al. 2013) could explain changes in measured ambient OA over distances of 10m–220m from from a freeway in North Carolina in both summer (26°C) and winter (6°C) conditions.

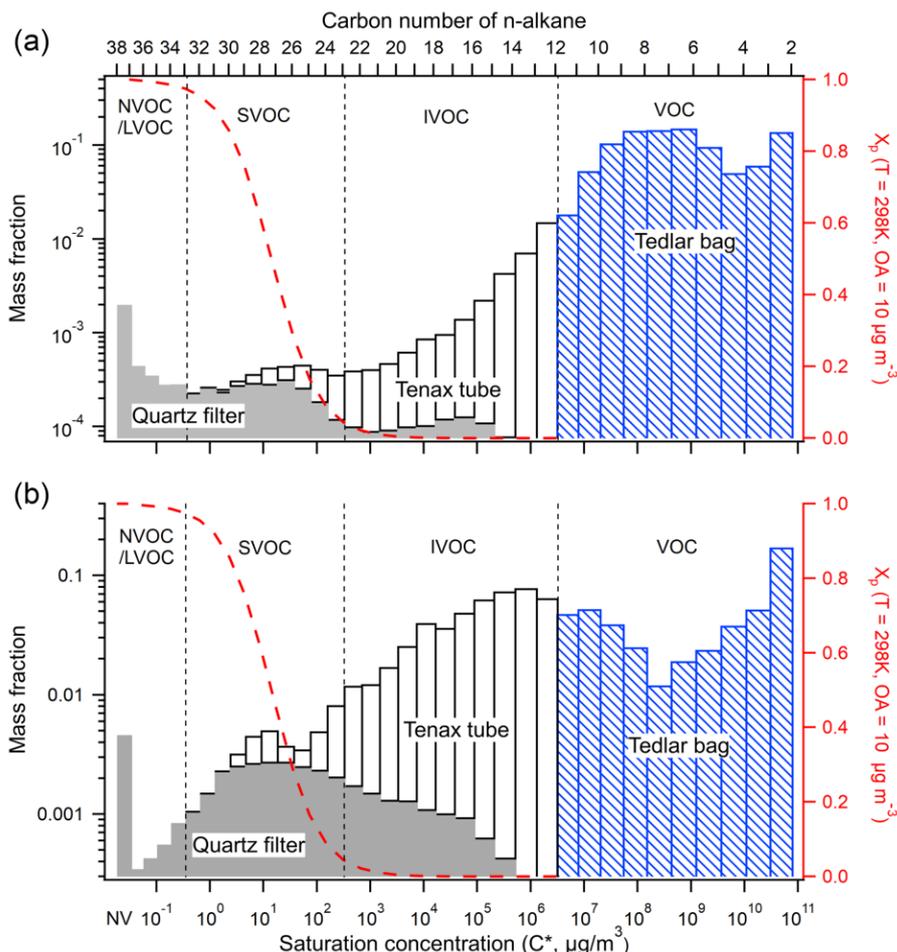


Figure 2.4: Volatility distribution of organic emissions for a typical (a) gasoline (b) diesel vehicle (both from USA fleets). The emissions are classified by sampling media (line 1: Tedlar bag, line 2: bare quartz filter followed by two Tenax tubes). The red dashed line indicates the partitioning fraction (X_p) assuming the emissions for a quasi-ideal solution at C_{OA} of $10 \mu\text{g}/\text{m}^3$ and 298K. From Lu et al. 2018.

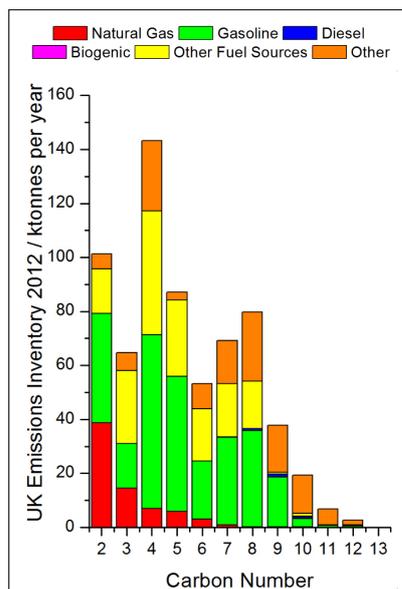


Figure 2.5: NMHC emissions by carbon number and functionality from UK 2012 emission inventories. The carbon number and functionality of emissions have been estimated by applying the speciated inventory of emission sources of Passant (2002) to estimates of 2012 NMVOC sources. From Dunmore et al. (2015).

The VBS scheme seems to offer a unifying framework for translating EFs measured in one set of conditions to other conditions. For example, Grieshop et al. (2009b) compared the gas-particle partitioning and VBS characteristics of a diesel engine, combustion of hard- and soft-woods in a stove, and also aerosols formed from flash-vaporised engine lubricating oil. Although differences were found (e.g. with wood smoke being somewhat less volatile than the diesel exhaust), the overall partitioning characteristics of diesel and wood smoke POA were similar (see also Fig. 2.2). Thus, current studies seem to suggest that different sources have different VBS profiles, but not too different. As emission factors vary so widely due to other conditions, e.g. for type of wood, moisture status, user-habits for RWC (Seljeskog et al. 2017b, Nussbaumer et al. 2008a), the biggest source of uncertainty is probably in the emission factor determinations themselves, rather than which VBS profile is applied. Still, proper accounting for systematic differences in EF measurements due to gas-particle partitioning can reduce discrepancies by factors of 2–10 (e.g. Figs. 2.2-2.3).

2.3. Can we define condensables?

Summarising the current knowledge as discussed in e.g. Robinson et al. (2010) or Nussbaumer (2010), it is clear that emission factors (EF) for condensables depend on:

1. source (stoves, diesel cars, etc.)
2. operating conditions
3. measurement conditions (stack C_{OA} , temperatures, dilution ratios, filters, ...)
4. ambient temperature
5. ambient C_{OA} .

Thus, measured EFs depend strongly on the protocol used to make the measurements, which implies that knowledge of these protocols and conditions is also required if we are to compare EFs across different studies, to make harmonised emission inventories, and to make sensible use of such EFs in atmospheric modelling studies.

As discussed in sect. 2.1, the EF of condensables measured in an exhaust plume tends to peak at dilution ratios of around 5–20. Indeed, the estimate of condensables in Japanese emissions made by Morino et al. (2018) made the pragmatic choice of defining the EFs as those occurring for DR=20.

If some kind of standard conditions could be defined (perhaps some ambient reference, $T_a = 295\text{K}$, $C_{OA} = 10 \mu\text{g m}^{-3}$, or based upon DT criteria, e.g. DR=10), it should be possible to define a procedure, and possibly a web-interface, where the analyst provides the calculated OM EFs, the measured C_{OA} and the temperature, and the total condensable EFs could be calculated based on an agreed volatility distribution. Using this same distribution in models would ensure consistency and comparability between results. There are unfortunately issues with this idea though. As stated by Robinson et al. (2010), one could conceivably address the phase partitioning issue by specifying the temperature and concentration at which dilution sampler measurements are made. At a minimum, these parameters need to be reported along

with emission factor data. However, specifying a concentration would further complicate the test procedure, not span the atmospherically relevant range of conditions, and not solve the problems of filter artefacts.

Robinson et al. (2010) went on to propose that in order to address emissions of POA and the contribution of low-volatility vapour emissions to SOA, one could simply define an emission standard for all low-volatility organics, regardless of phase. The source test would be similar to existing total hydrocarbon measurements except that a sorbent would be used to isolate the less-volatile fraction ($C_i^* < 10^6 \mu\text{g m}^{-3}$, essentially SVOC+IVOC) of the hydrocarbons. By specifically targeting the less-volatile fraction, this proposed test could address the components of the emissions that contribute to atmospheric organic PM mass. The measurements could be made without dilution, which would greatly simplify the existing test procedures. It would also better focus testing on the components of the emissions that contribute most substantially to atmospheric PM rather than on those somewhat arbitrarily collected based on their gas-particle partitioning inside of a dilution sampler.

Of course, there is a large infrastructure already in place for measuring EFs from different sources and in different countries, and any changes or ‘correction-factors’ (to convert EFs made by one method to any other) would involve a lot of discussion and work.

There is unfortunately no easy answer to these basic issues, but increased knowledge of the measurement techniques, capabilities, and assumptions behind the numbers used for national EFs are needed. With increasing transparency, and more use of common assumptions (through for example the EI guidebook), the situation should improve. A systematic harmonisation effort within the Air Convention framework would be very beneficial in this respect, in order to (a) understand the EFs used by different countries, (b) convert them to a common standard, (c) to recommend more unified and consistent methodologies in the future, and (d) more accurately apply detailed chemical speciation data across sources and conditions. These issues are discussed further in sect. 5.

