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New European AQ Directive: Volatile Organic Compounds

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ACTRIS CiGas

REVISION OF EU AMBIENT AIR QUALITY DIRECTIVES: "CLEANER AMBIENT AIR BY 2030, ZERO POLLUTION AIM BY 2050" - VOC

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The proposed revision of the Ambient Air Quality Directives will set **interim 2030 EU air quality standards,** aligned more closely with World Health Organization guidelines, while putting the EU on a trajectory to achieve **zero pollution for air at the latest by 2050,** in synergy with climate-neutrality efforts.

Press release on 26 October 2022: European Green Deal: Commission proposes rules for cleaner air and water

Directive 2008/50/EC

11.6.2008 EN Official Journal of the European Union L 152/29

ANNEX X

MEASUREMENTS OF OZONE PRECURSOR SUBSTANCES

A. **Objectives**

The main objectives of such measurements are to analyse any trend in ozone precursors, to check the efficiency of emission reduction strategies, to check the consistency of emission inventories and to help attribute emission sources to observed pollution concentrations.

An additional aim is to support the understanding of ozone formation and precursor dispersion processes, as well as the application of photochemical models.

B. **Substances**

Measurement of ozone precursor substances shall include at least nitrogen oxides (NO and NO_2), and appropriate volatile organic compounds (VOC). A list of volatile organic compounds recommended for measurement is given below:

	1-Butene	Isoprene	Ethyl benzene
Ethane	Trans-2-Butene	n-Hexane	m + p-Xylene
Ethylene	cis-2-Butene	i-Hexane	o-Xylene
Acetylene	1,3-Butadiene	n-Heptane	1,2,4-Trimethylebenzene
Propane	n-Pentane	n-Octane	1,2,3-Trimethylebenzene
Propene	i-Pentane	i-Octane	1,3,5-Trimethylebenzene
n-Butane	1-Pentene	Benzene	Formaldehyde
i-Butane	2-Pentene	Toluene	Total non-methane hydrocarbons

C. **Siting**

Measurements shall be taken in particular in urban or suburban areas at any monitoring site set up in accordance with the requirements of this Directive and considered appropriate with regard to the monitoring objectives referred to in Section A.

- **Only NMHC, TNMHC, and formaldehyde** (although not widely measured in Europe)
- **Oxy-VOCs?**
- **Terpenes?**

Only benzene is regulated (annual average < 5 µg/m^3) - Directive 2000/69/CE

Directive 2008/50/EC: 30 ozone precursors

		Butane	*Natural gas, fuel evaporation*	**Trans-2-butene**	*Combustion, fuel evaporation*	**1,3,5-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*
		i-pentane	*fuel evaporation*	**Cis-2-butene**	*Combustion, fuel evaporation*	**Isoprene**	*Biogenic mainly, combustion*
Formaldehyde	*Photochemistry of isoprene, anthropogenic*	**Pentane**	*fuel evaporation*	**1-pentene**	*Combustion, fuel evaporation*		
		i-hexane	*fuel evaporation, exhaust emissions*	**Trans-2-pentene**	*Combustion, fuel evaporation*		
		Hexane	*fuel evaporation, exhaust emissions*	**Cis-2-pentene**	*Combustion, fuel evaporation*		
		Heptane	*fuel evaporation, exhaust emissions*	**Benzene**	*Combustion*		
		i-octane	*fuel evaporation, exhaust emissions*	**Toluene**	*Combustion, fuel evaporation, solvent use*		
		Octane	*fuel evaporation, exhaust emissions*	**Ethylbenzene**	*Combustion, fuel evaporation, solvent use*		
Ethyne	*Combustion*	**Ethene**	*Combustion*	**M,p-xylenes**	*Combustion, fuel evaporation, solvent use*		
Ethane	*Long-lived species, natural gas*	**Propene**	*Combustion*	**O-xylene**	*Combustion, fuel evaporation, solvent use*		
Propane	*Long-lived species, natural gas*	**1,3-butadiene**	*Combustion, industrial sources*	**1,2,4-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*		
i-butane	*Natural gas, fuel evaporation*	**1-butene**	*Combustion, fuel evaporation*	**1,2,3-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*		

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AQUILA Air Quality Reference Laboratories

