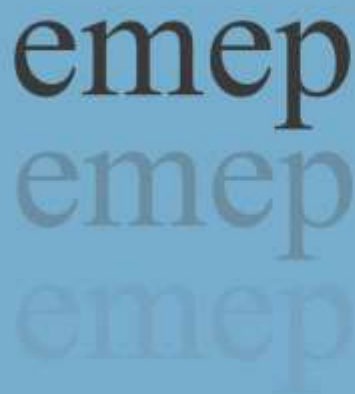


Convention on Long-range Transboundary Air Pollution



*Co-operative programme for monitoring
and evaluation of the long-range
transmission of air pollutants in Europe*

EMEP/CCC-Report 4/2025

VOC measurements 2023

Authors: Sverre Solberg (NILU), Paul D. Hamer (NILU), Anja Claude (Deutscher Wetterdienst) and Stefan Reimann (EMPA)

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EMEP/CCC-Report 4/2025

EMEP Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe

VOC measurements 2023

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EMEP/CCC-Report 4/2025

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Short summary (English)

This report documents the EMEP VOC monitoring carried out in 2023. The levels of the measured species in 2023 are presented as well as the 2023-status and history of the VOC programme. The geographical pattern of the species in Europe is discussed as well as the long-term trend during the last 20 years.

Short summary (Norwegian)

Denne rapporten dokumenterer EMEP VOC-overvåkingen utført i 2023. Nivåene av de målte komponentene i 2023 presenteres sammen med status i 2023 og historikken for VOC-programmet. Den geografiske fordelingen av komponentene i Europa diskuteres samt den langsiktige trenden de siste 20 årene.

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EMEP, VOC, NMHC, monitoring

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Summary

This report presents VOC (volatile organic compound) measurements carried out during 2023 at EMEP monitoring sites. In total, 19 sites reported VOC-data from EMEP VOC sites this year.

The monitoring of VOCs has become more diverse with time in terms of methods and instrumentation. Starting in the early 1990s with standardized methods based on manual sampling in steel canisters and adsorption tubes with subsequent analyses at the lab, the methods now consist of a variety of instruments and measurement principles, including automated continuous monitors and manual flask samples.

Within the EU infrastructure project ACTRIS, data quality issues related to measurements of VOCs are important topics. Most of the institutions providing VOC-data to EMEP are participating in the ACTRIS infrastructure, either as formal partners or on a voluntary basis. Participation in ACTRIS means an extensive effort with data-checking including detailed discussions between the ACTRIS community and individual participants. There is no doubt that this extensive effort has benefited the EMEP-program substantially and has led to improved data quality in general.

In general, the levels in 2023 agree well with the previous 5-years period, but the lack of sites with long and consistent time series poses a substantial problem for making reliable long-term trend assessments for VOCs. Breaks in the time series are caused by shifts in instrumentation/method or stations being closed or moved etc. In this report, we have estimated the trends in individual NMHCs over the 2001-2023 period for one single station (Hohenpeißenberg), using a so-called GAM (generalized additive model), that removes the effect of anomalies in the meteorology.

The results indicate substantial reductions in the observed levels of the NMHCs over the 2001-2023 period at this site. Furthermore, significant differences in the trends for the various species are found. The strongest declines are found for toluene, ethene, m-p-xylene and i-pentane which in 2023 are seen at levels only 35-45% of that in 2001 meaning a drop of the order of 55-65 % since 2001. The other NMHC have dropped to around 70-90% from the 2001 levels meaning a reduction of 10-30% during this 22-years period. These trend numbers refer to the trends adjusted for variations in meteorology.

These reductions are in line with the emission data from the so-called EMEP-West region although a quantitative comparison species-by-species could not be done since the EMEP emission data are given for the sum of aggregated NMHCs and not for individual substances.

VOC measurements 2023

1 Introduction

1.1 Historical background

The EMEP VOC monitoring programme was initiated at the EMEP Workshop on Measurements of Hydrocarbons/VOC in Lindau, 1989 (EMEP/CCC, 1990). A three-fold objective of the measurement programme was defined at the workshop:

- Establishing the current ambient concentrations
- Compliance monitoring (“Does the emission control programme lead to a reduction of atmospheric concentrations?”)
- Support to the transboundary oxidant modelling (prognostic and diagnostic)

The Workshop recommended that as a first step it would be sufficient with VOC monitoring at 10-15 rural sampling sites and taking two samples per week centred at noon GMT at each station. Collection in stainless steel canisters and analyses by high resolution gas chromatography was recommended for the detection of light hydrocarbons, whereas impregnated adsorbent tube sampling combined with high performance liquid chromatography (HPLC) was recommended for the detection of carbonyls.

VOC measurements within EMEP started with the collection of grab samples of light hydrocarbons in mid-1992 and measurements of carbonyls in 1993. Initially, five stations were included in the monitoring programme: Rucava (LV0010), Košetice (CZ0003), Waldhof (DE0002), Tänikon (CH0032) and Donon (FR0008). Since then, the number and selection of VOC measurement sites have changed several times.

EMEP VOC measurements are reported annually and presented in reports for consideration by EMEP-TFMM and the EMEP Steering Body. Previous results from the EMEP VOC programme have been presented in annual reports (e.g. Solberg, 2023 and references therein). An EMEP expert meeting on VOC measurements was organised in Berlin, 1994 (EMEP/CCC, 1995), and an evaluation of the measurement programme was made in 1995 (Solberg et al., 1995).

VOC-data from the EMEP-network have been published and documented in numerous publications, e.g. Waked et al. (2016), Hellen et al. (2015), Hoerger et al. (2015), Malley et al. (2015), Tørseth et al. (2012), Worton et al. (2012), Sauvage et al. (2009), Plass-Dülmer et al. (2009), Plass-Dülmer et al. (2002), Hakola et al. (2006), Borbon et al. (2004), Solberg et al. (2001), and Solberg et al. (1996). Details about the VOC monitoring program are given in the EMEP annual reports such as Solberg (2024) and references therein.

2 Status of the measurement programme

2.1 The station network

The locations of the EMEP monitoring sites for VOCs in 2023 are shown in Figure 1 and an overview of the measurement programme and the responsible laboratories is given in Table 1. In total, 18 measurement sites and 11 laboratories were included in the EMEP VOC program in 2023.

The measured VOCs consist of various groups of species which could be split into non-methane hydrocarbons hereafter named NMHCs and oxygenated species hereafter named OVOCs. Monitoring of NMHCs are carried out at all sites except ES0001, whereas OVOCs are measured at fewer sites.

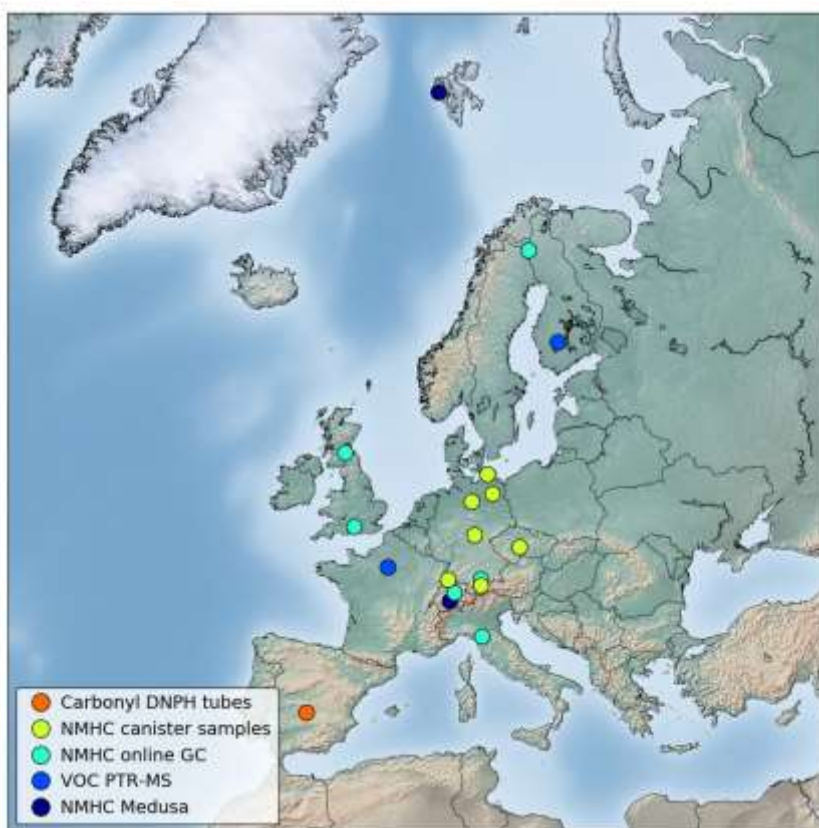


Figure 1: Monitoring sites for VOC in 2023.

The VOC monitoring at EMEP sites has become more diverse with time in terms of methods and instrumentation. This is illustrated in Figure 2. The EMEP VOC monitoring programme started in 1992 with one standardized method for NMHCs based on manual sampling in steel canisters and subsequent lab analyses and another standardized method for sampling of OVOCs by DNPH tubes followed by analyses with HPLC in the lab. Over the years a variety of instruments and measurement principles have been applied, including PTR-MS, Medusa monitors, specialized online GC monitors and adsorption tubes for aromatic species.

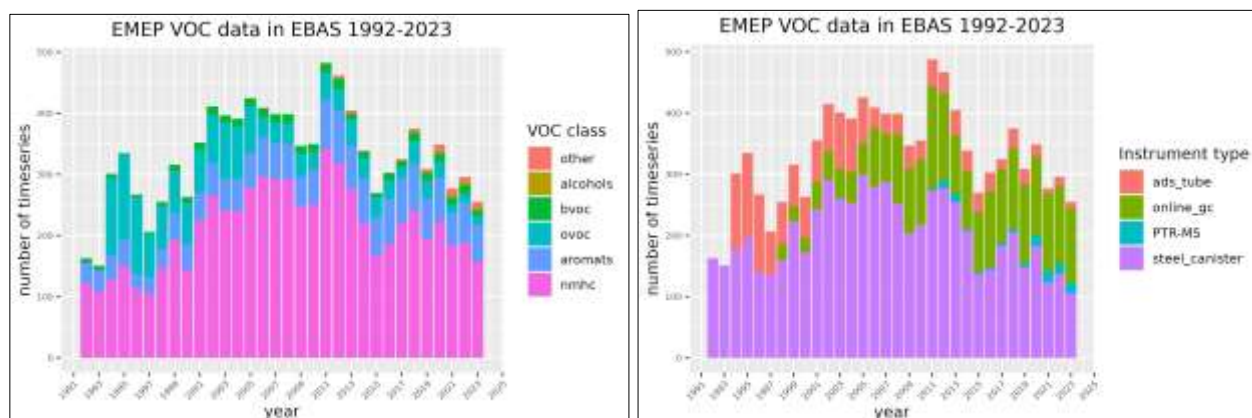


Figure 2: History of the VOC monitoring as a function of chemical group (left) and instrument (right).

The types of VOC species have also expanded from C_2 - C_6 NMHCs and a few light OVOCs in the start of the period, to a very wide spectrum of species including terpenes, alcohols, C_7 - C_9 NMHC, acetonitrile and numerous branched saturated and unsaturated NMHC as given in the Appendix.

For OVOCs the original EMEP method based on sampling in DNPH adsorption tubes with subsequent lab analyses is still the method used at one site in Spain (ES0001). In addition, OVOCs are also measured by PTR-MS and by the GC-GC FID/FID system at Beromünster in Switzerland.

Although many sites have contributed to the EMEP VOC programme since the early 1990s, very few sites have long and continuous time series. Additionally, shifts in instrumentation imply breaks in the time series. At some sites these shifts are a matter of upgrading the GC monitor, with minor effects on the measured values, while at other sites they represent significant breaks in the data time series. The VOC network has also changed due to stations being closed or moved (like e.g. CH0005 that was replaced with CH0053 in 2018).

The history of the VOC station network is illustrated in Figure 3 showing the monitoring at the various sites as a function of time since the onset in 1992. The changing monitoring network with few sites having a long-term continuous time series is clearly seen from this figure and poses a major obstacle for doing long-term trend studies.