3. Sources

3.1. Residential Biomass Combustion

It is clear that emissions from residential wood combustion (RWC) account for a major fraction of PM emissions in Europe, and differences in countries handling of condensables in their reported RWC emissions are probably the greatest source of problems in comparing national inventories (Denier van der Gon et al. 2015). The basic EFs are also very hard to specify for this category because of the large variations in burning conditions and emission-measurement protocols, especially in ‘real-world’ conditions (e.g. Nussbaumer et al. 2008a, Seljeskog et al. 2017b, Kindbom et al. 2018, Reichert and Schmidl 2018, see also Fig. 3.1). Emissions depend heavily on many factors, including technical (e.g. type of stove), fuel (type of wood, moisture content), and usage. Emissions change significantly over a burning cycle in

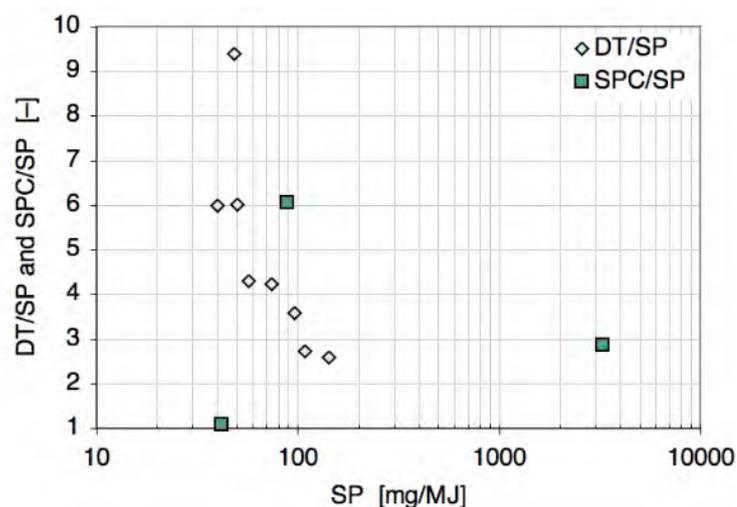


Figure 3.1: Ratios of dilution tunnel (DT), solid particle and impinger (SPC) and solid particle (SP) emission rates for wood stoves, illustrating high and variable contributions of condensables (DT, SPC methods) compared to solid particles. From Nussbaumer et al. (2008a).

Table 3.1: Wood use by appliance type in Europe in 2005 and related solid particle (SP) and dilution tunnel (DT) particle emission factors. Table from Denier van der Gon et al. (2015)

Appliance type ^a	Wood use in Europe in 2005 (PJ)	Fraction of wood consumption	Emission factor (g GJ ⁻¹) ^b			
			SP		DT	
			Avg	Range	Avg	Range
Fire place	140	6 %	260	23–450	900	^d
Traditional heating stove	1167	52 %	150	49–650	800	290–1932
Single house boiler automatic	198	9 %	30	11–60	60	^d
Single house boiler manual	348	15 %	180	6–650	1000	100–2000
Medium boiler automatic	267	12 %	40	^c	45	^c
Medium boiler manual	141	6 %	70	30–350	80	30–350
Total Europe	2262	100 %				

^(a) Following IIASA GAINS stove type definition (Klimont et al. 2002a).

^(b) Derived from Nussbaumer et al. 2008a,b.

^(c) Range in emission factor is determined by end-of-pipe emission control.

^(d) Not enough data available to indicate range.

even well-maintained and well-run appliances, and they increase dramatically if sub-optimal combustion conditions occur. Concerning condensables, measurements under both lab- and real-world conditions show high EF of total PM (solid+condensables) compared to EFs for solids alone, as seen in Fig.3.1 and Table 3.1. As noted in Sect. 1.1, the question is much more complex than just “are condensables included”, but also “how are they included?”. Even where included, countries use very different methodologies to estimate their EFs. For example, France, Germany, Sweden and Norway use different EF methods. Various reports have compared emission factors (e.g. Reichert and Schmidl 2018), but still each country uses its own data and its own methods (see Sect. 3.1.1). We give a few examples below.

3.1.1 National Approaches

Some examples of national approaches give a flavour for the variation in emission factor characteristics:

Germany:

Emission factors for total suspended particulates (TSP) and PM are mainly based on measurements without condensed compounds, according to CEN-TS 15883 (CEN 2009), annex I. This measurement standard is also the basis for the national limit values. All measurements, also historical values are based on this national standard. In addition emission factors for small combustion systems are determined in accordance with device design, age level, output category and typical mode of operation. The age level is important since the German legislation requires the decommissioning of old stoves which were produced before 2010 until 2024. Test bench values cannot be used directly, since they do not reflect reality. Nevertheless it is important that emission factors used for the inventory are comparable with the limit and the test bench values and historical data in order to be able presenting the technical development and the emission trend respectively. Poor combustion conditions were not considered in the inventory, since emission level and trend are difficult to estimate. A reliable data source is not

available. Due to the amendment of the 1st Federal Immission Control Act, which applies for small combustion plants, a trend in this respect can be expected. Therefore the inclusion of poor combustion conditions would have an effect on compliance issues.

Norway: In Norway condensables have been included for many years (Hansen 1998, Seljeskog et al. 2013, 2017a, Kausch et al. in preparation), using EFs defined by a set of Norwegian Standards (NS). Emission factors using NS procedures can be much higher than EFs using procedures from other European countries. As described in Seljeskog et al. (2013) differences are primarily due to testing with non-optimal burning conditions (as described in NS 3058-1:1994), with condensables collected with a dilution tunnel (method described in NS 3058-2:1994). The final emission factor is weighted in accordance to NS 3059:1994. See Seljeskog et al. (2013) for more details of the standards and methods.

Sweden:

Prior to 2019, condensables were not included in Swedish EFs, which gave rise to the large discrepancies between Swedish and Norwegian EFs as highlighted in Sternhufvud et al. (2004) and Denier van der Gon et al. (2015). From 2019 onwards, condensables were added. The EFs were based upon measurements made on Nordic wood-burning appliances using dilution tunnel according to Norwegian standard 3058. Poor combustion conditions were simulated including measurements during part load and with moist fuel. When implemented in the Swedish emission inventory, it was assumed that 10% of the wood fuel was combusted in poor combustion conditions, based on interviews with Swedish chimney sweepers (Kindbom et al. 2018).

USA:

Regulated sources of wood-burning emissions, including residential and industrial wood heaters, are characterised by test methods 5G¹ for sampling emissions passed through a dilution tunnel and 5H² for sampling directly from a stack location. Method 5G employs a dilution tunnel to cool flue gas to 32°C prior to filtration and collection with variable dilution ratio recommended not to exceed 150:1. Method 5H uses two filters one required to be at a temperature less than 120 °C and a second required to be less than 20°C. Residential wood appliance results are reported to the U.S. EPA Office of Enforcement and Compliance Assurance (OECA) where a database of compliant products is maintained. Efforts are underway to pass these wood heater data through the Compliance and Emissions Data Reporting Interface (CEDRI).

The stationary source PM_{2.5} test methods have evolved significantly since 1990. Older test methods may have missed CPM or overestimated CPM, depending on the method. In the most heavily used databases for emission factors (AP-42 and WebFIRE) different PM measurement techniques have been used for different sectors; consequently, the emission factors that result may not include the condensable component even when they should.

¹U.S. Environmental Protection Agency (a), Method 5G – Determination of particulate matter emissions from wood heaters (dilution tunnel sampling location). 40 C.F.R. Part 60 Appendix A-3, as of November 23, 2020.

²U.S. Environmental Protection Agency (b), Method 5H – Determination of particulate matter emissions from wood heaters at a stack location. 40 C.F.R. Part 60 Appendix A-3, as of November 23, 2020.