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Chemical family	Substance			
	Trivial name	IUPAC name	Formula	CAS number
Alcohols	Methanol	Methanol	CH_4O	67-56-1
	Ethanol	Ethanol	C_2H_6O	64-17-5
Aldehyde	Formaldehyde	Methanal	CH_2O	50-00-0
	Acetaldehyde	Ethanal	C_2H_4O	75-07-0
	Methacrolein	2-Methylprop-2-enal	C_4H_6O	78-85-3
Alkynes	Acetylene	Ethyne	C_2H_2	74-86-2
Alkanes	Ethane	Ethane	C_2H_6	74-84-0
	Propane	Propane	[illegible]	74-98-6
	n-Butane	Butane	[illegible]	106-97-8
	i-Butane	2-Methylpropane	[illegible]	75-28-5
	n-Pentane	Pentane	[illegible]	109-66-0
	i-Pentane	2-Methylbutane	[illegible]	78-78-4
	n-Hexane	Hexane	[illegible]	[illegible]0-54-3
	i-Hexane	2-Methylpentane	[illegible]	[illegible]-83-5
	n-Heptane	Heptane	[illegible]	142-82-5
	n-Octane	Octane	[illegible]	111-65-9
	i-Octane	2,2,4-Trimethylp[illegible]	[illegible]	540-84-1
Alkenes	Ethylene	Ethe[illegible]	[illegible]	75-21-8
	Propene / Propylene	Pr[illegible]	[illegible]	115-07-1
	1,3-Butadiene	Bu[illegible]	[illegible]	106-99-0
	1-Butene	[illegible]	[illegible]	106-98-9
	Trans-2-Butene	[illegible]	C_4H_8	624-64-6
	cis-2-Butene	[illegible]	C_4H_8	590-18-1
	1-Pentene	[illegible]	C_5H_{10}	109-67-1
	2-Pentene	[illegible]	C_5H_{10}	627-20-3 (cis-2 pentene) 646-04-8 (trans-2 pentene)
Aromatic hydrocarbons	Benzene	[illegible]	C_6H_6	71-43-2
	Toluene / Methylbenzene	[illegible]	C_7H_8	108-88-3
	Ethyl benzene	[illegible]	C_8H_{10}	100-41-4
	m + p-Xylene	[illegible]ne (m-Xylene) 1,4-[illegible]nzene (p-Xylene)	C_8H_{10}	108-38-3 (m-Xylene) 106-42-3 (p-Xylene)
	o-Xylene	1,2-Dim[illegible]benzene (o-Xylene)	C_8H_{10}	95-47-6
	1,2,4-Trimethylebenzene	1,2,4-Trimethylbenzene	C_9H_{12}	95-63-6
	1,2,3-Trimethylebenzene	1,2,3-Trimethylbenzene	C_9H_{12}	526-73-8
	1,3,5-Trimethylebenzene	1,3,5-Trimethylebenzene	C_9H_{12}	108-67-8
Ketones	Acetone	Propan-2-one	C_3H_6O	67-64-1
	Methyl ethyl ketone	Butan-2-one	C_4H_8O	78-93-3
	Methyl vinyl ketone	3-Buten-2-one	C_4H_6O	78-94-4
Terpenes	Isoprene	2-Methylbut-1,3-diene	C_5H_8	78-79-5
	p-Cymene	1-Methyl-4-(1-methylethyl)benzene	$C_{10}H_{14}$	99-87-6
	Limonene	1-methyl-4-(1-methylethenyl)-cyclohexene	$C_{10}H_{16}$	138-86-3
	β-Myrcene	7-Methyl-3-methylene-1,6-octadiene	$C_{10}H_{16}$	123-35-3
	α-Pinene	2,6,6-Trimethyl-bicyclo[3.1.1]hept-2-ene	$C_{10}H_{16}$	80-56-8
	β-Pinene	6,6-Dimethyl-2-methylenebicyclo[3.1.1]heptane	$C_{10}H_{16}$	127-91-3
	Camphene	2,2-dimethyl-3-methylenebicyclo[2.2.1]heptane	$C_{10}H_{16}$	79-92-5
	Δ^3-Carene	3,7,7-Trimethylbicyclo[4.1.0]hept-3-ene	$C_{10}H_{16}$	13466-78-9
	1,8-Cineol	1,3,3 trimethyl 2 oxabicyclo[2,2,2]octane	$C_{10}H_{18}O$	470-82-6

45 VOC recommended for measurement depending on the objectives

Comments from ACTRIS and (RI-URBANS) to DG ENV and AQUILA on the proposal for reviewing the EC Air Quality (AQ) Directives - February 2023

“The Annex VII of the EC AQ Directive proposes an updated list of VOCs including oxygenated compounds and terpenes. VOCs are of interest considering their role in O_3 formation but also in Secondary Organic Aerosols (SOA) formation. The collocated measurement of VOCs together with UFP measurements can be recommended at **supersites** in order to better identify/apportion sources and understand processes of new particle formation and growth. Additional species can be measured depending on the objectives sought, and on the instrument deployed for VOCs measurements (like Proton Transfer Reaction Mass Spectrometry / PTR-MS). **ACTRIS implements a Topical Centre Unit for VOCs measurements able to provide the most suited recommendation for the specific VOCs to be measured.**”

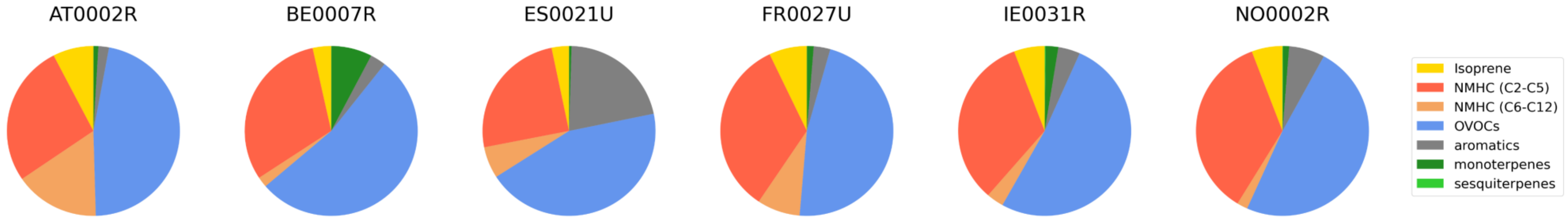
Recommendation #5: *Propose measurement protocols for VOCs species capitalising from the scientific and technical experience of CEN WG13, AQUILA and ACTRIS.*

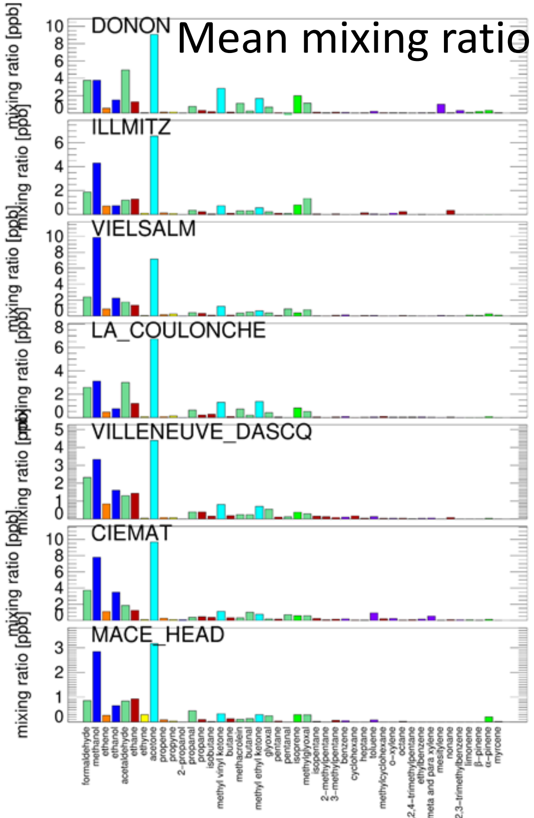
New suggested list: precursors of ozone and SOA