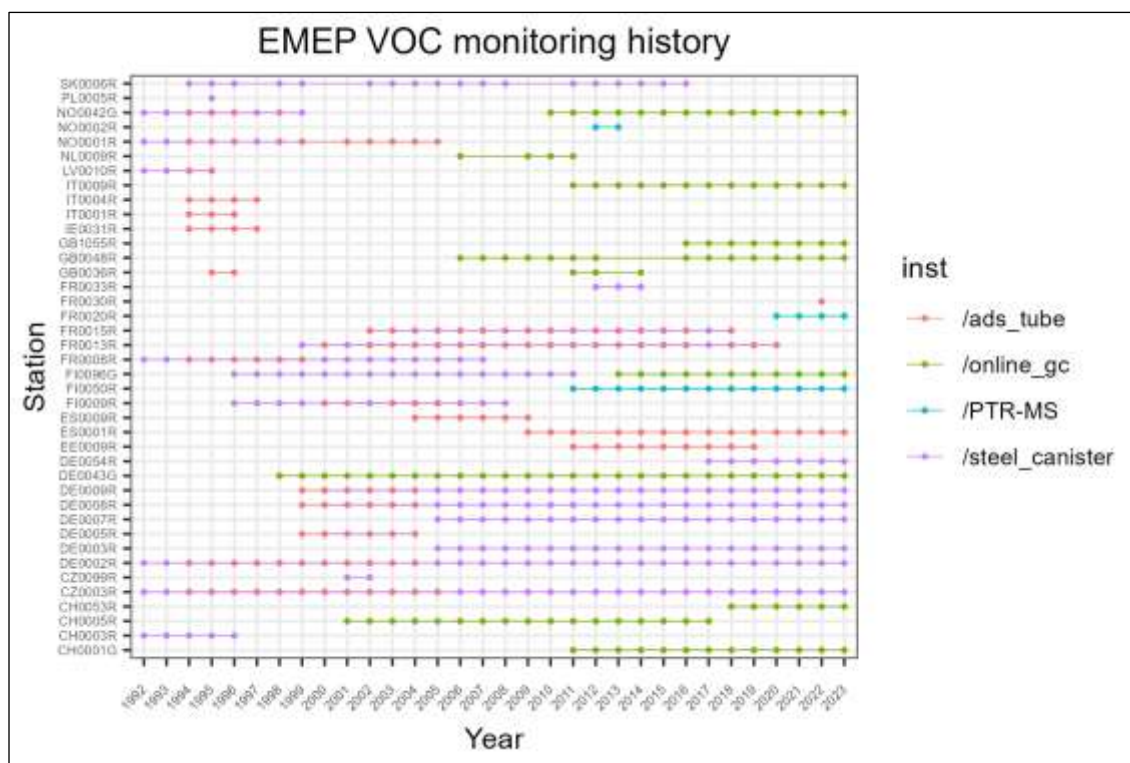


Figure 3: Monitoring history as a function of site 1992-2023.

All VOC data are reported annually to EBAS at NILU/EMEP-CCC through the EBAS submission tool (<https://ebas-submit-tool.nilu.no/>). For sites taking part in ACTRIS (see Table 1), the initial data submission is followed by a rigorous data quality checking procedure through an issue tracker (<https://mantis.nilu.no/>), involving a dedicated QA tool that is based on ratios of species (e.g. ethane:propane) and comparisons of the levels at different sites. Based on the results from this tool, the ACTRIS topic centre adds comments and questions to the data series followed by a discussion with the data provider and NILU/EMEP-CCC regarding outliers, unexpected shifts in the data etc., and then, finally, which data to invalidate or flag. Since this discussion is done in an issue tracker, the history of bugs, outliers and breaks in the time series is easily available at a later stage.

This QA system has been in operation for some years and has clearly led to an improvement in the overall data quality of the part of the monitoring program included in ACTRIS. The ACTRIS community is open to include non-ACTRIS sites in this exercise which has been the case for, e.g., the sites run by UBA in Germany.

An example of the QA tool for data from DE0054 (Zugspitze) and DE0043 (Hohenpeissenberg) is given in Figure 4, showing submitted data for ethene and ethyne for 2023 together with previous years of data. The marked outliers in the plot were inspected by the data providers.

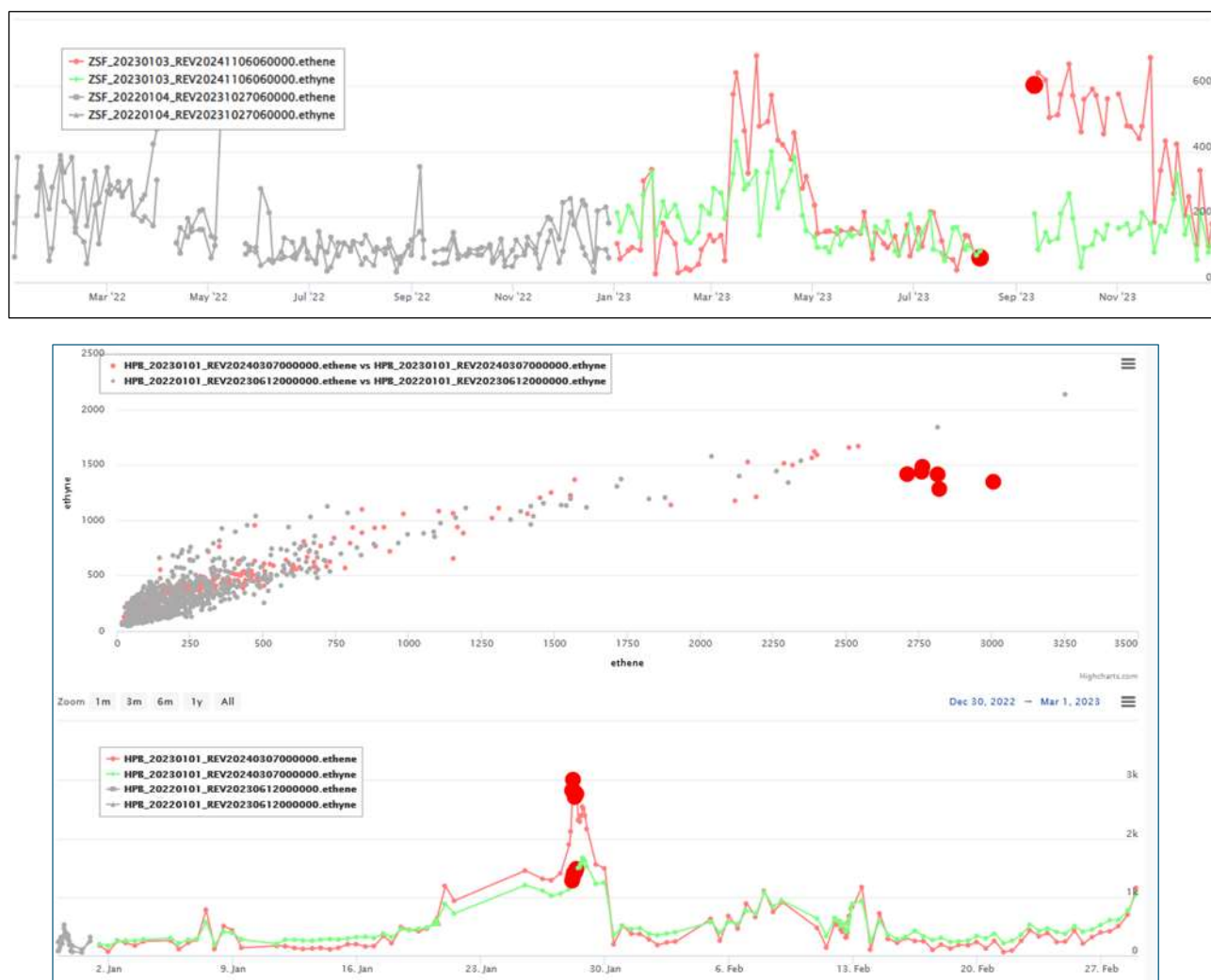


Figure 4: Example of the so-called «@VOC@ tool» used for quality checking of the submitted NMHC data within ACTRIS. This example shows data from DE0054 (Zugspitze) on top and DE0043 (Hohenpeissenberg) on the bottom as copied from the issue tracker

The EMEP VOC monitoring sites are by definition located in rural background areas, but in practice the sites are exposed to more nearby emissions to a varying extent. The data from Chilbolton observatory, located in southern England, reflect the influence of populated areas in the whole of southern England, including road traffic emissions, and show a number of short-term (hourly) spikes in the data. Data from Auchencorth Moss in Scotland, a rural location around 20 km south of Edinburgh also show very spiky hourly time series with peak levels of propane, n-butane and other species. The high concentrations at that site are mainly seen during periods with winds from the northern sector, pointing to fresh anthropogenic emissions from the urban area around Edinburgh as well as from petroleum refineries in the same direction. It turns out that for both these sites, most of the spikes are of short duration while daily average levels seem fine.

NMHC data for the sites from UBA in Germany, i.e. Waldhof, Schauinsland, Neuglobsow, Schmücke, Zingst, and Zugspitze have been subject to problems in 2023 as in previous years. This concerns alkenes with three or more C-atoms (propene and higher) but also aromatics (o-xylene) and some higher

alkanes. Dubious data have been invalidated by NILU in 2023 as in preceding years. Due to late data delivery, data from the UBA sites for 2023 could not be included in the ACTRIS QA procedure.

As in previous years, NMHC data from San Pablo (ES0001) for 2023 have been submitted to NILU but are not included in this report. The reason for this is, as discussed in previous annual reports, is that the data show levels that differ substantially from expected values indicating major problems with the sampling or chemical analyses. There is an ongoing dialogue between NILU and MMA about this issue, and a shift in technical equipment (pump, etc.) and in localisation of the sampling are represent promising changes carried out at the site.

Table 1: VOC monitoring at EMEP sites in 2023. The columns give the station names, site code, and the sampling frequencies for hydrocarbons (HC) and carbonyl compounds (Carb). The institute responsible for the chemical analyses is also given. Whether the station is part of the ACTRIS-2 project is also indicated.

Station	Code	HC ¹⁾	Institute ²⁾	Carb ¹⁾	Method	ACTRIS	Comment
Zeppelin Mtn.	NO0042	cont	NILU	-	MEDUSA	y	
Pallas	FI0096	cont	FMI	-	GC/MS	y	
Hyytiälä	FI0050	cont	UHel	-	PTR-MS	y	
Auchencorth Moss	GB0048	cont	Ricardo	-	GC/MS	y	
Chilbolton Obs.	GB1055	cont	Ricardo	-	GC/MS	y	
Waldhof	DE0002	reg	UBA	-	canisters	n	twice/week
Schauinsland	DE0003	reg	UBA	-	"	n	"
Neuglobsow	DE0007	reg	UBA	-	"	n	"
Schmücke	DE0008	reg	UBA	-	"	n	"
Zingst	DE0009	reg	UBA	-	"	n	"
Zugspitze	DE0054	reg	UBA	-	"	n	"
Hohenpeissenberg	DE0043	daily	DWD	-	GC/FID	y	twice/day
Košetice	CZ0003	reg	CHMI	-	canisters	y	twice/week
Jungfraujoch	CH0001	cont	EMPA	-	MEDUSA	y	
Beromünster	CH0053	cont	EMPA	-	GC/FID	y	
SIRTA	FR0020	cont	LSCE-CNRS-CEA		PTR-MS	y	
Mt. Cimone	IT0009	cont	UU	-	GC/MS	y	
San Pablo	ES0001	-	MMA	reg	DNPH	n	twice/week

- 1) reg = regularly (2-3 samples per week), cont = continuously
- 2) CHMI = Czech Hydrometeorological Institute
DWD = Deutscher Wetterdienst
IMT LD = Institut Mines Telecom Lille Douai
EMPA = Swiss Federal Lab. for Materials Testing and Research
FMI = Finnish Meteorological Institute
UHel = Univ. Helsinki
UBA = Umweltbundesamt (Germany)
UU = University of Urbino
MMA = Ministerio de Medio Ambiente
LSCE-CNRS-CEA = Laboratoire des Sciences du Climat et de l'Environnement
CNRS-LaMP = Laboratoire de Meteorologie Physique

3 VOC concentrations in 2023

3.1 General levels

Time series plots of the diurnal medians of all compounds at all stations during 2023 are given in the Appendix. Figure 5 shows the spread of data values for all stations and species in 2023 in box and whisker plots together with the corresponding data during the preceding 5-years period (2018-2022). The sites are arranged from north to south going from left to right in the panels. Thus, the panels in Figure 2 indicate both the north-to-south differences, the deviation of the 2023 concentration levels relative to the previous five years' climatology as well as the spread in 2023 data at each site separately.

In general, the 2023 data show similar mean levels and distribution compared to previous years with some exceptions. Data from CZ0003 (Kosetice) are examples of such exceptions, where the levels of some aromatics (ethylbenzene, m+p-xylene, o-xylene, toluene) and n-heptane show higher levels than seen in the previous 5 years' period.

For FR0020 (SIRTA) the amount of EBAS data in the years before 2023 are scattered, so the discrepancies seen when comparing 2023 to the historical data could be hampered by poor data capture. Apart from that, the levels in 2023 agree very well with the levels in the previous 5-years period in general although with a few exceptions.