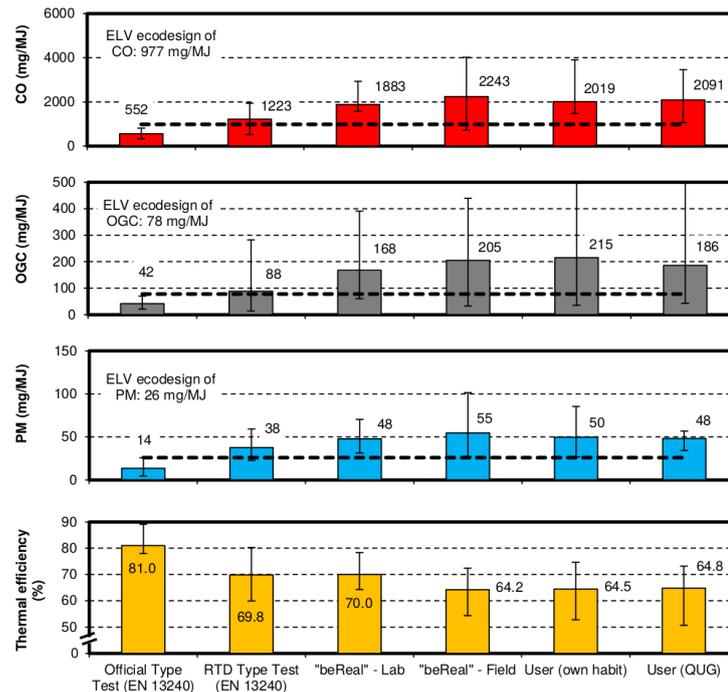


Figure 3.2: Comparison of official type test results for thermal efficiency, PM, organic gas compounds (OGC), and CO with lab and field test results. Lab and field test results were measured with the same appliances (all EN 13240). Error bars represent the minimum and maximum values determined. The dashed black lines represent the ELVs of future ecodesign requirements for local space heaters (closed fronted) and cookers. Reproduced from Reichert and Schmidl 2018, with permission.

3.1.2 Standards of firing procedures and measurement methods

The lack of standards and agreed methods is a serious problem. Current standards for RWC are more industry-needs oriented, and have not been designed with air pollution emissions or air quality issues concerning condensables in mind. Suitable, i.e., more representative standards in terms of performance and impact on air quality, should include and distinguish:

- firing procedures i.e. the operating conditions under which the appliance should run during the tests. The firing procedure should include start-up and low output phases,
- measurement methods to be used to characterise the emissions of pollutants. Measurement methods that include condensables are necessary to better evaluate combustion quality and impact ambient air concentrations of PM.

The influence of firing procedures can be observed in Fig. 3.2 which compares official type test (oTT) results with laboratory and field test results obtained for the same appliances under the 'BeReal' project. The results obtained from oTT are substantially lower than those obtained from the 'BeReal' approach (designed to reproduce real-world conditions better), or field data.

Emissions from solid fuel space heaters, such as wood stoves, are regulated in the Ecodesign Regulation (EU) 2015/1185³, and the Construction Product Regulation 305/2011 which regulates wood stoves as a construction product. There is currently no harmonised test method for PM emission measurements, and the Ecodesign Regulation allows manufacturers to select one of three different measurement methods with corresponding limit values, including the Norwegian standard NS 3058/59.

In 2020 the EN-PME validation project, which was initiated by CEN TC295 working group 5, was finished and proposed the EN-PME test method as test method to include in the European standard EN 16510 (Residential solid fuel burning appliances) to replace these three existing methods. The idea of the EN-PME method is to make a clear distinction between solid particles and VOCs, with both components being measured separately. This method was intended for the evaluation of new stoves, but the values cannot be used for an emission reporting system where the condensable fraction should be included.

In addition, there are no precise requirements in the non harmonised standard EN 16510 for the test design to reflect actual real-life conditions, i.e. testing under different fuel load and firing rates. This only requires testing under optimal conditions, and hence underestimates real-life emissions.

Another issue is the problem of efficiency of reduction techniques. For wood combustion, according to recent French studies, the most advanced wood appliances have good efficiency on filterable particles but not on condensables. We do not have good information on other sectors (except road traffic).

There has been much activity with regard to measuring condensables in Europe in recent years. Reliable methods do exist, and are well known in most European countries. Some simplified methods (e.g. HF-IPA impingers) also show very good agreement with the dilution tunnel methods, and allow the sampling of a large number of appliances under different conditions (Fraboulet 2016). More recently a simple method of dilution aiming at taking into account condensables has been developed by ENEA⁴/ISSI⁵ and evaluated using woodlog and pellet stoves within the EMPIR Impress 2 project⁶ by five European partners (ENEA/ISSI, DTI, RISE and INERIS). Increased use of these new firing procedures and methods should expand the availability and comparability of data, and lead to much more realistic and comparable EFs for European RWC emissions. It also seems clear that EFs differ more from burner to burner than between countries, thus there would be benefit in pooling the data from various European (and where comparable, North American) measurements to get better statistics.

³https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=uriserv%3AOJ.L_.2015.193.01.0001.01.ENG

⁴Italian National Agency for New technologies

⁵Innovhub Stazioni Sperimentali Per L'Industria

⁶e.g. <http://empir.npl.co.uk/impress>

3.2. Road traffic

Road vehicles are major sources of PM and VOC in all regions, and of course these emissions have received much attention, and are subject to increasingly stringent emission standards in Europe and North America (e.g. Drozd et al. 2019). Figure 2.4 has already demonstrated the wide range of compounds and volatility from both gasoline and diesel vehicles from the US car fleet, and similar ranges have been demonstrated in Europe (e.g. Xu et al. 2020). Characteristics of vehicles differ between the US and Europe though (Sect. 3.2.3), and emissions have also changed dramatically over the years. As noted in Jiang et al. (2019), diesel vehicles constitute nearly half of the total passenger car registrations in Europe (ACEA 2017). Diesel vehicle emissions were traditionally considered more efficient in generating SOA than gasoline exhaust (Gentner et al. 2012). However, vehicles equipped with a diesel particle filter (DPF) have been found to effectively reduce SOA production (Gordon et al. 2014, Kim et al. 2016, Gentner et al. 2017, Platt et al. 2017). In Europe, diesel particle filters (DPFs) were introduced in some Euro-4 vehicles in 2005, and in all Euro-5 vehicles since 2009 (Jiang et al. 2019). These DPF-equipped vehicles have very low emissions of SVOC and Jiang et al. (2019) set their SOA formation yield to be zero. For such vehicles IVOC, rather than SVOC, are by far the most important source of SOA precursors, but as noted in Jathar et al. (2014) and Jiang et al. (2019), IVOC are often associated with the NMVOC inventory for vehicles rather than the PM inventory.

It is still unclear though how far laboratory-based emission measurements reflect real-world usage, where different engine loads cause variable catalyst temperatures which can lead to limited effectiveness, as opposed to dynamometer tests where the catalyst is held at optimum operating conditions (Carslaw and Rhys-Tyler 2013, Dunmore et al. 2015). Dunmore et al. (2015) compared real-world urban composition in London with regulatory emissions inventories, and highlighted a previously unaccounted for, but very significant, under-reporting of diesel-related hydrocarbons; an underestimation of a factor 4 for C9 species rising to a factor of over 70 for C12 during winter. These observations suggested that hydrocarbons from diesel vehicles dominated gas phase reactive carbon in London, which has a high fraction of diesel vehicles, and this was further shown to have significant impacts on SOA formation over the region (Ots et al. 2016b). More recently, Xu et al. 2020 found that diesels accounted for most of the measured alkanes (acyclic and cyclic) and aromatic S/IVOC compounds at a central London roadside site during 2017, with gasoline contributing to the lower molecular weight S/IVOCs. It is not known to what extent these results from London can be transferred to other urban areas with high diesel fractions. Fig. 3.3 (from Platt et al. 2014) shows a comparison of POA EFs from a wide variety of vehicles and sources, as measured either in laboratory conditions or estimated from roadside and tunnel data. These results also seem to suggest that real-world emissions (the road/tunnel data) produce higher EFs than current European vehicles, but it should be noted that most of the road/tunnel data came from US measurements.

Although most focus has been on four-wheeled diesel and gasoline vehicles, two-wheeled vehicles also need to be considered. Platt et al. (2014) suggests that two-stroke scooters can have POA and NMVOC emission rates far higher than their fuel consumption would suggest. These vehicles often have rather poor maintenance standards and are subject to tampering, and evidence from air quality measurements before and after bans on scooters in Asian cities

Figure 3.3: POA emission factors plotted as box-and-whiskers (median line, red: 25th and 75th percentile, box: 10th and 90th percentile, whiskers) (g kg fuel^{-1}) for different vehicle types (2S = two-stroke), Grey markers are from test bench, whole coloured markers are from road tunnels in different regions. Figure from Platt et al. (2014), who also provide further details.