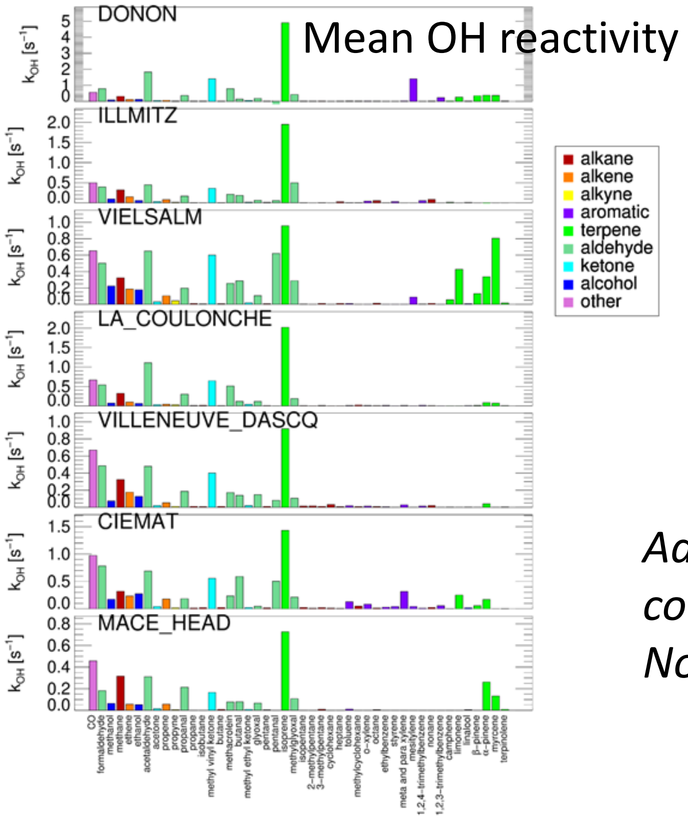
Methanol	***Biogenic, solvent use, combustion***	**Butane**	*Natural gas, fuel evaporation*	**Trans-2-butene**	*Combustion, fuel evaporation*	**1,3,5-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*
Ethanol	***Biofuels, solvent use***	**i-pentane**	*fuel evaporation*	**Cis-2-butene**	*Combustion, fuel evaporation*	**Isoprene**	*Biogenic mainly, combustion*
Formaldehyde	*Photochemistry of isoprene, anthropogenic*	**Pentane**	*fuel evaporation*	**1-pentene**	*Combustion, fuel evaporation*	**P-cymene**	***Biogenic mainly***
Acetaldehyde	***Photochemistry, solvent use***	**i-hexane**	*fuel evaporation, exhaust emissions*	**Trans-2-pentene**	*Combustion, fuel evaporation*	**Limonene**	***Biogenic mainly***
Methacrolein	***Photochemistry of isoprene***	**Hexane**	*fuel evaporation, exhaust emissions*	**Cis-2-pentene**	*Combustion, fuel evaporation*	**β-Myrcene**	***Biogenic mainly***
propanone	***Biogenic, solvent use***	**Heptane**	*fuel evaporation, exhaust emissions*	**Benzene**	*Combustion*	**α-pinene**	***Biogenic mainly***
MVK	***Photochemistry of isoprene***	**i-octane**	*fuel evaporation, exhaust emissions*	**Toluene**	*Combustion, fuel evaporation, solvent use*	**β-pinene**	***Biogenic mainly***
MEK	***Biogenic***	**Octane**	*fuel evaporation, exhaust emissions*	**Ethylbenzene**	*Combustion, fuel evaporation, solvent use*	**Camphene**	***Biogenic mainly***
Ethyne	*Combustion*	**Ethene**	*Combustion*	**M,p-xylenes**	*Combustion, fuel evaporation, solvent use*	**Δ-Carene**	***Biogenic mainly***
Ethane	*Long-lived species, natural gas*	**Propene**	*Combustion*	**O-xylene**	*Combustion, fuel evaporation, solvent use*	**1,8-cineole**	***Biogenic mainly***
Propane	*Long-lived species, natural gas*	**1,3-butadiene**	*Combustion, industrial sources*	**1,2,4-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*		
i-butane	*Natural gas, fuel evaporation*	**1-butene**	*Combustion, fuel evaporation*	**1,2,3-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*		

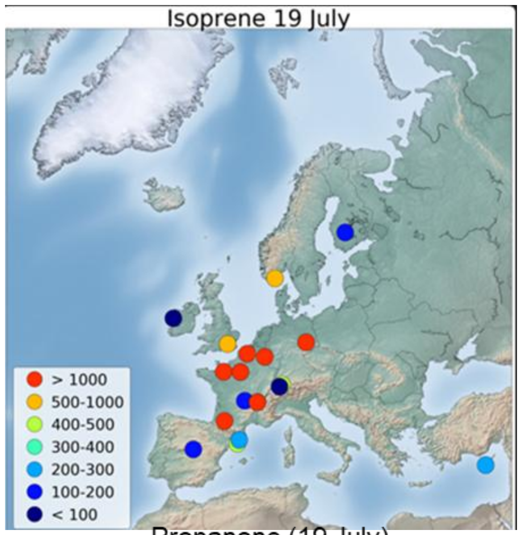
Importance of OVOC & terpenes measurements

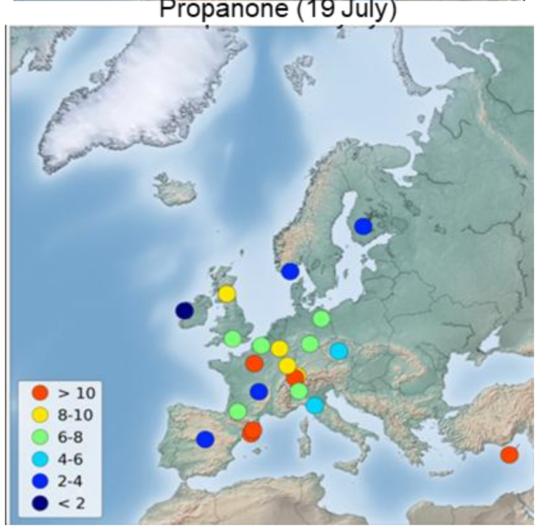
All the sites are dominated by OVOCs & C2-C5 NMHCs ; relative contribution does not vary very much between the sites with different typology