PTR-MS (in 2023 applied at FI0050 and FR0020) is not capable of distinguishing isomers and thus only the concentration of the sum of the isomers with the same molecular weight is measured. In the plots in this report, these groups are named 'mass_<WT>_organic_compounds' where WT is equal to the molecular mass + 1 (since the PTR-MS adds a hydrogen atom with mass 1 to the species). Thus, in the following the species applies to the plots and names listed in Table 2.

Table 2: Name of species measured by PTR-MS (see text for explanation)

Name (MW+1)	Species
59	propanone, propanal
69	isoprene, cyclopentene, furan
71	3-buten-2-one (MVK), 2-methylpropenal (MACR), 2-butenal
73	methylglyoxal, acrylic acid, butanal, 2-methylpropanal, 2-butanone (MEK), tetrahydrofuran
107	benzaldehyde, o-xylene, m-xylene, p-xylene, ethylbenzene, C ₈ -alkylbenzenes

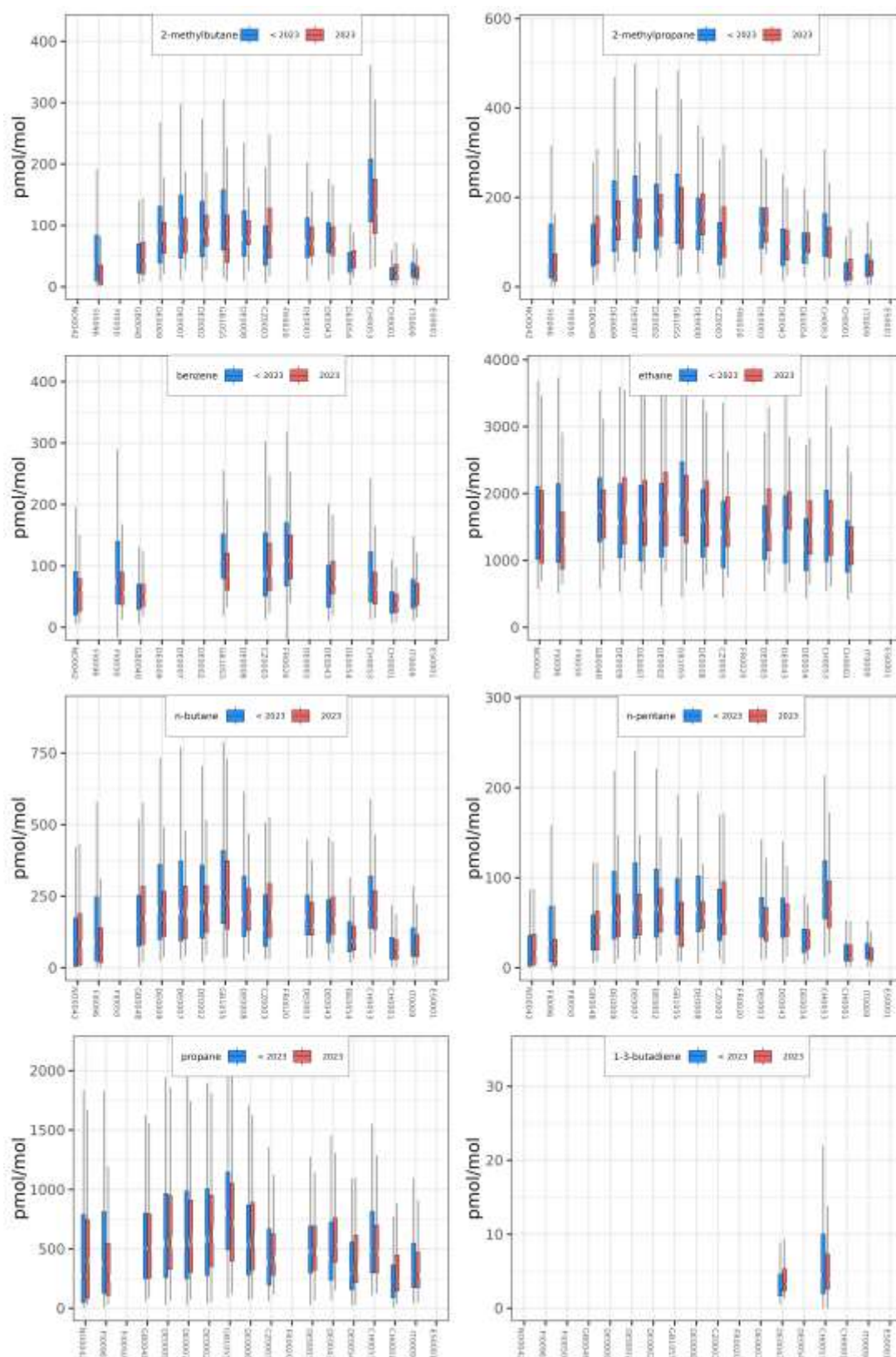
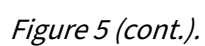


Figure 5: Box- and whisker-diagrams for VOCs based on daily median levels in 2023 (red) compared to the levels during 2018-2022 (blue). The boxes enclose the 25- and 75-percentile with the median marked inside.



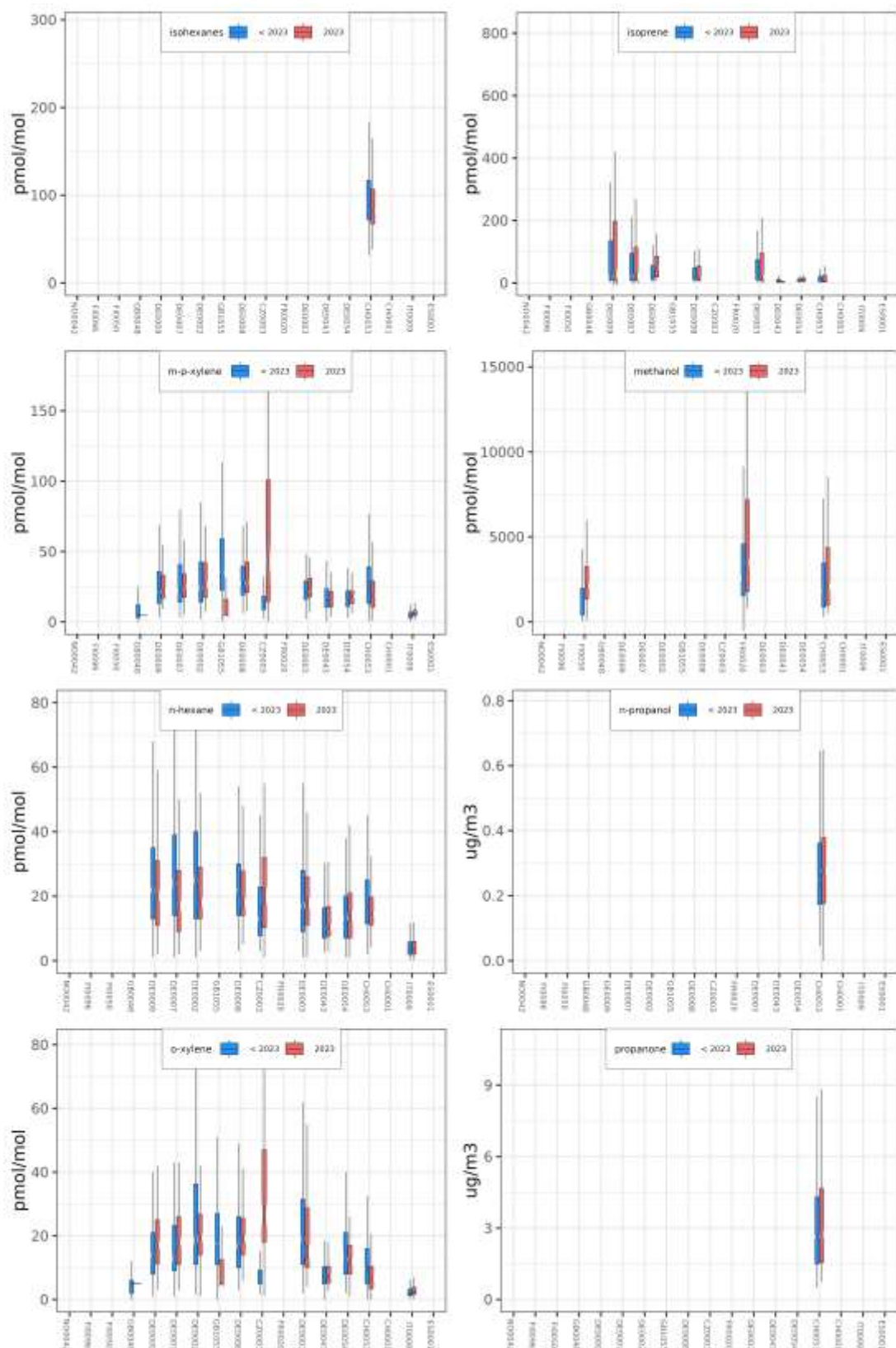
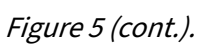


Figure 5 (cont.).