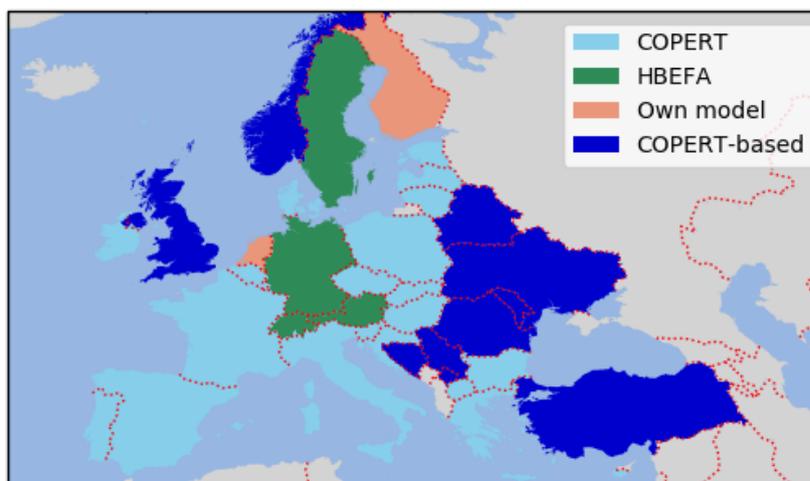
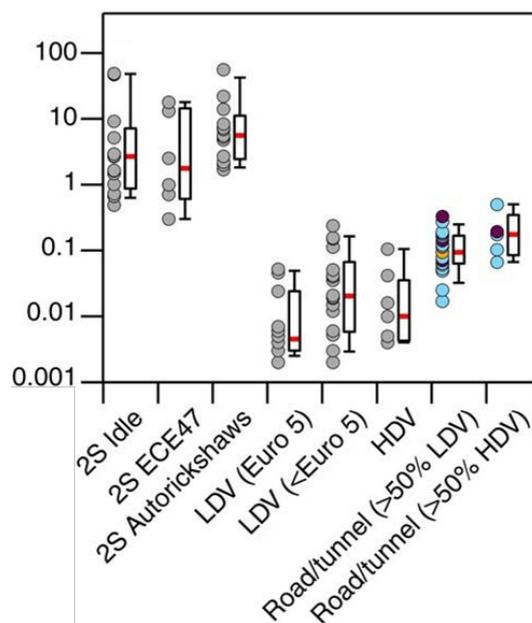


Figure 3.4: Vehicle emission models used in Europe. (Redrawn from ERMES group illustration, https://www.ermes-group.eu/web/leading_EU_models).

suggest they may dominate vehicular pollution despite their relatively small numbers (*ibid.*). Platt et al. (2014) did not present compound or VBS-classified POA emissions, but this class of vehicle warrants further attention in terms of both the POA and NMVOC emissions.

3.2.1 Road transport emission models/COPERT

The emission inventories from most European countries tend to make use of a few vehicle emissions models, as seen in Fig. 3.4. The COPERT system (<https://emis.com/c>

opert) seems to be the most widely used, but HBEFA (HBEFA 2017, Keller et al. 2017) is used in Austria, Germany, Sweden and Switzerland. COPERT and HBEFA cooperate through the European Research Group on Mobile Emission Sources (ERMES, <https://www.ermes-group.eu>). The models overseen by the ERMES group are based on a common set of measurements in order to produce consistent emission factors and emissions estimates. The ERMES emission factors are then fed in the most widely used inventory models across Europe. Finland and the Netherlands have their own national systems. COPERT is discussed in more detail below.

However, despite the harmonising nature of such vehicle emission models, estimation of POA emissions is still fraught with uncertainty. Emissions of POA from vehicles vary a lot with fuel, combustion principle and operation mode, and the SOA/POA ratio varies with vehicle type (e.g. Simonen et al. 2019). Transient effects (e.g. cold starts with gasoline, or occasional DPF regeneration with diesel, e.g. Gordon et al. 2014, Pereira et al. 2018) can also heavily influence POA emissions and the SOA-formation potential of emissions.

Emission factors for PM originate from laboratory measurements following exhaust dilution that brings the sampling temperature to 20–52°C. Dilution ratios in the order of 10:1 for passenger cars and 100:1 for heavy duty vehicles without a diesel particle filter are used. These EFs are validated with on road measurements as well, following ambient dilution. COPERT EFs are therefore considered to contain most of the condensable material. The filters are conditioned for humidity before and after the measurement. However, as the filter weight increases after the measurement, inevitably the water content adsorbed on the filter increases as well. If condensables are determined by PM evaporation, this extra humidity can cause a positive bias to the mass of condensables determined (Giechaskiel et al. 2014).

EC/OC ratios for COPERT are determined from the analysis of quartz filters, and then OM estimated as 1.2 times OC. Final values of EC/PM_{2.5} and OM/EC are determined from averages of collected data, and cross-checked and corrected with tunnel measurements. The latest revision of these values was conducted in 2006.

It should be noted that there are significant measurement uncertainties that originate from the filter material use during the above process. There are three different filter types used for collection of PM. In the US PTFE-membranes are used, whereas in the EU PTFE-coated, or (for chemical analysis) quartz filters are used. Because of material affinity, condensables tend to increase in that order, i.e. quartz>PTFE-coated>PTFE membrane (although all can measure CPOA if the concentrations are high enough).

COPERT also provides tables to speciate NMVOC emissions, and mainly the range C1–C10 is covered. A C>13 group is included, accounting for 3.5–6% of gasoline cars, 13% of diesel passenger cars and light-duty vehicles, and 20% of heavy-duty vehicles. No data are given however on SVOCs, and the data provided are in need of an update.

3.2.2 Volatility and links to PM/NMVOC?

Referring again to Fig. 2.4, gasoline and diesel vehicles are seen to be sources of SVOC and IVOC, on top of, or as partial subsets of, the traditional PM and NMVOC categories.

Although technologies such as DPF should have reduced many of these emissions significantly in recent years, Kim et al. (2016) suggest that SVOC emissions can still be significant from European vehicles. They also point out that inventories in Europe (and COPERT) do not include the full contribution of SVOC emissions in their PM EFs. For France, Kim et al. (2016) estimated that the gas/particle ratio of SVOC emissions for passenger cars is about 1.5, which would imply that current European emission inventories may underestimate SVOC emissions from passenger cars by about 60%. However, these estimates were based upon data from three vehicles under specific experimental conditions, so need to be confirmed by other studies.

Further to OC, organic material with the propensity to form PM may also be determined in vehicle exhaust emission measurements as part of the total hydrocarbons (THC). The current measurement setup underlying COPERT is not well designed to characterise such condensables species. For diesel vehicles, the measurement is done online by heating to 190°C but for gasoline the measurement is offline after collecting the sample in a Tedlar bag at room temperature. The temperature conditions are completely different and these cannot be used to draw consistent conclusions between the two vehicle types. Moreover, THC is often used interchangeably with VOC which may not be the case for some oxygenated alternative fuels. Still, a number of studies associated SVOC emissions with the non-volatile PM components, and IVOC with the gaseous NMVOC emissions (e.g. Murphy et al. 2017). For example, Zhao et al. (2015, 2016) measured IVOC emissions from diesel and gasoline vehicles (and small off-road gasoline engines) and found them strongly correlated with NMHC emissions.

3.2.3 European vs North American situation

As noted above, diesel vehicles constitute nearly half of the total passenger car registrations in Europe (ACEA 2017), and over 50% of the total vehicle fleet. This is far more than in the US fleet in which practically no diesel passenger cars are in circulation. In addition, the latest generation of gasoline vehicles in the EU (Euro 6d-temp and Euro 6d) are equipped with gasoline particle filters (GPFs) which are not used in the US. GPFs have a high efficiency in filtering out non-volatile particles but they also reduce condensables due to their catalytic activity.

Mobile sources of $PM_{2.5}$ are characterised in the United States with dilution samplers where the dilution ratio is modulated to keep the filter temperature at $47 \pm 5^\circ C$. – see descriptions in the Code of Federal Regulations Title 40 Part 1065 (Engines) and 1066 (Vehicles). As noted in Lu et al. (2020), a challenge is that most existing POA emission factors used to inform the US NEI inventory are based on filter measurements, which do not quantitatively collect all SVOCs. Lu et al. (2020) evaluated the potential biases, and estimated that gasoline POA emissions needed to be corrected by a factor 1.4 to account for missing SVOC mass, but no correction was needed for diesel and gas-turbine emissions.

3.2.4 Issues with road transport emissions

In general, more data on POA emissions are available from U.S. studies (e.g. Lu et al. 2018, Drozd et al. 2019) than European ones, and there is a need to update the European databases to the latest data from all such sources. However, the European vehicle fleet is different in many ways to that of the U.S. – for example gasoline particle filters and diesel passenger cars with selective catalytic reduction (SCR) are not available in the US. Further measurements are needed to complement and confirm any transfer of U.S. POA and NMVOC information to European databases.

The methodologies used in Europe could become more comprehensive, but it is unclear if it is better to measure by volatility (e.g. SVOC, IVOC) or by chemical species.

In general, the issue of condensable SVOCs from vehicles is probably less important than the issues associated with IVOCs (Jiang et al. 2019), whether condensable or not. These IVOCs are emitted in greater amounts, but as noted above they are likely in most cases better correlated with the (more volatile) NMVOC emissions than with the PM emissions, and thus are not the main focus of this project.