Aas et al. Under preparation, contribution from Julich, FMI, & IMT Nord Europe. EMEP report 2023

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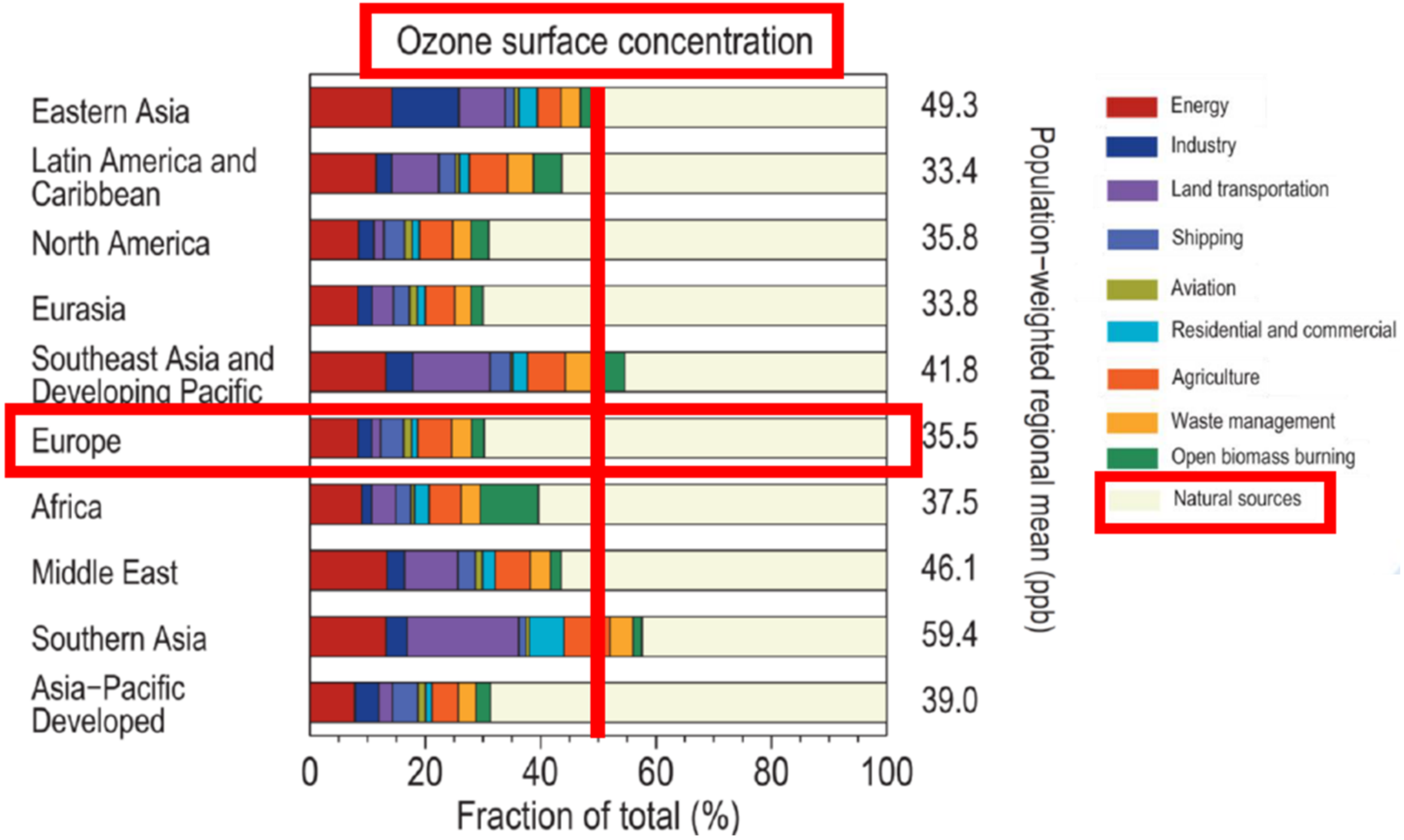
In Europe, 70 % of surface ozone is from natural sources *(Szopa et al., 2021)*