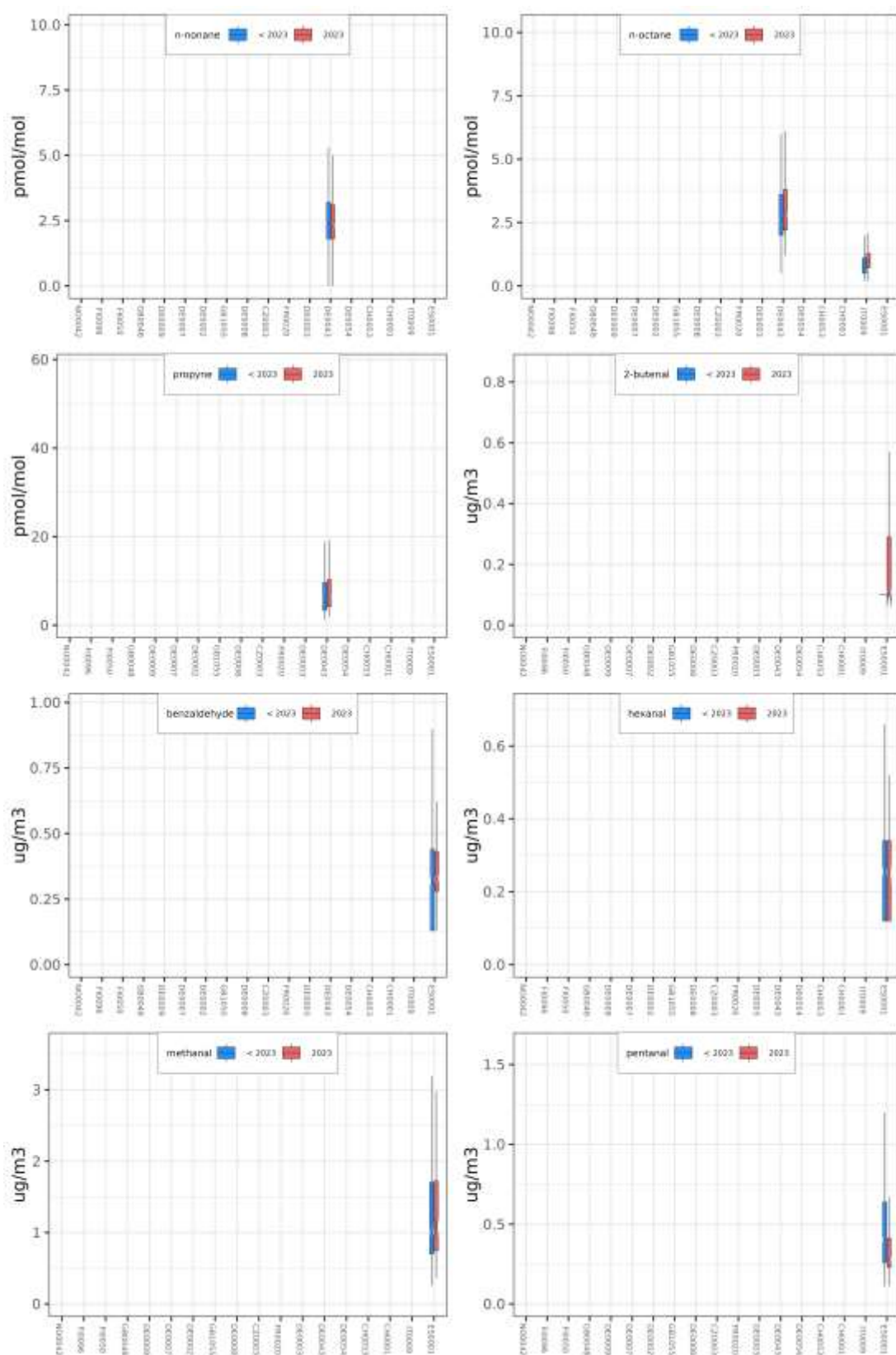


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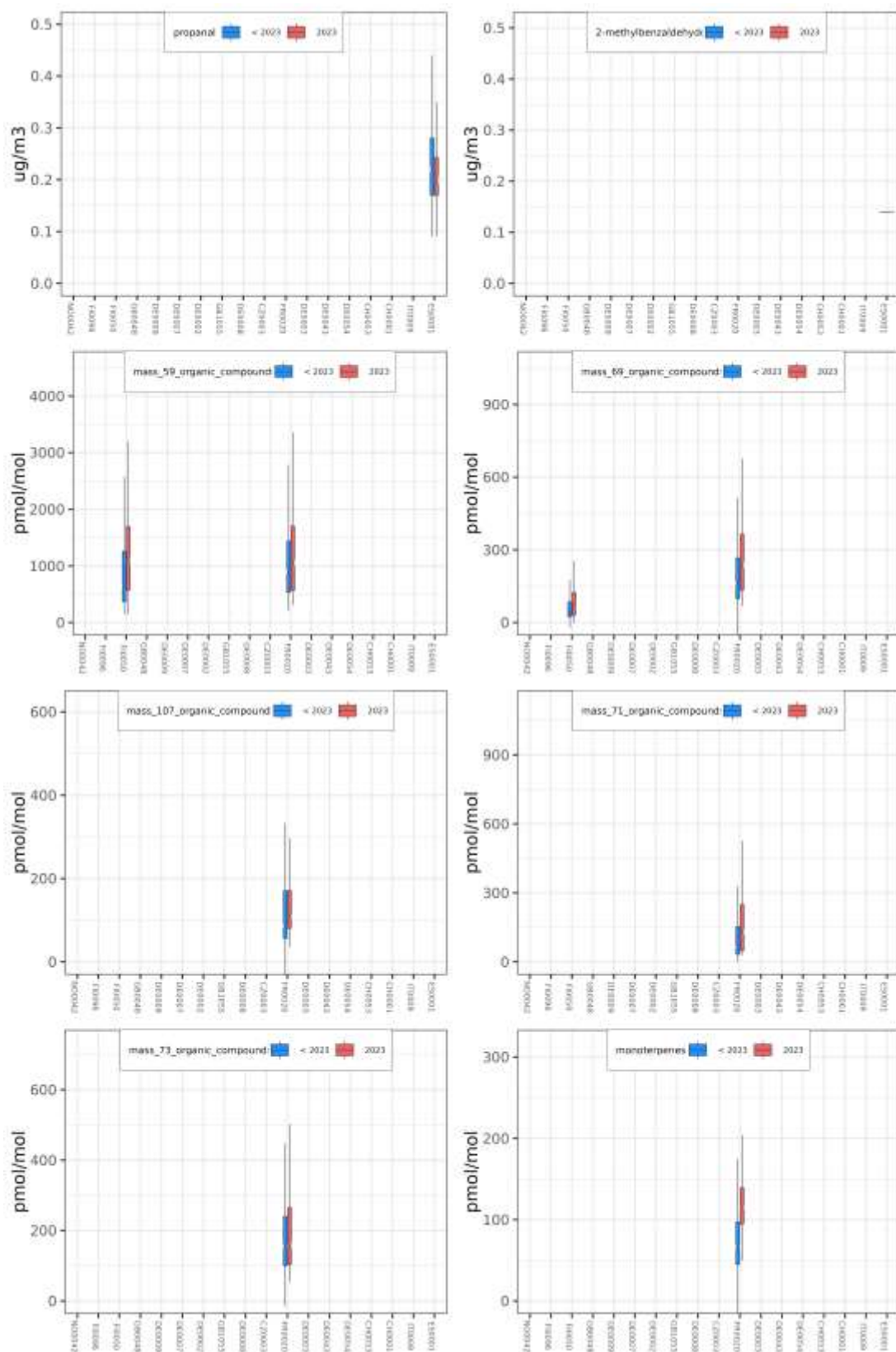


Figure 5 (cont.).

3.2 Regional distribution of VOC

Figure 6 shows maps with the stations' annual median concentrations of VOCs in 2023. Note that since the steel canisters are all sampled at daytime (normally at noon), a bias could be inherent in these plots when compared with the medians of the 24 h based data from online GCs. A bias for other species is also likely to be seen to a varying extent. Some of the mountain stations (Zugspitze, Hohenpeissenberg and Mt Cimone) are influenced by diurnal venting of the planetary boundary layer and will receive upslope polluted air masses during daytime when the vertical mixing is sufficiently strong and see cleaner free tropospheric air masses at night. The station at Jungfraujoch (3578 m asl) will on the other hand most of the time be located in the free troposphere, above the planetary boundary layer.

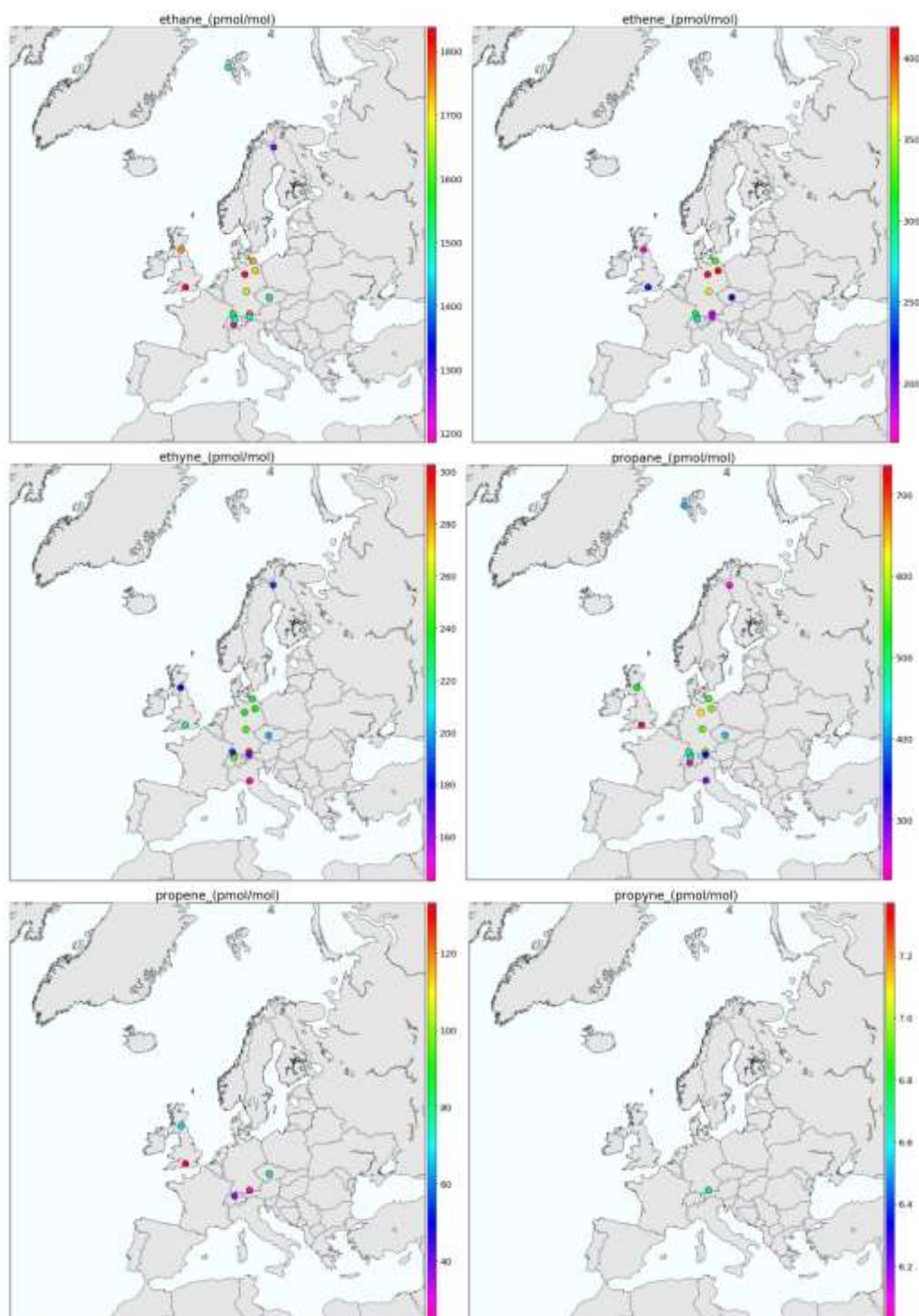


Figure 6: Annual median concentrations of VOCs in 2023.