Despite the complexity of road transport emissions though, understanding of condensables from road-transport is much further along than for other sectors because 1) there is a robust regulatory and testing framework built for vehicle emissions and 2) the fuel composition is tightly constrained and thus emissions complexity is relatively easier to characterise. The activity statistics are also much better known than for many other sources.

3.3. Others sources

There seems to be only limited studies on condensables from sources other than RWC or road transport, especially in Europe. Here we briefly mention some of the sources which need to be looked at in more detail in terms of condensable emissions. The most common terminology in much of this literature seems to consist of FPM for filterable particulate matter and CPM for condensable particulate matter (CPM), but as noted in Sect. 1 these components include inorganic matter.

One of the more extensive efforts to estimate CPM from other sources has been made in Japan, where the default emissions do not include condensables. Morino et al. (2018) investigated the impact of condensables on the Japanese inventory, based upon emission surveys made in the Tokyo metropolis. They compared emission factors from FPM and filterable+condensable (FCPM, with the latter determined for dilution ratios of 20), and also

estimated the gas-phase SVOC components in the primary emission. With this methodology, overall emission rates of OA increased by a factor of 7 over Japan, with stationary combustion sources in the industrial and energy sectors estimated to be the largest sources of OA.

Reviews of sources such as industrial stacks and coal-fired plants can be found in e.g. Feng et al. (2018), Corio and Sherwell (2000) or Gong et al. (2016), but in all these studies inorganic emissions made large contributions to CPM. Indeed, it should be noted that many of the papers on the subject of industrial and power-generation emissions stem from Asia or from older North American studies, but it is then important to note that the fuels may include higher-sulphur content than those used in European situations, which will lead to higher inorganic contributions to CPM.

Off-road diesel engines, largely comprised of industrial, agricultural, marine, and stationary sources, can also be a large source of POA and NMVOC (Jathar et al. 2014, McDonald et al. 2015), and certainly deserve attention in the context of CPOA. As noted in McDonald et al. (2015), the less efficient off-road diesel engines may be the primary source of mobile POA in Los Angeles.

Unregulated fire source emissions (e.g. wildfires, prescribed burns, agricultural fires) can be major sources of SVOC and IVOC, at least locally and during specific periods. In the USA measurements on these unregulated fire emissions are performed in both the lab and field. Lab measurements are often based on burning small batches of fuels at about 20°C and experimental conditions like dilution are highly variable dependent on measurement technology (filters, calibrated nephelometers, and aerosol mass spectrometers, etc.). Extensive studies of US wildfire composition, with detailed chemical analysis have been reported by for example Jen et al. (2019) and Hatch et al. (2018).

In Europe the available data on chemical composition seems to be sparser, but there are also problems in the basic reporting of fire source PM emissions. As noted in Amann et al. (2017), Member States are obliged to report emissions from the open burning of agricultural residuals in their emission inventory submissions to EMEP/EEA. However, such burning is banned in the EU, and there are indications that some inventory agencies do not include these illegal emissions. By combining satellite data with emission factors from literature (consistent with the EI Guidebook), IIASA estimated that burning of agricultural waste accounted for 3.3% of total PM_{2.5} emissions in the EU-28 during 2015, but with particularly large contributions (10–20%) in some Mediterranean and Eastern European countries.

Emissions from food-related sources (meat cooking/frying/etc.) may be significant sources of OA and also of SVOC (Schauer et al. 1999b, Robinson et al. 2006, Huffman et al. 2009, Sun et al. 2011, Mohr et al. 2012, Crippa et al. 2013, Ots et al. 2016a), although there are still challenges in source-apportionment studies related to this emission source (Reyes-Villegas et al. 2018). It is not clear how cooking-related emissions are included in inventories, and to what extent condensables might be accounted for.

Some studies of the IVOC (and SVOC) emissions from shipping and ship engines have been published in recent years (e.g. Huang et al. 2018, Lou et al. 2019, Su et al. 2020), investigating among other things how emissions depend on the fuel type and operating conditions. Huang et al. (2018) found larger IVOC emissions from low-sulphur fuel than high-sulphur fuel (HSF) and that the total IVOC emissions (from HSF) were better correlated with POA emissions than with "total hydrocarbon" emissions, though only a small number of samples were investigated. More studies are clearly needed for this source.

The Canadian Oil Sands operations are a major non-combustion source of OA in Northern Canada (Liggio et al. 2016). Reported filterable PM is biased low when compared to top-down

emissions inventories of POA (Stroud et al. 2018). However, the top-down emissions inventories of POA were measured slightly down-wind of emission sources and are a combination of condensed POA as well as SOA from IVOC sources. The relative distributions of SVOC and IVOC from the Oil Sands are uncertain, though IVOCs are likely the more important precursors for OA from the Oil Sands (Liggio et al. 2016, Li et al. 2019).

Refineries may also contribute to emissions of condensables and IVOC in Europe, but more data are needed. As an example, Concauwe (see [/www.concauwe.eu](http://www.concauwe.eu)) is planning to undertake a stack testing monitoring campaign for the measurement of these emissions in a gas-fired stack. The monitoring campaign will consist of 3 pairs of tests which will be carried out through the parallel operation of two sampling systems deployed on the stack. Each sampling system will be operated under a different technique and according to one of the two following Standards: a) US EPA Method 202, and b) ISO 25597:2013.

The importance of another potentially important source of SOA precursors, volatile chemical products (VCPs) has recently been highlighted by McDonald et al. (2018). However, these are non-combustion sources, and thus outside the scope of this work on CPOA.

Finally, a recent study (Khare et al. 2020) has demonstrated that asphalt-based sources can be a major source of S/IVOC in urban areas, but noted that their emissions are essentially absent from inventories.

4. Emission inventories

This chapter briefly discusses the status of current emission inventories, some issues with these, and a discussion of possible future inventories. Emission inventories as reported to the Air Convention are primarily developed on a national scale, with each country free to use its own methodologies. The status for condensables in European reporting is presented in Sect. 4.1. Many European countries make use of methods and data from the EMEP/EEA air pollutant emission inventory guidebook (EMEP/EEA 2019, hereafter EI Guidebook, Sect. 4.2). The EI Guidebook acts as a central source of information for all Parties to the Air Convention, with the exception of Canada and the USA who primarily use reference material from the US Environmental Protection Agency (US EPA AP-42¹).

In addition to the national reporting, two important sets of more centralised emission data exist: the GAINS inventory of IIASA, and the inventories of TNO which are currently supported by and heavily used in the CAMS framework (Kuenen et al. 2014, Granier et al. 2019). These inventories provide profiles by country by source sector for precursors which include NMVOC, PM_{2.5} and PM₁₀. Both TNO and GAINS provide data aggregated to the SNAP² and/or GNFR³ emissions classifications. With regard to condensables and ongoing EMEP approaches, the TNO inventory (and especially so-called ‘Ref2’ scenarios) are of particular importance - this is discussed in Sect. 4.3.

4.1. Condensables in current EMEP emissions

As documented in Matthews et al. (2020), Parties to the Air Convention were asked to include a table with information on the inclusion of the condensable component in PM emission factor reporting in 2019. Seventeen Parties responded, though some reported the information only on an aggregated level. This reporting showed that in many cases Parties do not know if the PM emissions of a specific source category include the condensable component. For the majority

¹US EPA AP-42: Compilation of Air Emissions Factors: <https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emissions-factors>

²SNAP: Selected Nomenclature for Air Pollutants – an emissions source classification that was previously used for reporting to the Air Convention. See the “mapping table” here: <https://www.ceip.at/reporting-instructions>

³GNFR: Geographical NFR – see Annex V in <https://www.ceip.at/reporting-instructions/annexes-to-the-2014-reporting-guidelines>

of the source categories the information was either “unknown”, not given, or the provided information was unclear. This is often because they use default emission factors from the EI Guidebook, and the inclusion/exclusion of the condensable component is not always clearly defined.

The status of inclusion or exclusion is best known for the emissions from road transport. For example for “1A3bi Road transport passenger cars” ten of the twelve Parties that provided information for this source category report condensable emissions to be included and only two Parties state that the status of inclusion is unknown.

For RWC⁴, which accounts for 44% of reported PM_{2.5} emissions, the status was less clear. Of the thirteen Parties that provided information, three parties reported the condensable component to be included and three Parties to be excluded. The other Parties reported “unknown”, “partially included” or provided information on a more detailed level with a different status of inclusion (see Matthews et al. 2020, for further details).

One of the reasons for the low quality of information for the small combustion sector is the missing documentation for the PM emission factor of coal in the Guidebook. In that case it is unclear whether condensable organics are included or not. In terms of wood there is a clear indication that the Guidebook EF includes condensable components. Considering this aspect and including additional information from the sector specific chapters of the ‘informative inventory reports’ (IIRs), it can be assumed that 26 countries most likely include condensable organics in their PM emissions for residential wood combustion. Most of these countries are using default values. Some of them are using country-specific emission factors which are based on dilution tunnel measurements. There are only 6 countries which do explicitly report PM emissions excluding condensed compounds for residential wood combustion. All these countries are using country-specific emission factors which are based on heated filter measurements. There are still several countries where the situation is unclear. But there are also countries where a mix of different factors is used for different fuels or appliance types.