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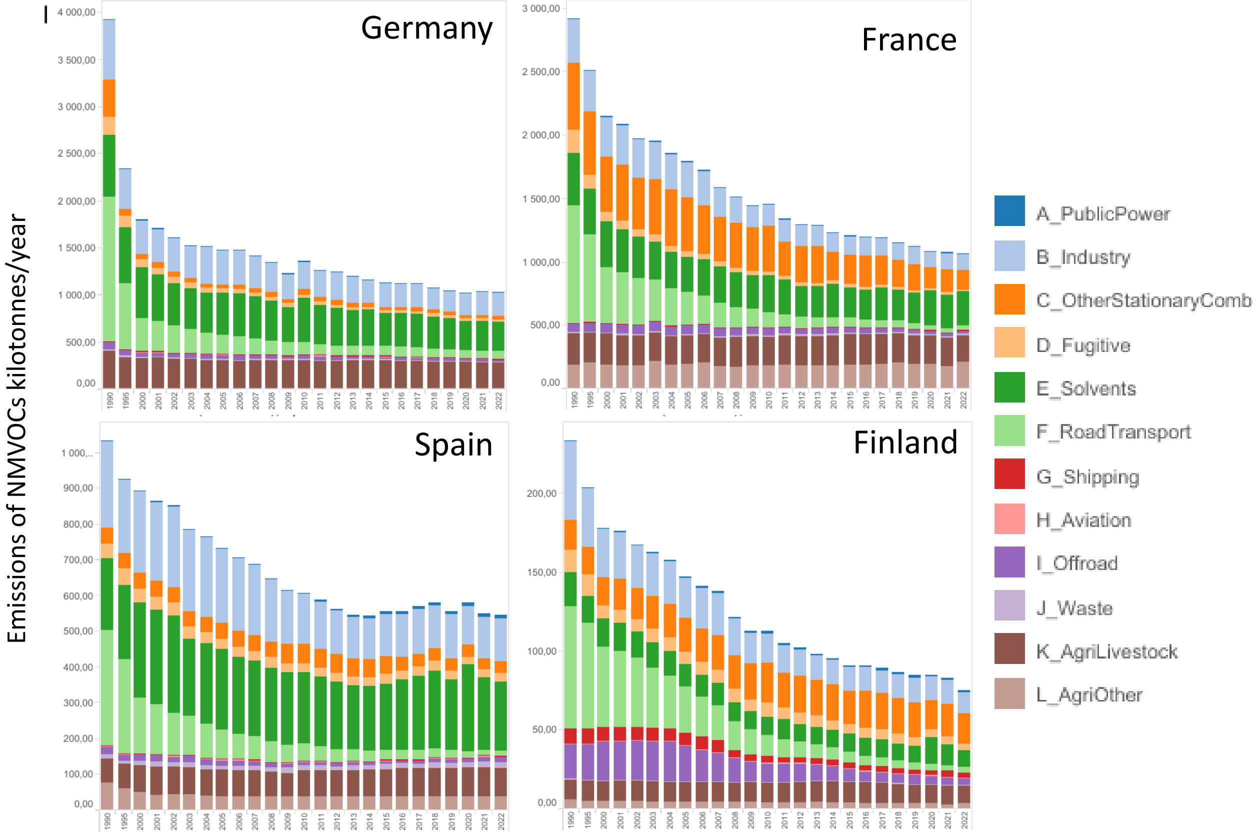
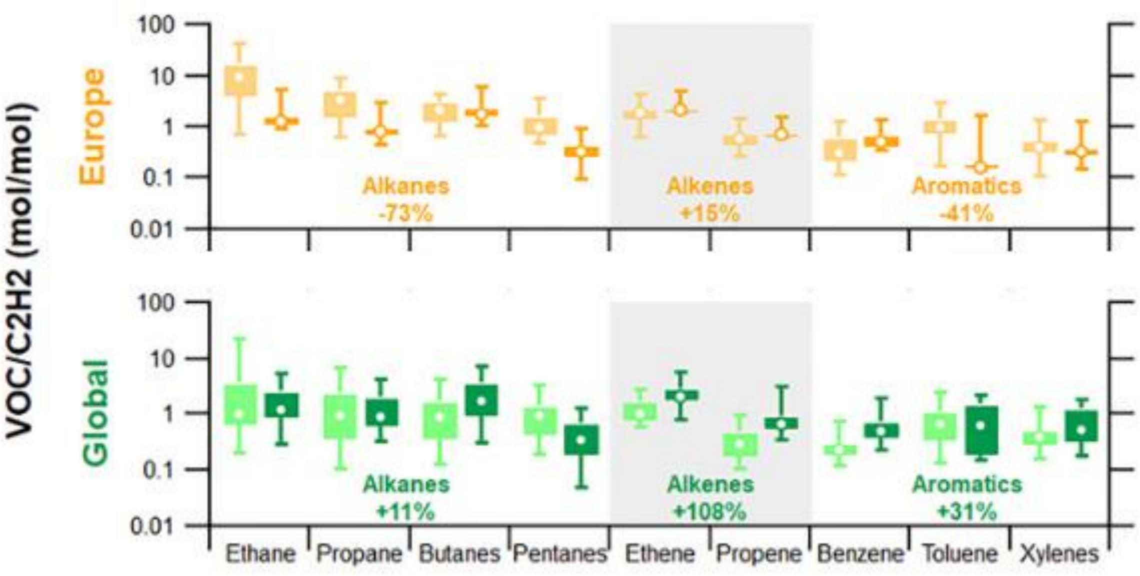
Decrease in VOC emissions, especially transport sector



Annual 1990–2022 emissions in kilotonnes of NMVOCs, showing a marked decrease of VOCs from road transport. Data from CEIP (2024).

But! remaining discrepancies with emission inventories ⇔ CTM models



Ratio COV/acétylène issus de plusieurs campagnes de mesures dans le monde (~80)(en clair) et de l'inventaire d'émission EDGAR v6.1 (en foncé). Les pourcentages indiquent les différences observées entre les mesures et l'inventaire d'émission EDGAR v6.1, ***von Schneidmesser et al., 2023***

VOC in the new AAQ Directive

Annex VII, Section 2, A.: the main objectives of measurements of ozone precursor substances are to:

- Analyse any trend in ozone precursors
- Check the efficiency of emission reduction strategies
- Check the consistency of emission inventories
- Support the understanding of ozone formation and precursor dispersion processes, as well as the application of photochemical models
- Help attribute emission sources to observed pollution concentrations

Member States may use the method which it considers suitable for the objective sought and methods that are being standardised by the European Committee for Standardization (CEN) shall be used once available.

MEASUREMENT OF VOC IS CHALLENGING: OVERVIEW OF VOC MEASUREMENT TECHNIQUES

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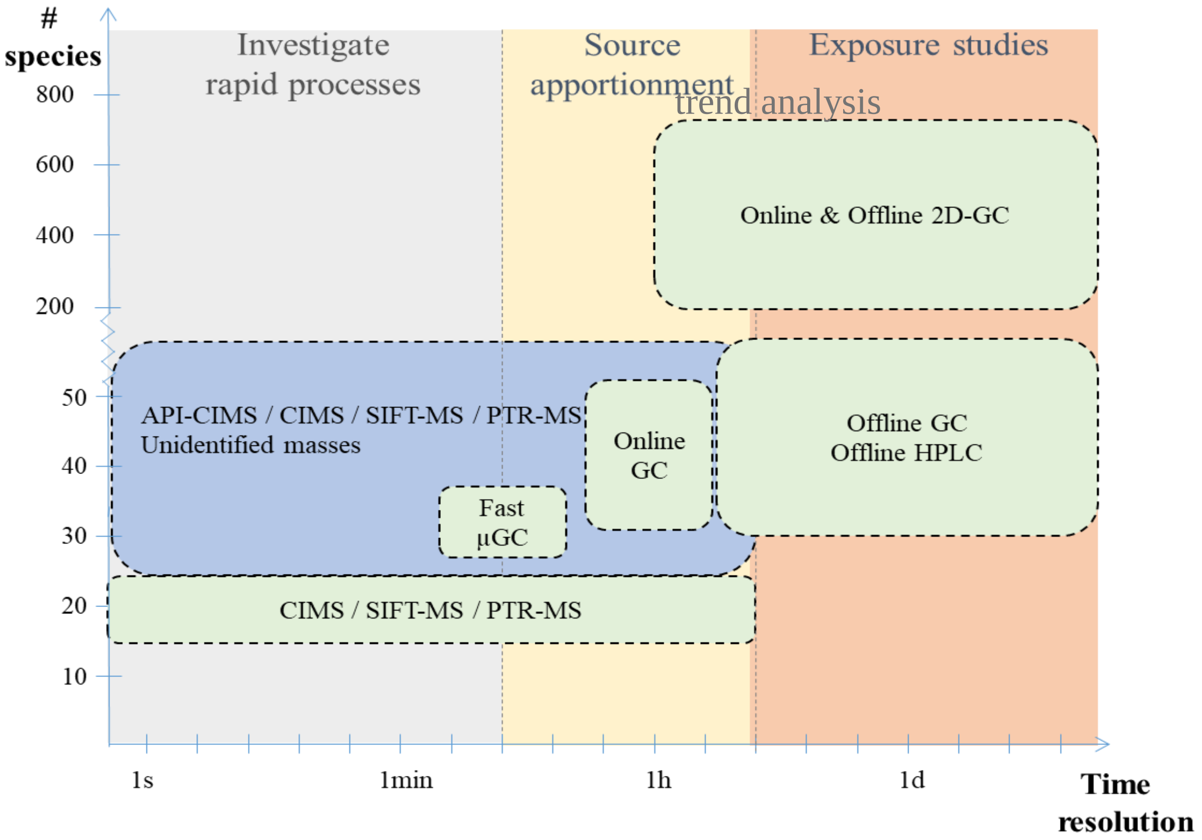