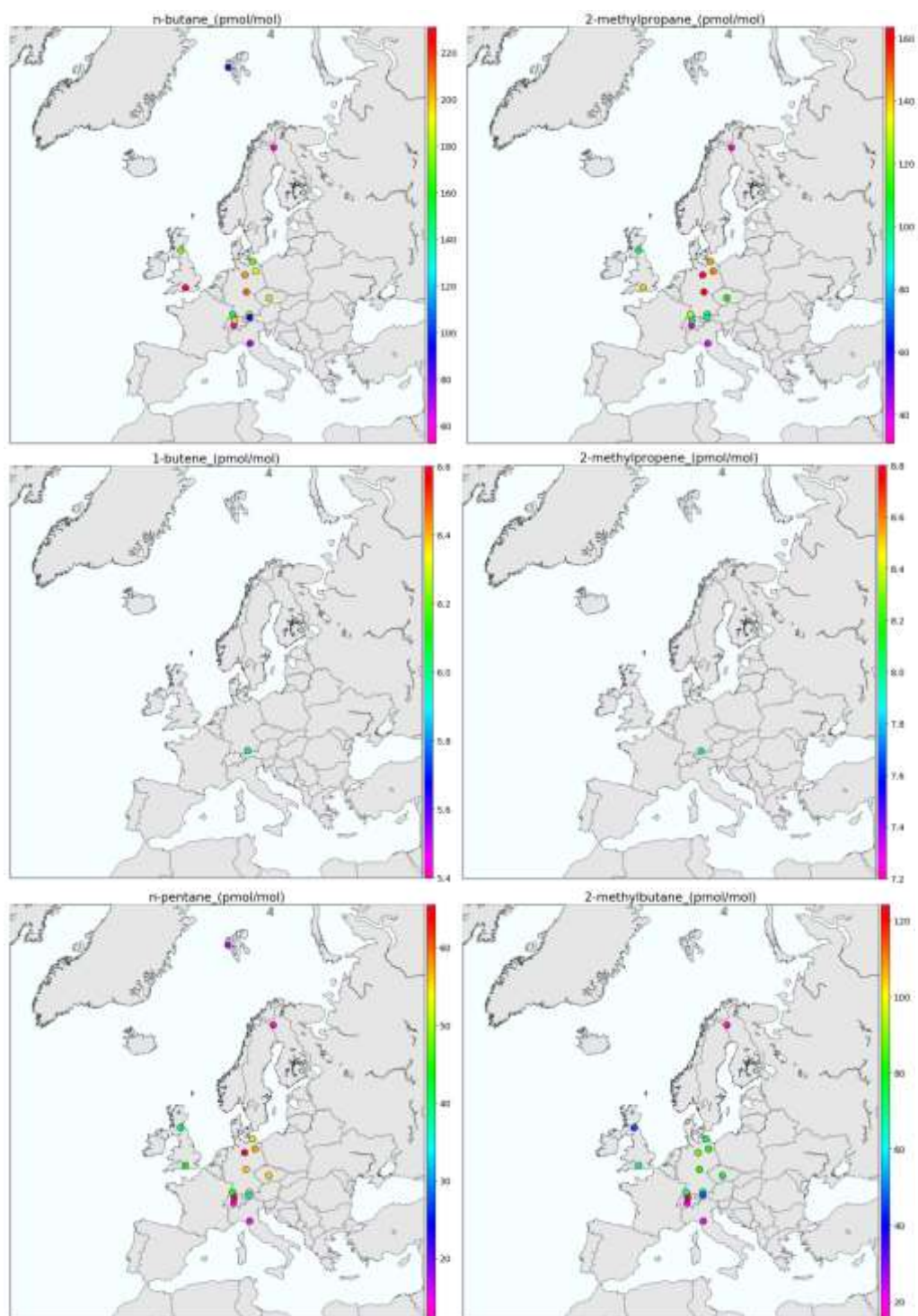


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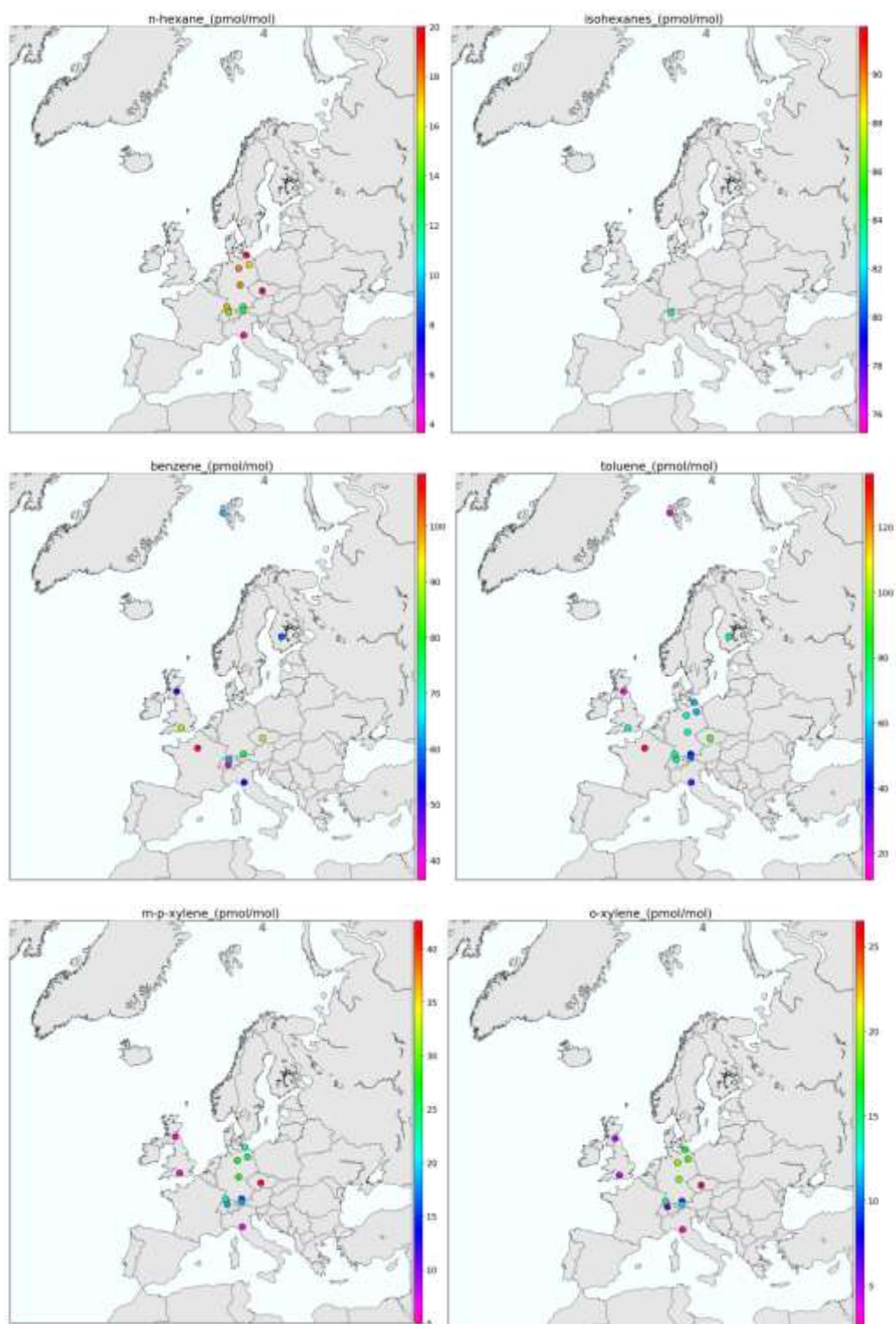


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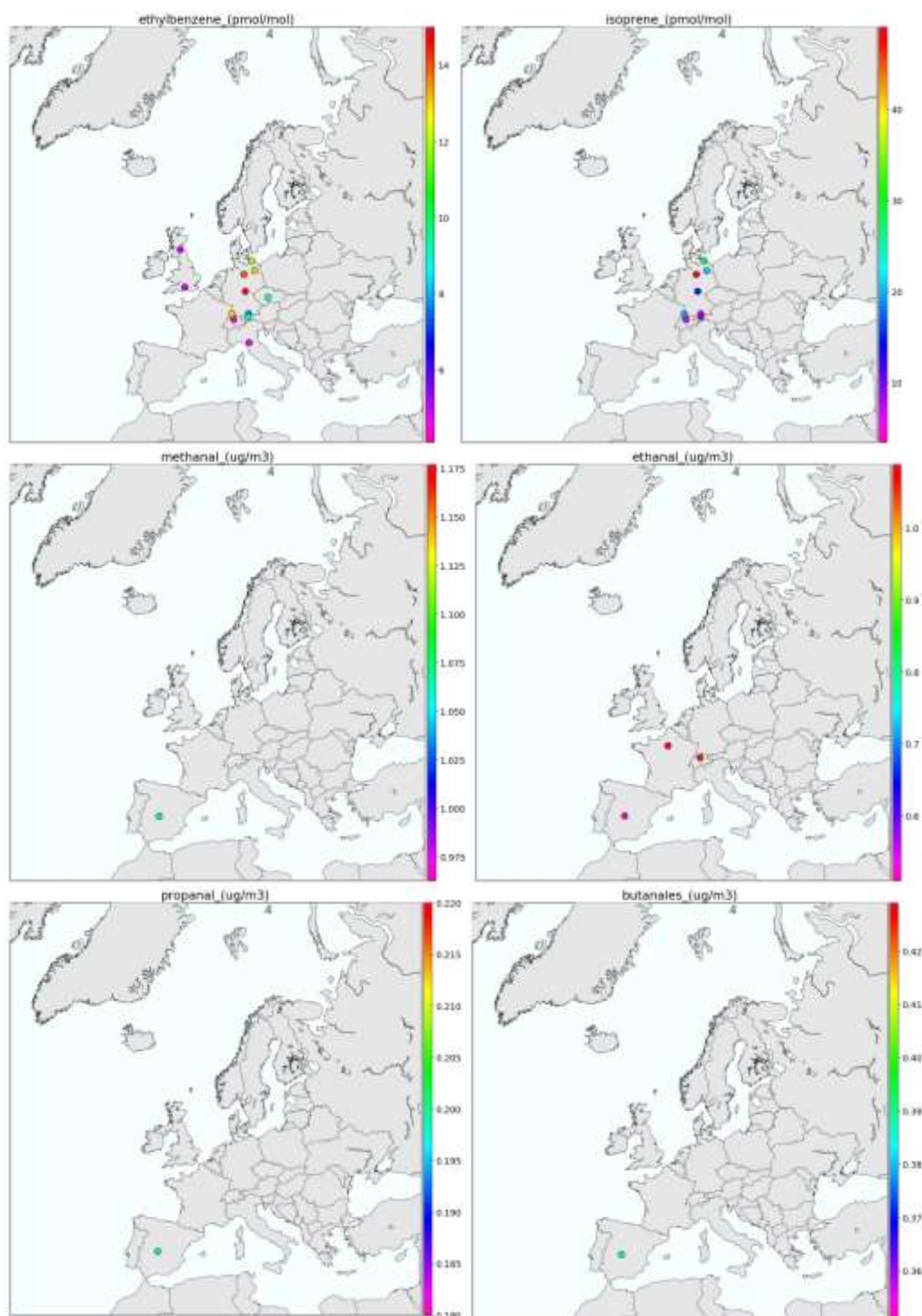


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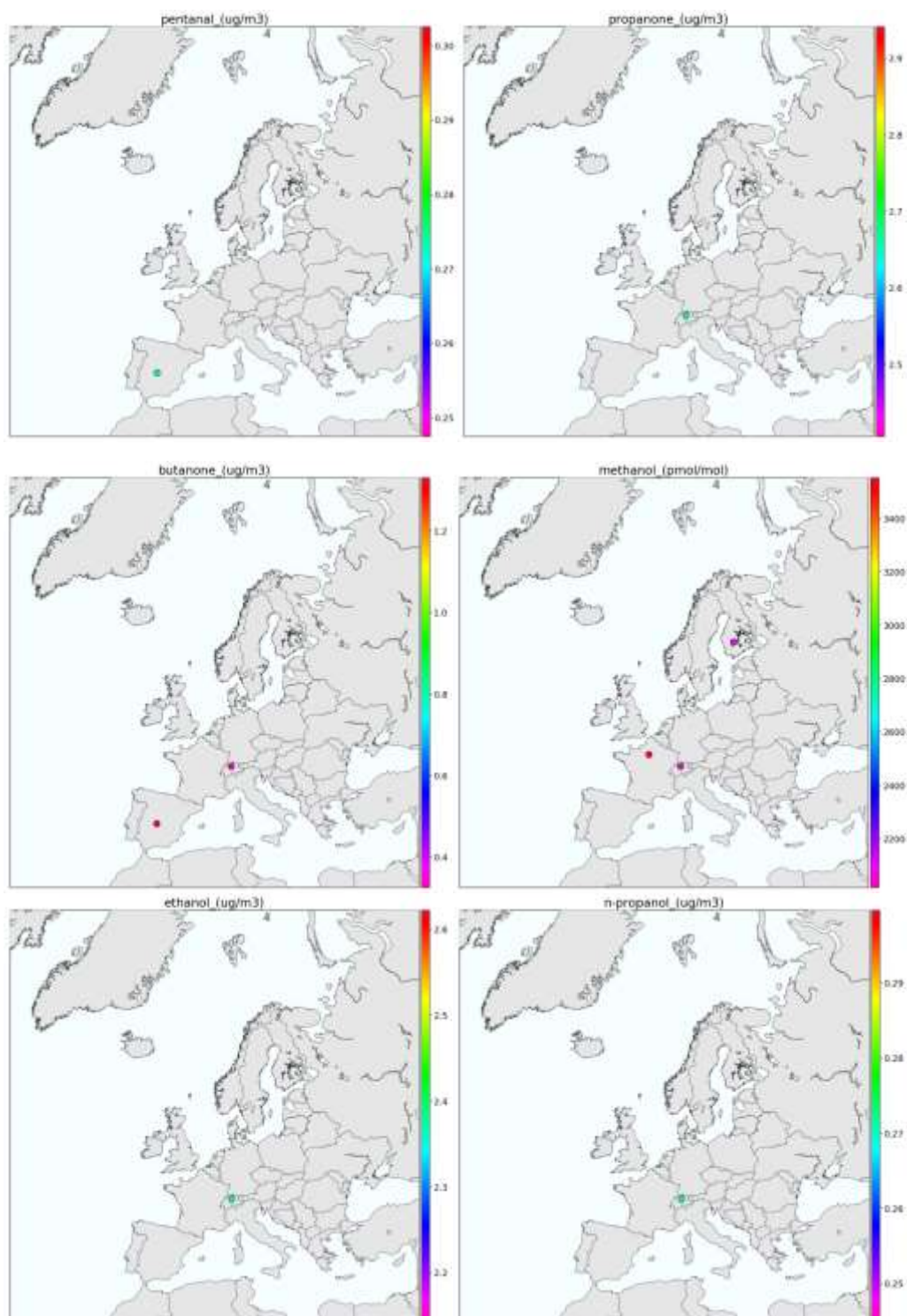


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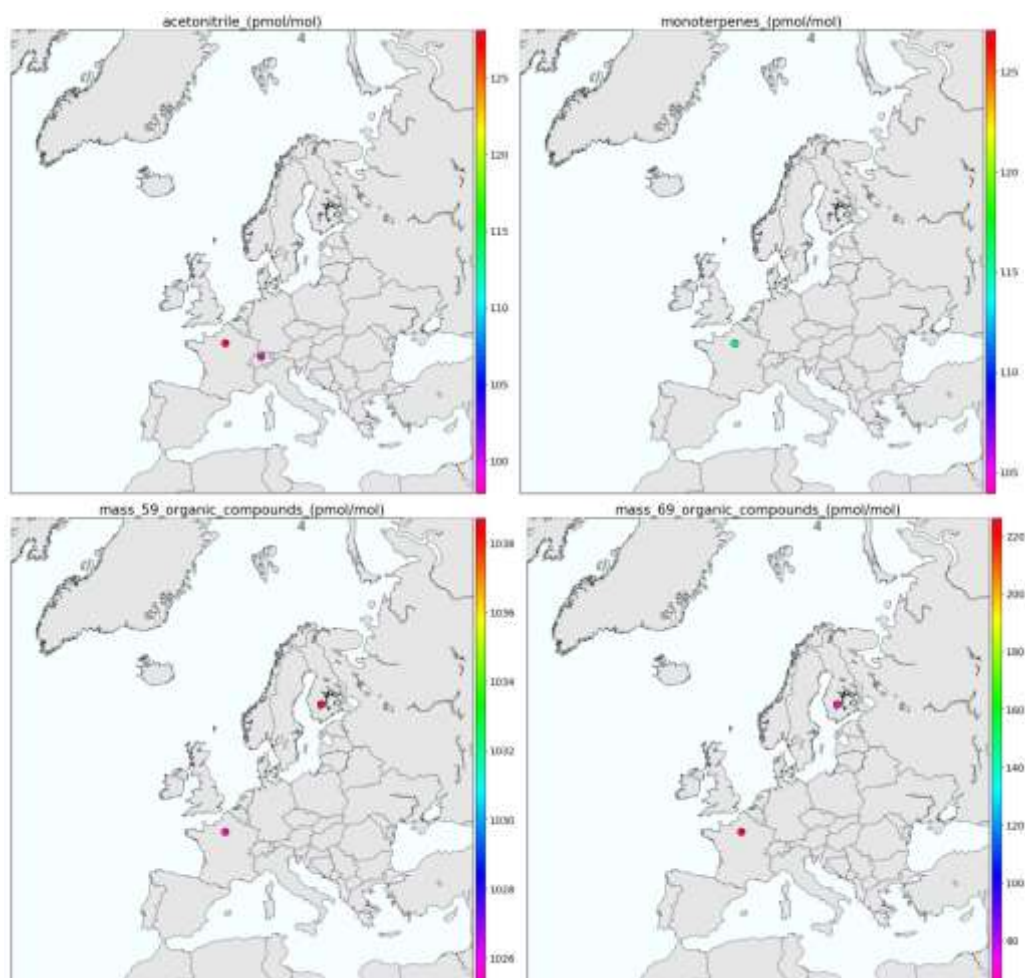


Figure 6 (cont.).