4.2. The EMEP/EEA Guidebook

The joint EMEP/EEA air pollutant emission inventory guidebook 2019 (EMEP/EEA 2019), hereafter referred to as the EI Guidebook, is the main source of technical guidance for preparation of national emission inventories for Parties to the Air Convention (with the exception of Canada and the USA). The EI Guidebook supports the reporting of emissions data under the Air Convention and the EU National Emission Ceilings Directive. The EI Guidebook is published by the EEA, with the Air Convention Task Force on Emission Inventories and Projections (TFEIP) responsible for the technical content of the chapters.

Although the EI Guidebook provides methodologies that can be used by each country, and is widely used, countries are free to use alternative methodologies if they can demonstrate that they are equally, or more, accurate in representing emissions. Given that EMEP has 51 parties, there is a wide diversity in detail and methodology across the Parties.

Until now, there has not been the need to assess whether the measurements that are used as reference material in determining PM emission factors from different sources include the

⁴RWC is associated with the GNFR sector “1A4bi Residential: Stationary”

condensable component. Furthermore, for many of the emission factors in the Guidebook the source material does not provide enough detail to be able to determine whether the measurements included or excluded the condensable component. So for many sources, retrospectively amending the emission factors quoted in the EI Guidebook is not a realistic task. However, it is possible to make some assumptions based on the knowledge that historically, particular measurement techniques have been used for different source sectors. For example, emission measurements for large combustion plants typically exclude the condensable component, and from road transport (and other smaller mobile sources) typically include the condensable component. Emissions from residential combustion typically make a large contribution to the condensables total, and unfortunately it is thought that historical measurements have used techniques which both include and exclude the condensable component, making it very difficult to interpret historical measurements, and some of the older emission factors in the EI Guidebook. Countries that have higher quality emissions inventories generally use country specific data to a much greater extent. The use of more recent and better characterised measurement studies means that they are much better able to identify whether emissions include or exclude the condensable component, compared to countries which rely heavily on information from the EI Guidebook.

4.3. The TNO approach and ‘Ref2’ scenarios

Denier van der Gon et al. (2015) demonstrated that current PM emission inventories for RWC in Europe do not account for wood combustion emissions in a consistent and comparable manner, and in particular that there is inconsistency in the emission factors used between countries. Denier van der Gon et al. (2015) generated a new emission inventory for RWC (termed then TNO-newRWC), in which they combined:

- A harmonised set of emission factors across Europe for RWC. The EFs, derived from the work of Nussbaumer et al. (2008a,b), were provided for both solid particles (SP) and for a case (DT) derived from dilution tunnel data where condensables were included (c.f. Table 3.1). On average the difference between the DT and SP cases was a factor 5.
- Activity data (wood usage and appliance types) were compiled for six appliance types from the GAINS and IEA data and from Klimont et al. (2002b) and Kupiainen and Klimont (2007).
- A new spatial distribution of RWC, based on statistics, surveys and country reporting, together with assumptions concerning urban/rural differences and local availability of wood (Visschedijk et al. 2009, Kuenen et al. 2014).

The result was a revised inventory with a consistent approach for residential wood combustion, independent of individual country emission factor choices used for official reporting. Denier van der Gon et al. (2015) noted that this TNO-newRWC inventory was a first-order approach because it neglects the importance of combustion conditions and “cultural” differences in how to burn wood. Nevertheless it leads to a more transparent and comparable emission inventory, which also led to improved performance of the chemical transport models in that study (see also Bergström et al. 2012).

More recently, the TNO estimates have been revised and improved (Denier van der Gon et al. 2020), and the new harmonised inventory has acquired the label ‘Ref2’ (to distinguish from the nationally reported data which can loosely be termed ‘Ref1’). Again it was noted that the revised TNO bottom-up estimate included substantial uncertainties for example concerning wood quality, appliance types in different countries, etc., but the merit is that it applies one consistent methodology to all countries. And again, model results were improved when applying the new inventory (Fagerli et al. 2020).

It should be noted that there are multiple factors that affect the ratio Ref1/Ref2, and just one of them is whether the emission factor includes condensables. Other factors include:

- appliance type splits may differ (old versus new stoves, share of open fireplaces, etc.)
- assumptions on burning practices (are the stoves operated in the proper way?)
- wood characteristics (dry/wet wood, type of wood)

All of these factors are probably different in Ref2 compared to national inventories. In addition, the activity data (how much wood is being burned in total in each country) can be different. Many countries may simply use the official statistics for this, while in some areas there may be a large proportion of the wood that is burned directly harvested from the forest (so not entering any statistics). Resolving these differences will require detailed comparison of activity and EF data from national inventories and TNO, and this work should form part of the short- and longer-term possibilities discussed in Sects. 5.1–5.3 below.

4.4. The USA

Emission inventories in the U.S. are very detailed with regard to sources and technologies. In the U.S., the EPA compiles and releases the National Emission Inventory (NEI) every 3 years. The NEI includes emissions for VOCs and PM for each county and for each emission source category. Emission sources are identified by Source Classification Codes (SCCs). EPA is responsible for setting emission reporting guidance, but ultimately, each individual state has responsibility for data they submit for the inventory process. This can lead to variability from state to state for how emissions in some source categories are calculated and attributed to SCCs.

While states are responsible for most emission source categories, there are a few exceptions, including large facilities, vehicles, and RWC. Large facilities, such as power plants, have their emissions directly measured using continuous emission monitors for CO₂, SO₂, and NO_x. For estimating mobile-source emissions, the U.S. EPA Office of Transportation and Air Quality develops and maintains the MOtor Vehicle Emission Simulator (MOVES5), a state-of-the-science modeling system that estimates national- and county-level emissions for criteria air pollutants, greenhouse gases, and air toxics. The most recent version, MOVES3, incorporates the latest data on vehicle populations, travel activity, and emission factors as well as updated fuel supply information at the county level. The model accounts for differences in the start and drive cycles, long-haul truck hoteling, off-network idling, and the implementation of recent requirements for emission control technologies. Lastly, for residential wood combustion, the EPA conducted a survey in 2018 that asked households about their wood burning appliances, how it is operated, and how much wood is burned. These data, along with county-level information about heating and cooling days, were used to estimate a regression model that predicts the prevalence and use of wood burning appliances for each county and for each relevant residential wood combustion SCC code in that county. While the each state is ultimately responsible for reporting emissions, for some source categories that can be standardized across states or are difficult to estimate comprehensively, EPA develops tools such as these to help standardize approaches, improve accuracy, and quantify variability. These are evolving methods and benefit from improved data from the scientific community.

Once all of the emission data are compiled into the National Emission Inventory, EPA applies a tool named SMOKE to create “Modeling Platforms”, which are the datasets needed for input to a chemical transport model. County level PM and VOC emissions from the NEI are allocated to the spatial grid and temporal timestep needed for the chemical transport model. PM and VOC emissions are assigned to the chemical species represented by the chemical transport model. This chemical speciation step is carried out using a database named SPECIATE, which maps each SCC code to a profile for that emission source. SPECIATE is described in the next section.

4.4.1 SPECIATE

The speciation of NMVOC and PM emissions is handled through the ‘SPECIATE’ database system (Bray et al. 2019, US EPA 2019, 2020). Version 5.0 of the database included major structural changes by merging common tables (PM, Gas, OTHER profiles) into one PRO-

FILES table, producing the following total number of profiles and unique species:

- 6,654 PM, GAS, and OTHER profiles;
- 2,814 unique species; and
- 198 PM-AE6 profiles.

which was increased to 6746 profiles in v5.1 (US EPA 2020). Table 4.1 summarises the types of profiles provided by the SPECIATE system.

SPECIATE 5.0 is the first version that includes profiles to support the VBS of CMAQ5.3 (<https://www.epa.gov/cmaq>). To do this, EPA added numerous fields to SPECIATE and a few SPECIATE PM-VBS and GAS-VBS profiles. The VBS serves to better characterize semi-volatile compounds in gas and PM profiles. The added fields include temp, RH, particle loading and organic loading of the sample, vapour pressure estimation (2 methods), and OM/OC ratio. Additionally, 20 VBS species (with C_i^*) were added. SPECIATE 5.0 also introduced profiles for mobile sources (vehicles, offroad mobile, and aircraft) that explicitly distribute organic mass among low volatility, semi-volatile, intermediate volatility and volatile organic compounds (LVOCs, SVOCs, IVOCs, and VOCs, respectively).