Additional techniques: Laser-based Systems (in-situ), remote sensing (FTIR), satellites…

*Schematic diagram of different techniques commonly used for ambient VOCs measurements. Technique performances are shown based on the number of speciated species and the measurement time resolution. Color coding corresponds to molecular and isomer identification (**green**), only molecular identification (**blue**). Units are in nmol/mol or µg/m³*

chemical ionization mass spectrometers (CIMS); proton transfer reaction mass spectrometry (PTR-MS); selected ion flow tube mass spectrometers (SIFT-MS); atmospheric pressure ionization CIMS (API-CIMS); gas chromatography (GC)

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VOC sensors:

- -Current evaluations show poor agreement with reference data
- -Current low-cost VOC sensors cannot distinguish between individual VOC species and instead report a measurement of total VOC (tVOC)
- -Development continues on speciated VOC measurements commonly with a focus on specific VOC species
- -Some successful applications include qualitative leak detection

ACTRIS deliverable 3.17. Updated Measurement Guideline for NOx and VOCs, 2018; WMO webinar Feb. 2021

EXISTING & ONGOING NORMS, GUIDELINES

Existing, upcoming norms, guidelines…

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No European reference methods available for VOCs (ongoing work), only for benzene

Only benzene is regulated (annual average < 5 µg/m³) - Directive 2000/69/CE
3.4 µg/m³ - Directive 2024/..

CEN-WG12, has published five norms related to **benzene** measurements:

- EN 14662-1:2024 : «Ambient air quality - Standard method for measurement of benzene concentrations - Part 1: Pumped sampling followed by thermal desorption and gas chromatography »
- EN 14662-2 :2005 : « Ambient air quality - Standard method for measurement of benzene concentrations - Part 2: Pumped sampling followed by solvent desorption and gas chromatography »
- EN 14662-3 :2015 : « Ambient air — Standard method for the measurement of benzene concentrations — Part 3: Automated pumped sampling with in situ gas chromatography »
- EN 14662-4 :2005 : « Ambient air quality Standard method for measurement of benzene concentrations - Part 4: Diffusive sampling followed by thermal desorption and gas chromatography »
- EN 14662-5 :2005 : « Ambient air quality Standard method for measurement of benzene concentrations - Part 5: Diffusive sampling followed by solvent desorption and gas chromatography »

Existing, upcoming norms, guidelines…

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CEN WG13 established in 2019, where ACTRIS contributes, with the objectives of developing validated standard measurement methods for the measurement and monitoring of **volatile organic ozone precursors in ambient air in order to ensure a harmonized implementation of the Directive 2008/50/EC**. Total non methane hydrocarbons are excluded. The Commission has requested a programme for Standard development for ozone precursors using the following techniques:

- Automatic pumped sampling, pre-concentration **and on-line gas chromatography** with flame ionisation detector (FID) and/or mass spectrometer detector (MSD);
- **Manual or automatic canister sampling** followed by off-line gas chromatography with FID and/or MSD;
- **Manual or automatic pumped sampling** followed by off-line thermal desorption and gas chromatography with FID and/or MSD;
- **Diffusive sampling** followed by thermal desorption by off-line gas chromatography with FID and/or MSD;
- Manual or automatic pumped sampling of **formaldehyde** on dinitrophenylhydrazine (DNPH) followed by off-line high-performance liquid chromatography (HPLC) / ultraviolet (UV) detection;
- Diffusive sampling of **formaldehyde** on DNPH followed by off-line HPLC/UV detection.

NEW DIRECTIVE: VOC LIST & SUGGESTED METHODS

New suggested list: How can we measure all these VOC species?

VOC(s)	Measurement technique – most common method or potential method	Primary standard (CCL)	CEN TS/norm	Existing guidelines/SOP – ACTRIS – WMO/GAW	Cost estimation (K€) for instrument purchase	Comments & remarks
NMHCs (alkanes, alkenes including isoprene, alkynes, aromatics)	Online measurement: GC-FID or FID/MS	NPL, UK	Ongoing work within CEN WG13	Yes, within ACTRIS and WMO/GAW=> input to CEN WG13	Around 100	Robust method, existing of compact GC; possibility to measure some terpenes; ozone scrubbers, filters, water elimination…
	Offline measurement: sampling with canisters, sorbent tubes; analysis with GC-FID/MS	NPL, UK	CEN WG12: **benzene** related norms for diffusive and pumped sampling + Ongoing work within CEN WG13	For canisters sampling by WMO/GAW	Around 100	A GC can serve multiple sites
	PTR-MS	NPL, UK – for PTRMS (Worton et al. 2023)	No	Ongoing within ACTRIS	~400-700	Mainly aromatics (ionisation mode); Price depending on the options; high time resolution => data treatment

New suggested list: How can we measure all these VOC species?