4 Long-term trends in VOC

According to the official emission data, there have been marked reductions in anthropogenic emissions of VOCs during the last decades in Europe. Overview tables with reported emission trends for individual countries have been published on the CEIP website at <https://www.ceip.at/>. Detailed information on the sectoral level can also be accessed in WebDab.

There are substantial differences in the emission trends between countries and regions. For the area defined as “EMEP-West”, there has been an overall reduction in VOC emissions of more than 40 % for the period 2000-2020 (Fagerli et al., 2022) and for individual countries such as Germany, France, UK, Italy, Spain, and Poland, the reductions have been 43%, 54%, 56%, 46%, 38% and 17%, respectively, as given in the same report.

For the area defined as “EMEP-East”, however, the emission data including so-called gap-filling indicate a nearly flat development from 2000 to 2020 for NMVOC. As stated in last year’s EMEP Status report (Fagerli et al., 2022), the emission estimates for EMEP-East are, however, much more uncertain than the data for EMEP-West.

Declines in the measured concentrations of hydrocarbons have been reported from suburban/urban sites at several locations. Based on a network of high-frequency continuous monitoring of C₂-C₈ hydrocarbons in the UK, mostly at urban/suburban locations, Derwent et al. (2014) found substantial declines in concentrations with recent levels close to an order of magnitude below the levels in the early 1990s. They estimated exponential declines in concentrations of the order of -11% y⁻¹ to -22% y⁻¹ for the period 1994-2012. They also found a marked difference between ethane and propane on one side which showed relatively stable levels, while other alkanes showed pronounced declines.

Long-term monitoring data from an urban network in Switzerland (Hüglin, pers. comm.) also show strong declines in the concentrations of NMHC and OVOC from the start of the 1990s to the present.

Various trend studies have been carried out for VOC-data from EMEP rural sites as well. Sauvage et al. (2009) and Waked et al. (2016) found clear decreases at the French EMEP sites of most NMHCs. Ethane was an exception to this and showed more stable levels.

Analyses of the twenty-year NMHC monitoring at the EMEP/GAW site Pallas in Northern Finland revealed a significant downward trend only for ethyne (Hellen et al., 2015). They concluded that other source regions than the EU were dominating the NMHC levels at the site. Based on source area estimates, they found that the Eastern parts of the continent were the main source regions for high concentrations at Pallas.

A simple 1:1 relationship between observed VOC concentrations at rural background sites and the overall European emission numbers is not to be expected. Interannual variations in atmospheric transport patterns, vertical mixing, photochemical oxidation as well as spatial differences in emission reductions complicate the analyses.

Furthermore, various procedures for trend analyses in terms of mathematical methods, selection of time periods and stations etc. could give different results. In previous VOC annual reports (Solberg et al., 2022) daily measurements of selected VOCs at five EMEP sites were analyzed with two separate statistical methods: The method used for trace gases in the AGAGE project (Simmonds et al., 2006) as well as best-

fit seasonal trend curves calculated by non-linear least squares fit using a standard statistical package (Markwardt, 2009). In addition to these two methods the Mann-Kendall/Theil-Sen's slope methodology (MK) was applied to the annual median concentrations (Bronaugh and Werner, 2019; Sen, 1968).

In the present report a GAM (generalized additive model) has been used based on the AirGAM model (Walker et al., 2023). This is an air quality trend and prediction model developed at NILU in cooperation with the European Environment Agency (EEA). AirGAM is based on nonlinear regression and is capable of estimating trends in daily measured pollutant concentrations at air quality monitoring stations, discounting for the effects of trends and time variations in corresponding meteorological data.

AirGAM has been applied to daily levels of hydrocarbons measured at Hohenpeissenberg (DE0043) during the period 2001-2022 together with daily values of temperature, humidity, wind speed, wind direction, and mixing height as compiled from ECMWF met data in the same way as described in Walker et al. (2023). The same model set-up as used for NO₂ (Solberg et al., 2021) was used for these hydrocarbons, implying that daily mean values of all the meteorological covariates were used in the regression and furthermore, that a log-based approach was assumed, i.e. the logarithm of the daily hydrocarbon levels was used as the dependent variable.

The trend during 2001-2023 in the relative concentration levels of the NMHCs with 2001 being the reference year is shown in Figure 7 while Figure 8 shows the daily mean levels in 2023 as observed and predicted with the AirGAM model. Please note that whereas the measurements are done twice a day (noon and midnight) the meteorological data input to AirGAM has been based on the daily averages of the values at 00, 06, 12 and 18 UTC.

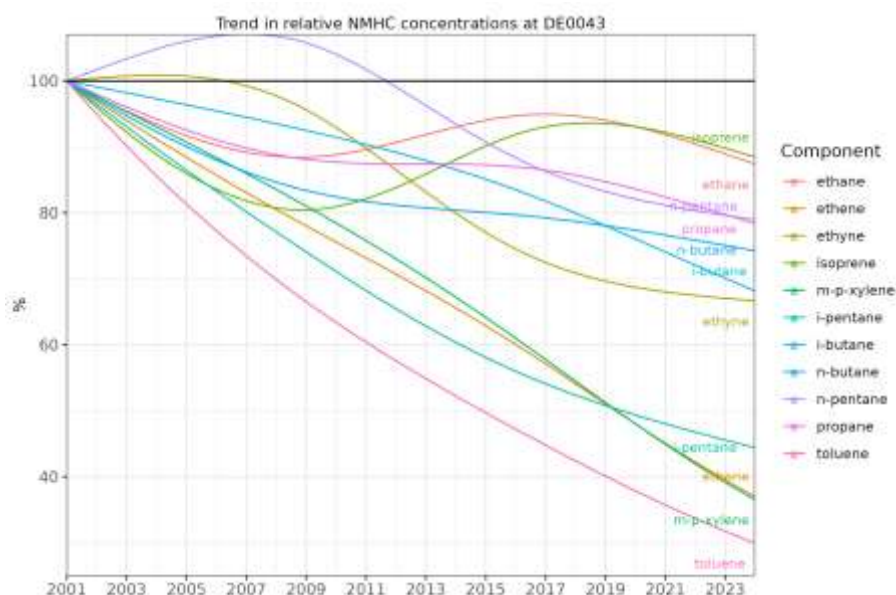


Figure 7: Meteorological adjusted trend in relative concentrations of NMHCs at Hohenpeissenberg 2001-2023 as calculated by the AirGAM model (Walker et al., 2023).

The results shown in Figure 7 indicate a substantial reduction in the NMHCs measured at DE0043 and, furthermore, significant differences in the trends for the various species. Strongest declines are found for toluene, ethene, m-p-xylene and i-pentane which in 2023 are seen at levels only 35-45% of that in 2001 meaning a drop of the order of 55-65 % since 2001. The other NMHC have dropped to around 70-90% of the 2001 levels meaning a reduction of 10-30% during this 22-year period.

These marked reductions in observed NMHC levels in Europe since 2001 are in line with the emission data from the EMEP-West region as mentioned above. A quantitative comparison species by species could not be done since the EMEP emission data are given for NMHC as a whole and not for individual substances.

The reliability of the AirGAM model could be judged from Figure 8 showing the agreement between the observed and modelled daily mean values during 2023 for nine anthropogenic NMHCs from ethane to toluene plus isoprene including some statistical metrics for the data. Unfortunately, there was a long period of missing observations in the summer season (June-September) at Hohenpeissenberg in 2023.

It is important to keep in mind that AirGAM is a statistical model (or tool) that is based purely on observed relationships between air quality data and meteorological parameters without any parameterization of physical or chemical processes. This makes it well suited for analysing historical data while the model could of course not be used for predicting the effects of emission changes. The results in Figure 8 show good agreements between observed and predicted daily values in 2023 although some spikes in the observed data are not reproduced by AirGAM.

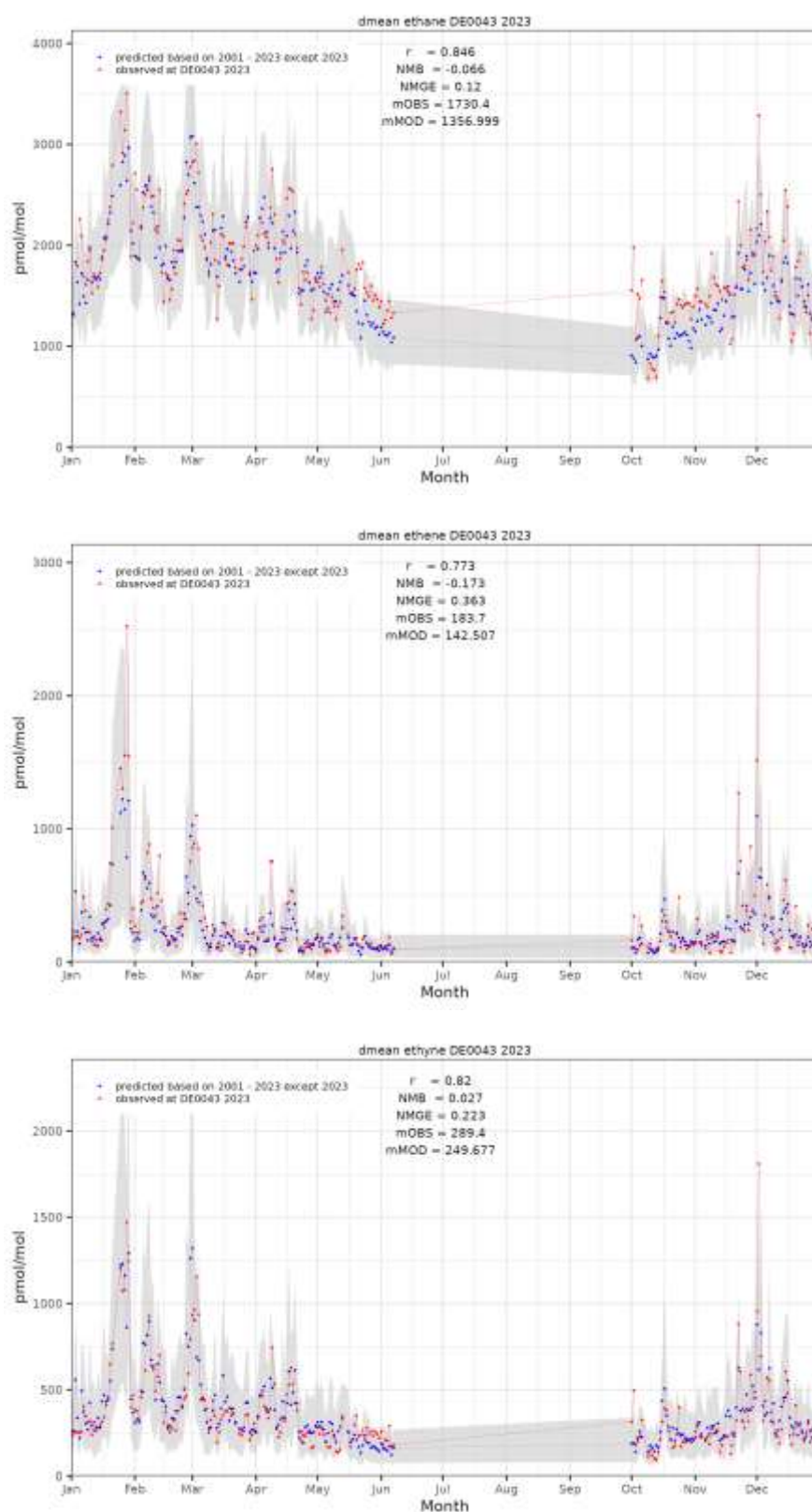


Figure 8: Daily mean concentrations of selected NMHCs as observed (red) and modelled by AirGAM (blue) at Hohenpeissenberg in 2023. Statistical metrics as the correlation coefficient (r), normalized mean bias (NMB), normalized mean gross error (NMGE), mean observed (mOBS) and mean modelled (mMOD) level are given in the panels.