The use of these data could be usefully explored for Europe. While emission factors, activities and technologies can differ significantly between the US and Europe the VOC and VBS profiles can be rather similar in many cases and therefore the SPECIATE data might be extremely helpful to build on. Funding would be needed for projects to support data generation for some of the technologies and fuels specific to Europe though (e.g. gasoline particle filters and diesel passenger cars with SCR).

Table 4.1: Description of SPECIATE PROFILE types (from US EPA 2019)

Profile type	Definition ^(a)
GAS	Organic gas profiles. They can be TOG, NMOG, THC, VOC, and NMHC profiles, depending on the available species and analytical methods.
GAS-VBS	Organic gas profiles to support the volatility basis set (VBS) approach in air quality modeling such as CMAQ. These are typically profiles for which the raw measurement data are aggregated and/or non-measured species are derived from the measured species.
PM	Particulate matter (PM) profiles include data for PM of various size classes, such as PM _{2.5} , which represents the mass of particles from 0 to 2.5 microns in diameter.
PM-SIMPLIFIED	PM _{2.5} profiles that support the AE5 Aerosol Module in CMAQ.
PM-AE6	PM _{2.5} profiles that support the AE6 aerosol module in CMAQ. PM-AE6 profiles include additional species that are not typically measured such as PNCOM and particulate water.
PM-VBS	PM profiles to support the VBS approach in air quality modeling.
OTHER	OTHER profiles are those that do not fit in the organic gas or PM categories. Examples of the OTHER profiles are nitrogen oxides (nitric oxide (NO), nitrogen dioxide (NO ₂), nitrous acid (HONO)) and speciated mercury (elemental and oxidized mercury).

^(a) See US EPA (2019) for explanation of acronyms used here.

4.5. Issues with current inventory and reporting system

Many issues exist with the current reporting system, in the context of condensables, and for review of the Gothenburg Protocol:

- Many countries rely heavily on the content of the EI Guidebook. In some cases the EI Guidebook indicates that the condensable component is included or excluded. But in several cases this information is not available, and for many EFs it is not realistic to expect this to be retrospectively added. Consequently, the situation will only be improved when new measurements become available.
- It is possible to make the assumption that EFs for large stationary sources do not include the condensable component, and road transport (and other smaller mobile sources) do. However, this does not help to define whether residential combustion does – which is a particularly important source. The content of the EI Guidebook is being updated to address this, but the next release is not scheduled until 2023.
- It is also the case that emissions from the use of wood in residential combustion is a particularly challenging estimate to determine with accuracy. Obtaining accurate wood consumption statistics is challenging in many countries, and the emission factor is highly dependent on many variables, such as the appliance, the condition of the wood and even the skill of the operator. So even if the EF is known to include condensables, the amount included has large uncertainties.
- Under the Air Convention and the EU's National Emissions Ceilings Directive (NECD⁵), emissions are reported for e.g. the residential sector, along with consumption of different fuels, but as the emissions are not reported/resolved by fuel type, it is not possible to determine the implied EF (emission/GJ biomass⁶). Some, but not all, Parties provide this information in their accompanying report (IIR), but it is difficult to 'harvest' information from individual reports. It would be better if the information was provided in a convenient data format for inter-country comparisons, inventory review, or for other Parties to develop their own inventories.
- Greenhouse gas (GHG) reporting requires much more detailed information with emissions split by fuel type (and hence implied EFs, IEFs) for example. More detailed reporting of emissions and activity statistics by fuel and appliance type would bring significant benefits to several Air quality stakeholder groups. However, EFs for GHG emissions are simpler in that there is a clear correlation between CO₂ emission and the carbon content of a fuel. Emissions of air quality pollutants, on the other hand, depend not only on fuel quality but also on the combustion process and abatement technology.

The current NFR structure requires the reporting of data for each sub-sector and fuel group (solid, liquid, gaseous fuels and biomass). However, for several countries fuel data are still missing or incomplete. In several cases the quality of the fuel data is not

⁵<https://www.eea.europa.eu/themes/air/air-pollution-sources-1/national-emission-ceilings>

⁶Biomass includes wood, wood pellets, straw and other non-fossil solid organic material

satisfactory. The reporting of fuel-group-specific emissions would increase the complexity of the NFR tables and therefore the operational effort for the preparation process and the quality checks.

Thus, there is a quandary about whether to expand the NFR requirements to include more details and transparency about the EFs used, or whether to retain current approaches but encourage more complete reporting.

- Compliance with Gothenburg Protocol:

Changes in emissions methodology and estimates imply changes in the agreements made as part of the so-called Gothenburg Protocol of UN-ECE⁷.

An “Adjustments” mechanisms allows non-compliant Parties to demonstrate compliance with commitments, even if they are reporting science-based inventories, and that this process could be used if we change the reporting requirements relating to PM. However, the current Adjustments process is not ideal as it is rather involved and time consuming for both Parties and review teams. (The process will likely be reviewed during 2020-2021). It is also not possible for such procedure to cover all possible implications. For example, issues arise if countries change from excluding to including condensable organics in the PM reporting, or where ‘cleaner’ appliances are not as clean as expected when condensables are included in the emissions.

- Problems with Emission Limit Values (ELVs):

When reporting emissions to demonstrate compliance with ELVs, businesses are allowed to subtract the uncertainty range from the mid-point. These data are then often used in emissions inventories, which is not good practice (the mid-point should be reported). This is being addressed as far as possible through inventory reviews, but there are challenges (Parties often don’t know whether the data has had the uncertainty range subtracted or not).

The definition of emission limit values has to be considered. TFTEI has to review existing annexes to the GP, implementing mandatory ELVs. The time frame to include SVOC should be assessed.

- Speciation and missing emissions?

The current split of organic emissions into either PM or NMVOC is artificial, in that some compounds can partition to both the gaseous and particulate phases. Further, some compounds fall into the volatility gap between the two phases, and may have been omitted from both types of inventory. The EI Guidebook currently includes NMVOC speciation profiles for different sources. This could be extended to include speciation profiles for e.g. SVOCs, IVOCs, in principle, but then the allocation of condensables (SVOC and even some IVOC) to PM or NMVOC needs to be defined in a clear way.

⁷1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, <https://www.unece.org/environmental-policy/conventions/envlirtapwelcome/guidance-documents-and-other-methodological-materials/gothenburg-protocol.html>

4.6. Future inventories and links to modelling

It has been suggested (e.g. Donahue et al. 2006) that instead of emissions being reported as either PM or NMVOC, what are really needed are emissions of compounds distributed across volatility-space; Donahue et al. (2012) gives for example an estimate of the emissions of the USA in such a scheme. However, as noted above, such estimates require both emission measurements and assumptions concerning the thermodynamic properties of the condensables. So far there is very little data with which to construct such an inventory for Europe, so there is a clear need for new measurements, as well as new ideas on how to constrain such an estimate.

And while we strongly argue for more and new measurements, we also realise that this is not going to solve the problem in the short term. In the meantime a pragmatic compromise should be found to make sure that European policies address the right sources and optimise the impact of emission factor assumption for air quality policies.

It should also be noted that if the relative importance of certain sources remains ‘hidden’ in the primary PM inventories, important emission reduction policies/instruments such as the NEC directive will not directly tackle these sources — causing clear problems for mitigation and abatement. One danger is that if the treatment of condensables is left entirely to modelling groups, it may be used as a form of bias correction which leads to better predictive skills but not necessarily quantifying sources correctly, and thus it will degrade the ability of both the models and policy instruments with regard to air quality assessments and ultimately PM exposure reduction.

5. Discussion and conclusions

The main intention of the NMR condensables workshop was to bring together experts in the fields of emissions, measurements, emission inventories, and policy from Europe and North America, and to create a much better understanding of the issues and possible approaches for dealing with this important class of compounds. In this respect the workshop was very successful.

The issues are, however, very complex, and further progress should involve many communities and Parties to the Air Convention. We cannot give a recommendation here on ‘how-to-define’ condensable emissions and emission factors as the issues are just too complex, and EMEP must find a system that can be accepted and applied across Europe and North America. Much work is still needed to improve the understanding of condensables in emission inventories. We recommend however that these issues are brought into the open, and that a new system is devised that provides more reliable data for modelling, and which ensures transparency in the methodologies and assumptions used to tackle the grey zone that exists between particle and vapour phases of organic compounds, and their classification as either PM_{2.5} or NMVOC.

The key messages from the workshop have been provided in Sect. . In addition, the meeting identified some short term actions, some longer term, and also suggested a framework for a road-map for future progress. These are discussed below.