VOC(s)	Measurement technique – most common method or potential method	Primary standard (CCL)	CEN TS/norm	Existing guidelines/SOP – ACTRIS – WMO/GAW - EMEP	Cost estimation (K€) for instrument purchase	Comments & remarks
OVOC	Online measurement: GC-FID	VSL (ongoing development); alternatives	Ongoing work within CEN WG13 **mainly for NMHCs**	Planned within ACTRIS	Around 150	water treatment needed; all OVOC of the new list can be measured excluding HCHO; & NMHC
	Offline measurement: sampling with DNPH; analysis with HPLC-UV	Liquid (not primary) or gaseous standards	No	EMEP guideline; planned within ACTRIS	Around 100	A HPLC can serve multiple sites; price of one cartridge is 20 €; QA/QC measures; OVOC including ketones, aldehydes (HCHO), excluding alcohols
	PTR-MS	NPL, UK – for PTRMS (Worton et al. 2023)	No	Ongoing within ACTRIS	~400-700	Alcohols, aldehydes excluding HCHO, ketones; &aromatics, high time resolution => data treatment
HCHO	DNPH/HPLC or a specific analyser	No, mixture in cylinders is not stable; possibility of permeation method	Ongoing work within CEN WG13	Planned within ACTRIS	50 - 150	Different techniques available and under development, side-by-side intercomparison 2022

New suggested list: How can we measure all these VOC species?

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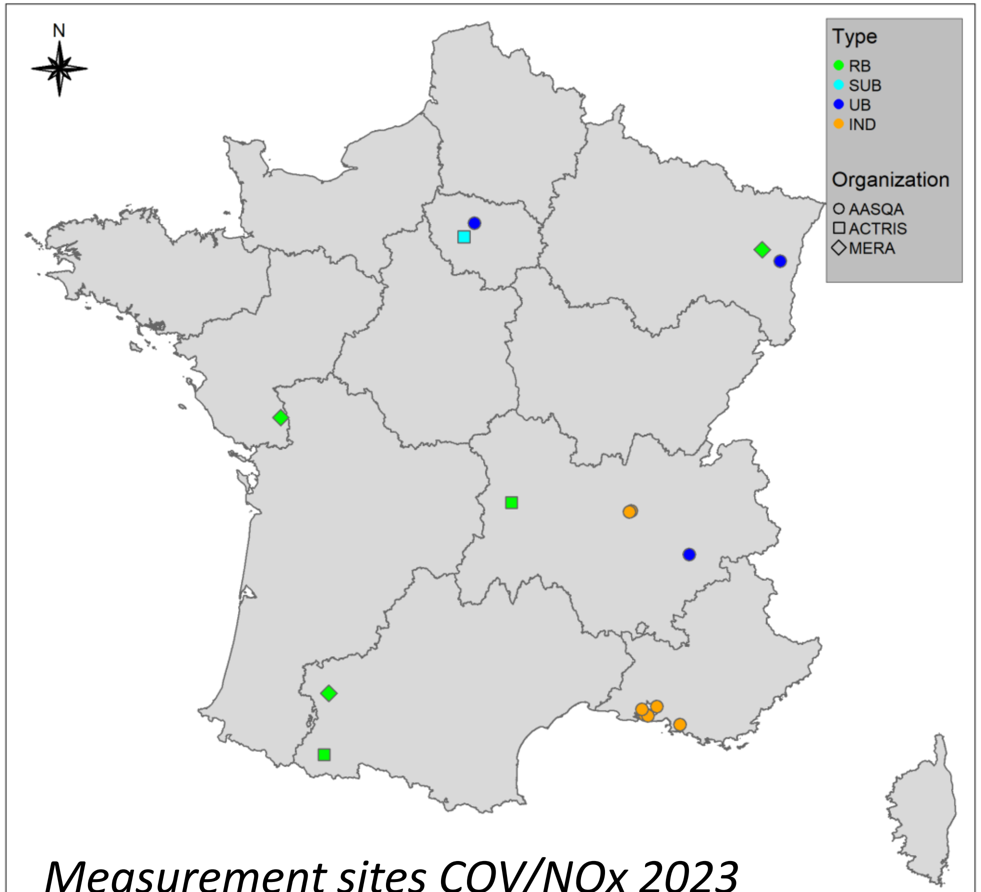
VOC(s)	Measurement technique – most common method or potential method	Primary standard (CCL)	CEN TS/norm	Existing guidelines/SOP – ACTRIS – WMO/GAW - EMEP	Cost estimation (K€) for instrument purchase	Comments & remarks
Terpenes	Online measurement: GC-FID or FID/MS	NIST; NPL for some terpenes can be applied	Not dedicated to terpenes	Planned within ACTRIS	Around 100-150	GC dedicated to terpenes; calibration standard not very stable
	Offline measurement: sampling with sorbent tubes; analysis with GC	Liquid (not primary) or gaseous standards	No		Around 100	A GC can serve multiple sites; QA/QC measures; Hellen et al. 2024
	PTR-MS	NPL, UK – for PTRMS	No	Ongoing within ACTRIS	~400-700	Sum of monoterpenes, no speciation; high time resolution => data treatment

THE CASE OF FRANCE

National strategy of VOC (&NOx, & methane) observations

Ongoing implementation



Measurement sites COV/NOx 2023

- Harmonized implementation of ozone & SOA precursors over France
- Twin-site : **Regional vs. Urban** contribution
- Source identification and contribution : **anthropogenic & biogenic**
- Evaluate and improve local emission inventories (=> CTM models)

VOC measured with the implemented TD-GC-FID system for NMHC and OVOC measurement in France