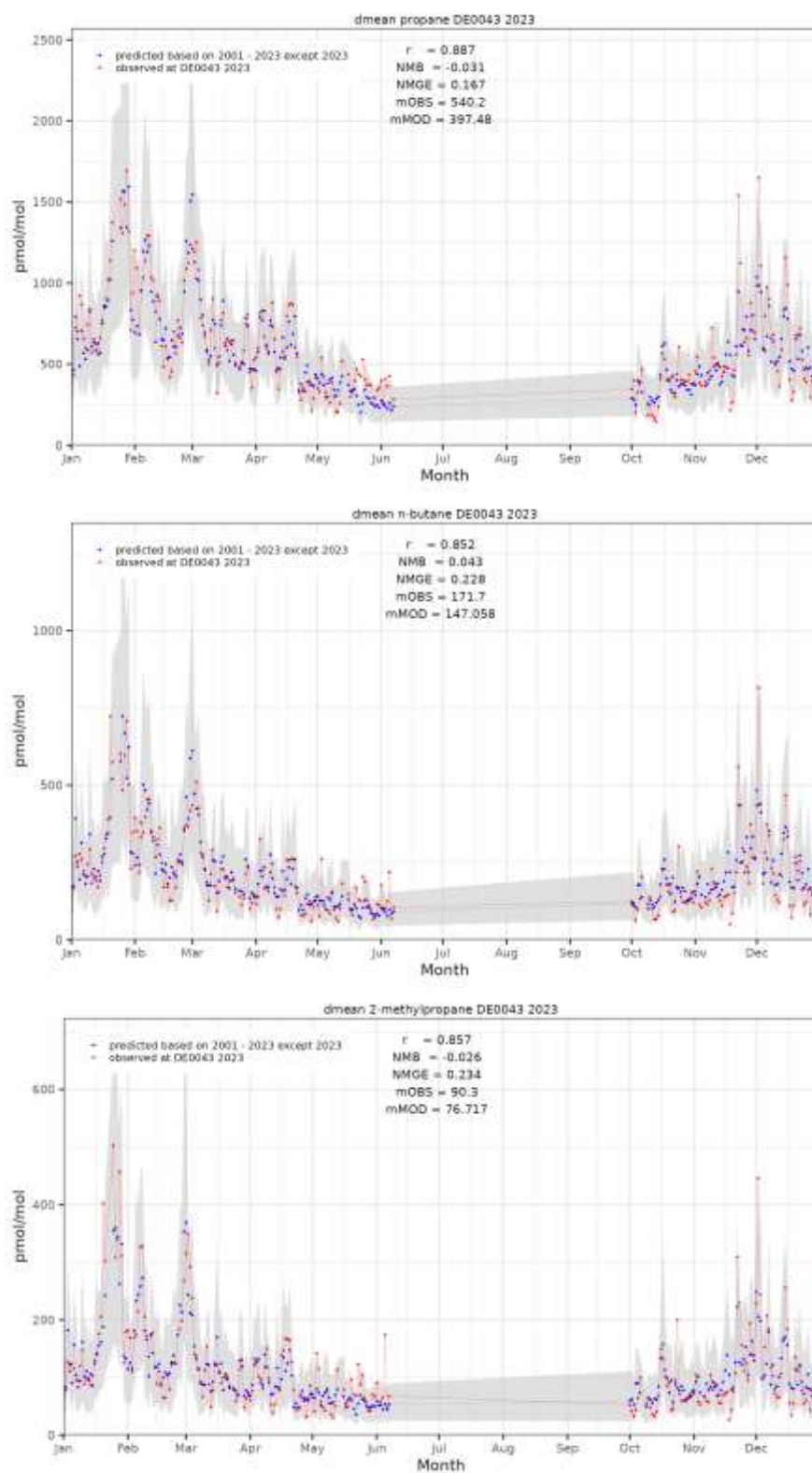


Figure 8 (cont.).

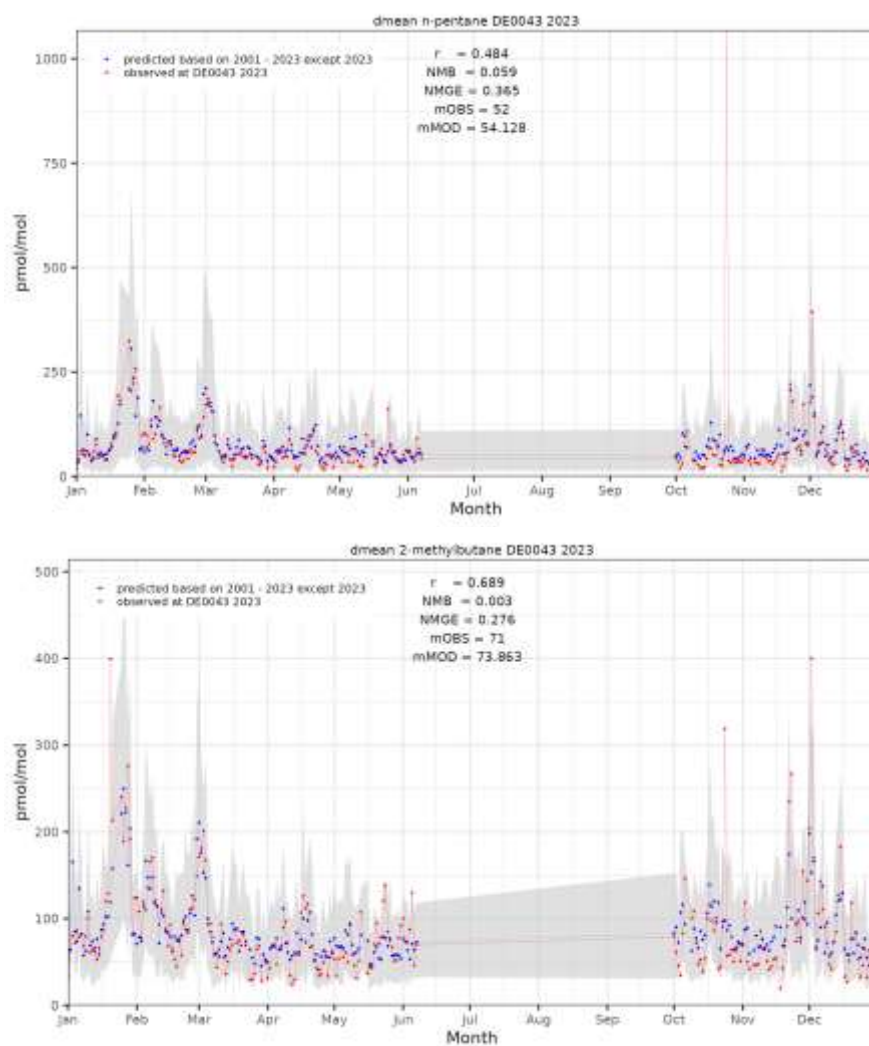


Figure 8 (cont.).

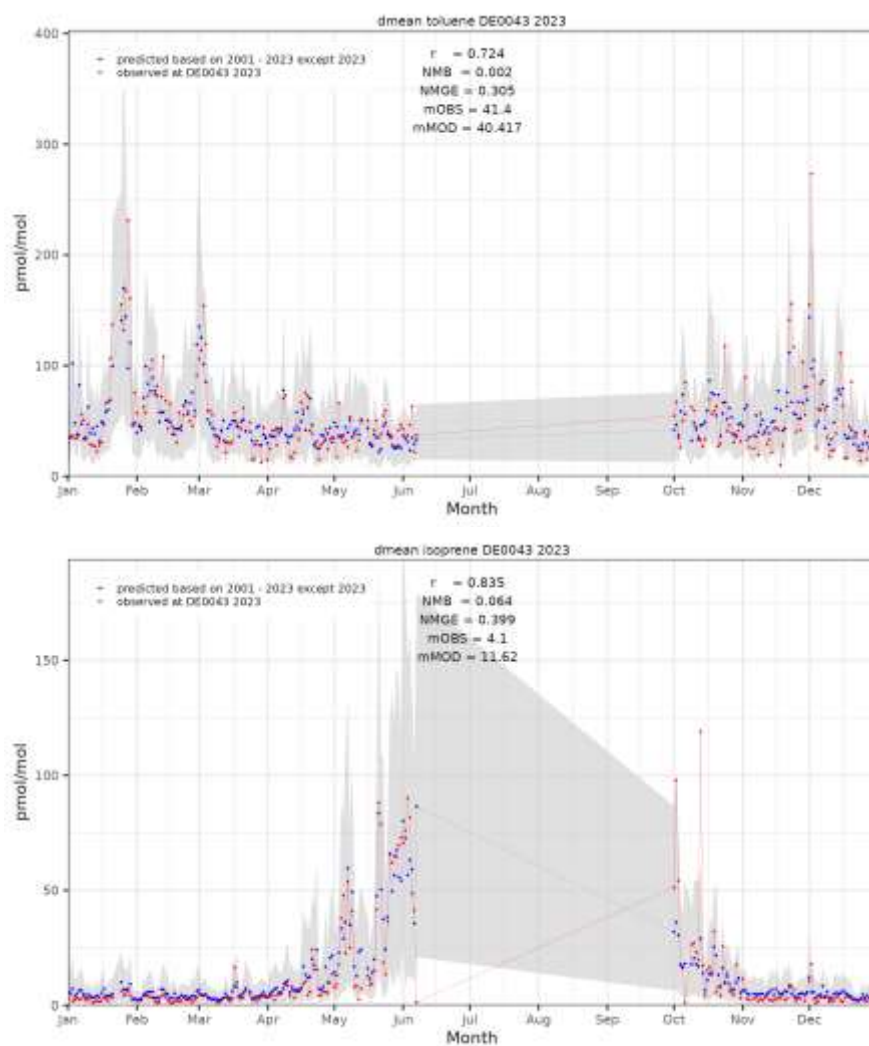


Figure 8 (cont.).