5.1. Short-term possibilities (2020–2021)

A number of activities have already started, or can be undertaken in the next year or so if the various Task Forces and experts have resources:

1. The workshop experts agreed that the TNO Ref2 emission inventory is a good first no-regret step for describing condensable emissions from residential wood combustion in emission dispersion modelling. It is to be considered as a temporary gap-filling strategy, necessary to provide better match between air quality measurements and air quality modelling,
2. However, increased transparency of the basis for TNO Ref2 is necessary, and these efforts need to be complemented by also studying the actual activity data (including fuel use) in residential wood combustion.

3. The extent to which condensables are included in the current inventories needs to be assessed. In addition, the methods/assumptions which lie behind the reported data need to be understood.
4. The best available parameters to construct a harmonised expert database for condensable needs to be assessed.
5. Need to consider which changes would be needed for future EI Guidebook changes, with regard to reporting templates and PM speciation.
6. Priority should be put first on the residential /wood combustion sector. The focus should be primarily on 1) harmonising the PM emissions factors used by the Parties, 2) improving activity data collection, and 3) compiling reliable information about the type of appliances used and fuel quality.
7. Need to identify sources other than RWC and road transport that have a significant potential to contribute to condensable PM.
8. Identify priorities for further research to reduce remaining potential systematic biases and increase scientific credibility.
9. Options need to be formulated to deal with the political consequences of changes in the emission reporting (with regard to existing national legislation, policies and reduction obligations).

5.2. Longer-term possibilities

Ideally we would deal with organic emissions as a spectrum ranging from inert PM mass to volatile gases, with the inventory providing emissions for each VBS bin (or explicit VOC compound). However, we also need to recognise the limited resources, time and data available to many inventory developers, and to avoid disrupting established and well-functioning links between scientific approaches and policy applications of EMEP.

Three possible approaches were suggested for improved inventory reporting in a medium- to longer-term reporting system, which all have pros and cons:

1. Report explicitly the solid and condensable organic fractions of POA included in the $PM_{2.5}$ emissions, i.e. FPOA and CPOA.
 - Pros: simple in concept. The EI Guidebook could be adapted. Easy to see if condensables are included or not. And to what extent.
 - Cons: the definition of CPOA, and the amount included, is still open and difficult. Even asking for BC alongside national PM inventories in recent years gave very variable results.
2. Report VBS-based emissions (e.g. LVOC, SVOC, IVOC or ideally for each C_i^* bin) and other NMVOC separately.

- Pros: makes sure that all organic emissions can be captured in the system. Provides a practical distribution of volatility properties that can be used in chemical transport modelling.
- Cons: this system is significantly different to the current EMEP emissions system. We do not know many of the emitted compounds (especially in the SVOC and IVOC fractions). The capture of some (especially gas-phase) SVOC and IVOC components requires advanced sampling, or assumptions based upon VBS distributions in combination with knowledge of the experimental conditions.

One possibility here would be to develop a web-interface where the analyst plugs in the calculated OM EFs, the measured concentration of OA and the temperature and the interface calculates the total condensable EFs based on a given volatility distribution. NMVOC EFs should also be used here to constrain the entire volatility distribution, and the complete volatility distribution should be the same as that used in models. This ensures consistency and comparability between results. This is indeed based on the assumption that the volatility distribution does not vary significantly between emissions, which may be a fair assumption when compared to variability in total emission factors.

3. Define an effective ambient PM emission factor, PMEA, which defines emissions at a standard temperature and ambient C_{OA} concentration.
 - Pros: allows consistent and harmonised PMEA factors across many sources (or for EMEP, countries) comparison. Gives a basis to calculate emissions at ambient temperature and C_{OA} values (models could do this).
 - Cons: PMEA is an emission potential, and not a real emission rate. Countries might find the concept too artificial. Calculated emissions in ambient conditions would be sensitive to assumptions concerning VBS behaviour. Does not capture the full quantity of SVOCs or IVOC which are needed by models to correctly predict partitioning. We do not know the volatility distributions for all sources that would be required, so it's probably not something that is ready to be operationalized at this time without some more very careful consideration.

These ideas represent differing ranges of practicality versus scientific completeness and flexibility. Option (1) is probably the easiest to consider within the next few years, since all approaches to include and improve the treatment of condensables (including use of the TNO Ref2 inventories) require estimates of both the solid and condensable fractions. All improvements will however pave the way towards better inventories in the future. Indeed, if efforts are made in the short to medium term for better and more transparent calculations of PM, the resulting databases and methodologies should improve the situation with condensables, and indeed other components of PM emissions, such as BC, to a significant extent.

5.3. The way forward

It is obvious that the issues surrounding condensables and national reporting are complex, and many aspects require both further discussion and more scientific knowledge. The workshop

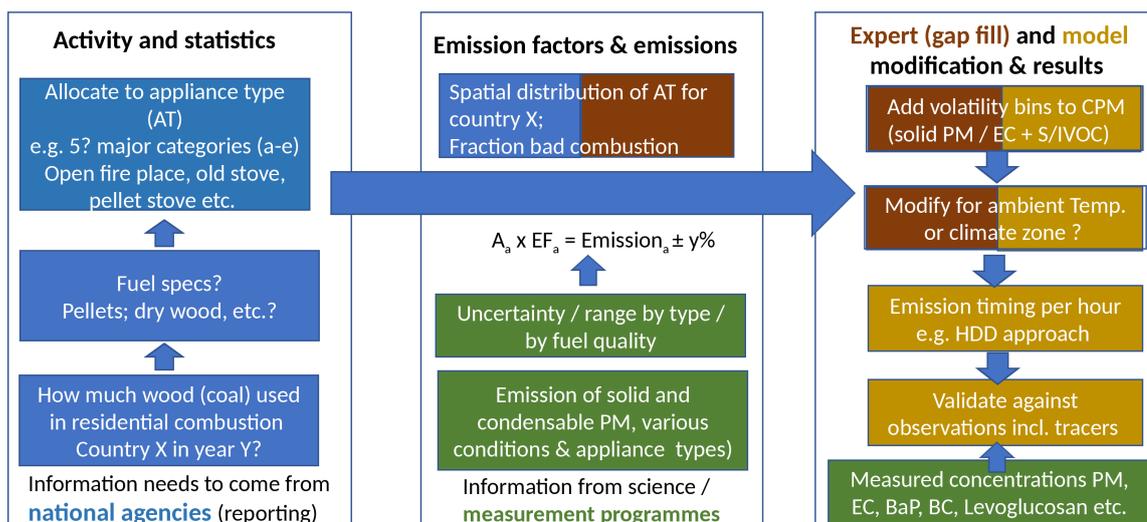


Figure 5.1: Flow-chart of emission factor components and possible responsibilities. Colours indicate different interdisciplinary groups: agencies, measurements, emission experts, and modellers.

has tried to identify both short term and long term approaches to improving the treatment of condensables in the EMEP system.

In order to clarify the roles of the involved parties, Fig. 5.1 was presented as a basis for further discussion. Briefly, the three main components of the emission inventory construction of Fig 5.1 are foreseen to be:

Activity and Statistics: here the national agencies (and/or we include international programmes) are responsible for collecting the basic activity data on fuels and technologies.

Emission factors and emissions: here national agencies have to combine activity data with emission factors derived from measurement programmes, guided by expert knowledge (which would include the EI Guidebook). At this stage the parties need to decide which emission factors are used, and in which way condensables are included.

Expert (gap fill) and model modification & results: in this third component, expert knowledge is likely applied to fill gaps in the emission data, and bring about complete and harmonised (as far as possible) emissions of the agreed POA emissions. Assuming that condensables are included in POA, the emission experts and chemical transport modellers need to assign volatility distributions, which might modify the extent to which the POA evaporate, and how they are treated in subsequent stages of atmospheric dispersion and oxidation.

There are many sources of confusion and discrepancy in the above, so a cyclic procedure was foreseen:

- In year 1 the TNO Ref2 data is used in an initial estimate for residential combustion emissions, with modellers making educated choices about SVOC emissions and the VBS framework

- (b) In subsequent years these top-down estimates should be increasingly replaced by national estimates once procedures for quantifying condensables in a more harmonised way are agreed on and implemented.
- (c) Such improvements will need detailed discussion among the emission inventory communities (e.g. TFEIP, TFTEI, national experts) as well as with modellers who will have to account for the complex volatility issues surrounding the condensables
- (d) Approach/updates should be tied to EMEP TFEIP meetings, etc

This approach will need guidance, coordination and support! Voluntary and/or country-specific contributions will lead to new mixtures of inconsistent assumptions. This can likely best be done within the frameworks of TFEIP, TFTEI, and the EI Guidebook, but the modelling and policy-oriented communities should be involved.

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Table A1: Workshop and/or report contributors^(a)

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^(a) Unless otherwise denoted, both workshop and report; ^(b) see also Table 2 for some acronyms; ^(c) Report only; ^(d) Workshop and extensive discussions

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