Methanol	***Biogenic, solvent use, combustion***	**Butane**	*Natural gas, fuel evaporation*	**Trans-2-butene**	*Combustion, fuel evaporation*	**1,3,5-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*
Ethanol	***Biofuels***	**i-pentane**	*fuel evaporation*	**Cis-2-butene**	*Combustion, fuel evaporation*	**Isoprene**	*Biogenic mainly, combustion*
Formaldehyde	*Photochemistry of isoprene*	**Pentane**	*fuel evaporation*	**1-pentene**	*Combustion, fuel evaporation*	**P-cymene**	***Biogenic mainly***
Acetaldehyde	***Photochemistry, solvent use***	**i-hexane**	*fuel evaporation, exhaust emissions*	**Trans-2-pentene**	*Combustion, fuel evaporation*	**Limonene**	***Biogenic mainly***
Methacrolein	***Photochemistry of isoprene***	**Hexane**	*fuel evaporation, exhaust emissions*	**Cis-2-pentene**	*Combustion, fuel evaporation*	**β-Myrcene**	***Biogenic mainly***
propanone	***Biogenic, solvent use,***	**Heptane**	*fuel evaporation, exhaust emissions*	**Benzene**	*Combustion*	**α-pinene**	***Biogenic mainly***
MVK	***Photochemistry of isoprene***	**i-octane**	*fuel evaporation, exhaust emissions*	**Toluene**	*Combustion, fuel evaporation, solvent use*	**β-pinene**	***Biogenic mainly***
MEK	***Biogenic***	**Octane**	*fuel evaporation, exhaust emissions*	**Ethylbenzene**	*Combustion, fuel evaporation, solvent use*	**Camphene**	***Biogenic mainly***
Ethyne	*Combustion*	**Ethene**	*Combustion*	**M,p-xylenes**	*Combustion, fuel evaporation, solvent use*	**Δ-Carene**	***Biogenic mainly***
Ethane	*Long-lived species, natural gas*	**Propene**	*Combustion*	**O-xylene**	*Combustion, fuel evaporation, solvent use*	**1,8-cineole**	***Biogenic mainly***
Propane	*Long-lived species, natural gas*	**1,3-butadiene**	*Combustion, industrial sources*	**1,2,4-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*		
i-butane	*Natural gas, fuel evaporation*	**1-butene**	*Combustion, fuel evaporation*	**1,2,3-trimethylbenzene**	*Combustion, fuel evaporation, solvent use*		

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CONCLUDING REMARKS

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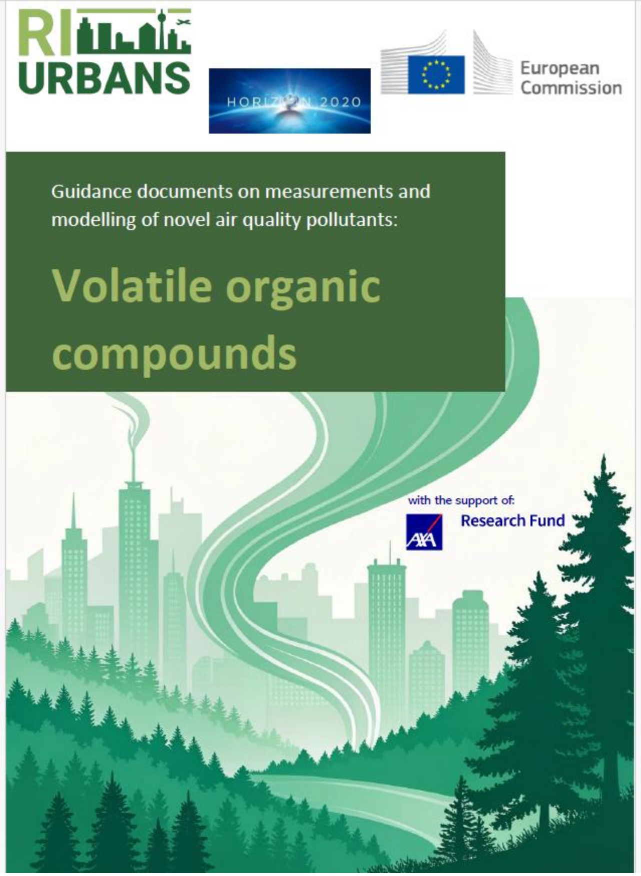
Key elements of good practice :

- A state-of-the-art quality assurance and quality control framework (QA/QC)
- Traceable network standards
- Suitable measurement guidelines and standard operating procedures (SOP)
- Trained personnel
- Regular audits, side-by-side intercomparisons, round robin exercises
- QA/QC workshops

Additional technical aspects for VOC measurements:

Water removal/management; Ozone removal; carbon dioxide removal; Particle filters; sampling lines materials

Additional QC*: use available online tools for trajectories calculations (NILU tool, Hysplit, ATMOACCESS tools, @VOC@ tool, TUCAVOC…)*



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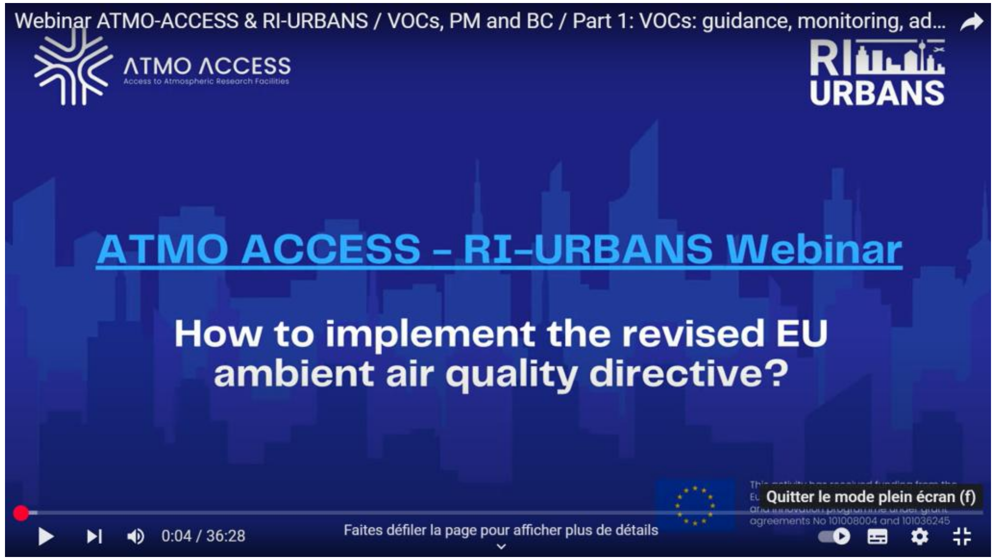

https://www.youtube.com/watch?v=ndDutvIWt6E

MERCI

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