5 Acknowledgement

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6 References

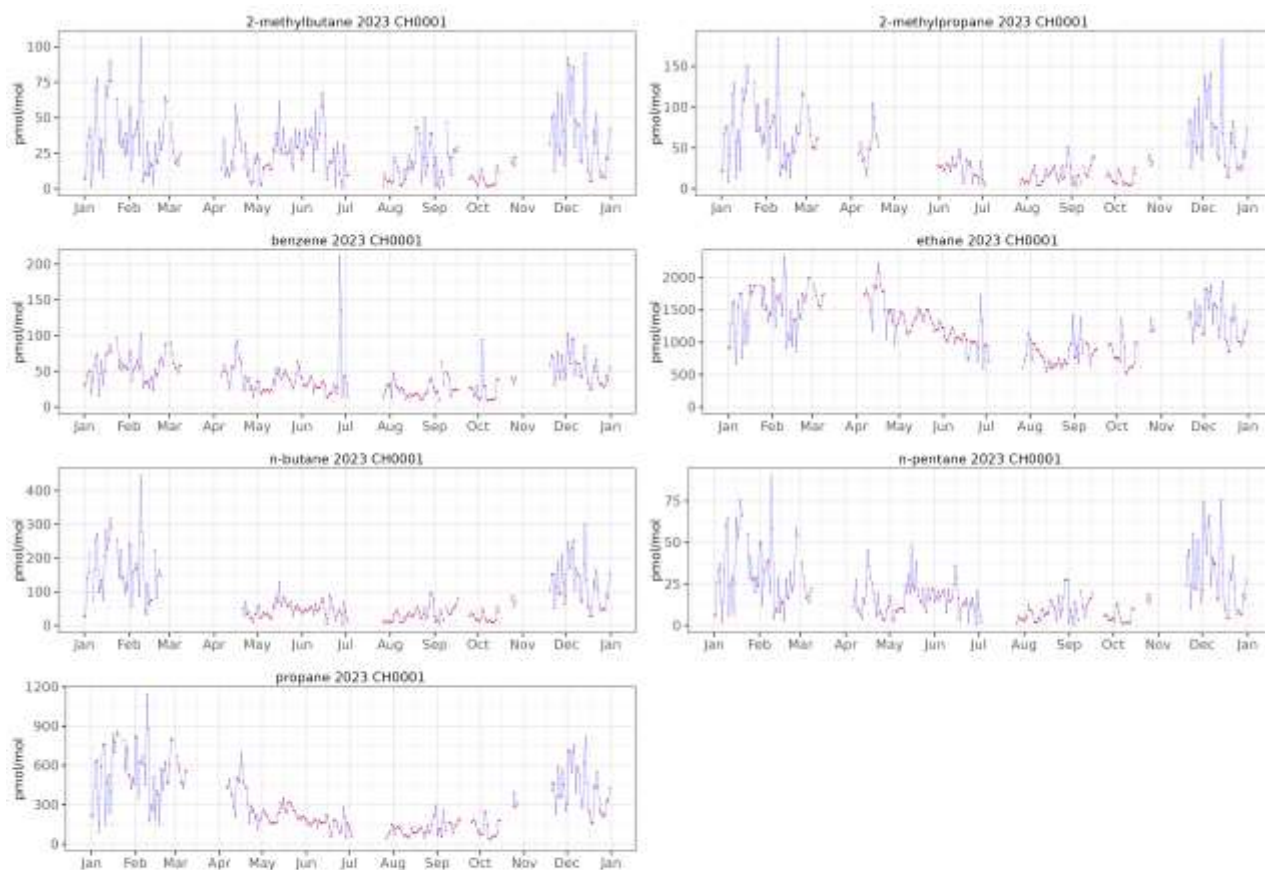
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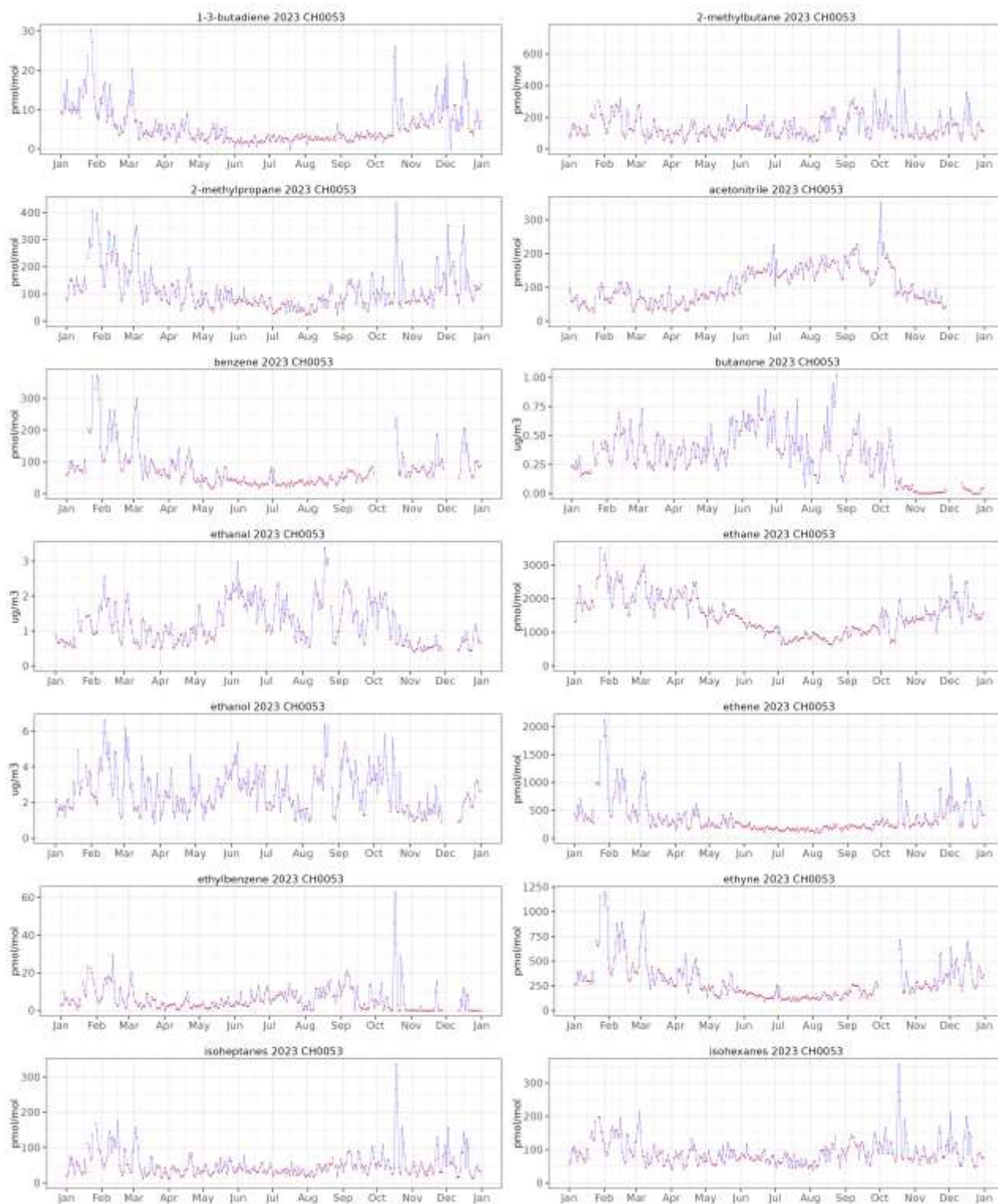
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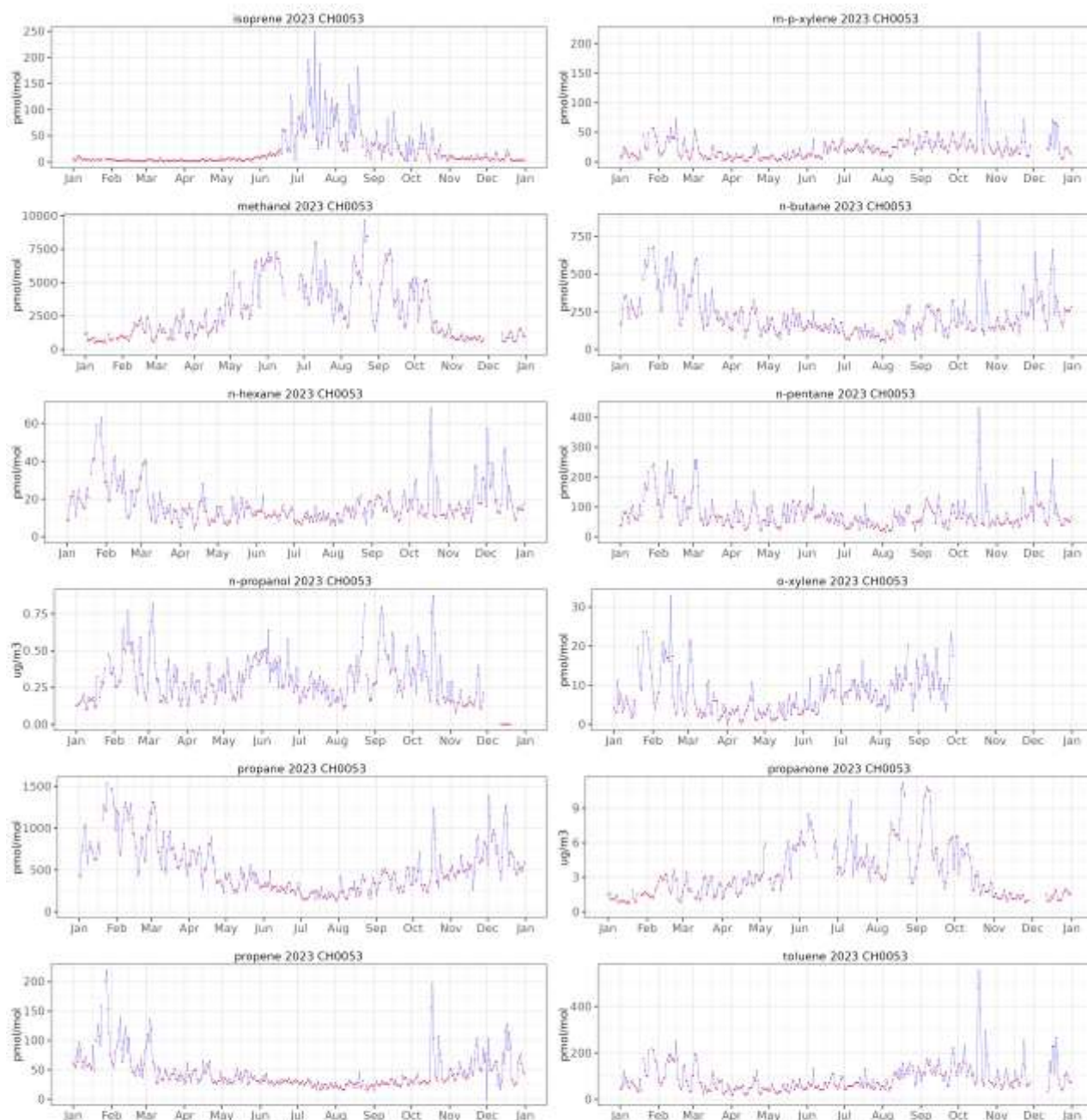
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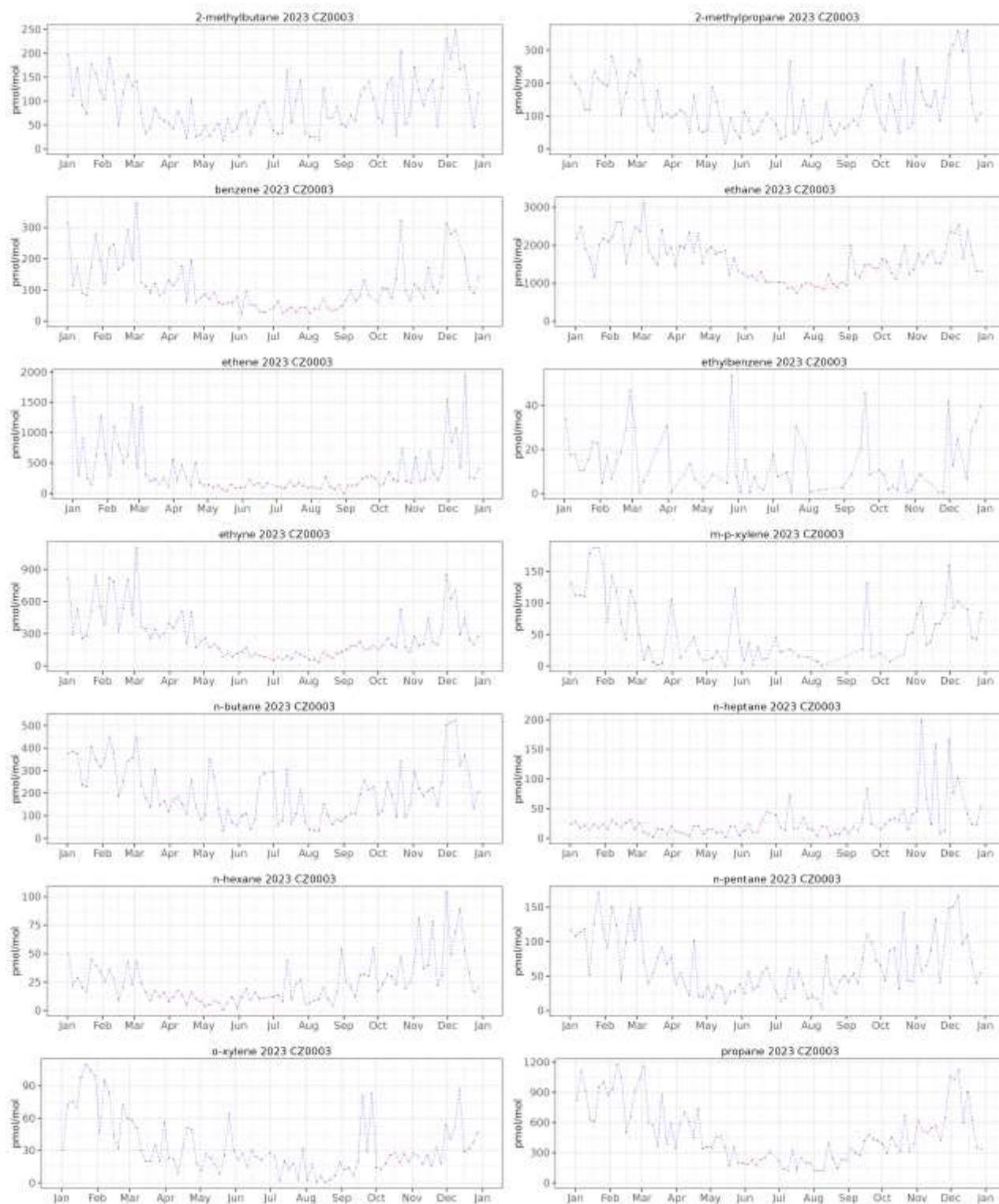
Appendix 1

Time series of daily medians of VOCs measured in 2023

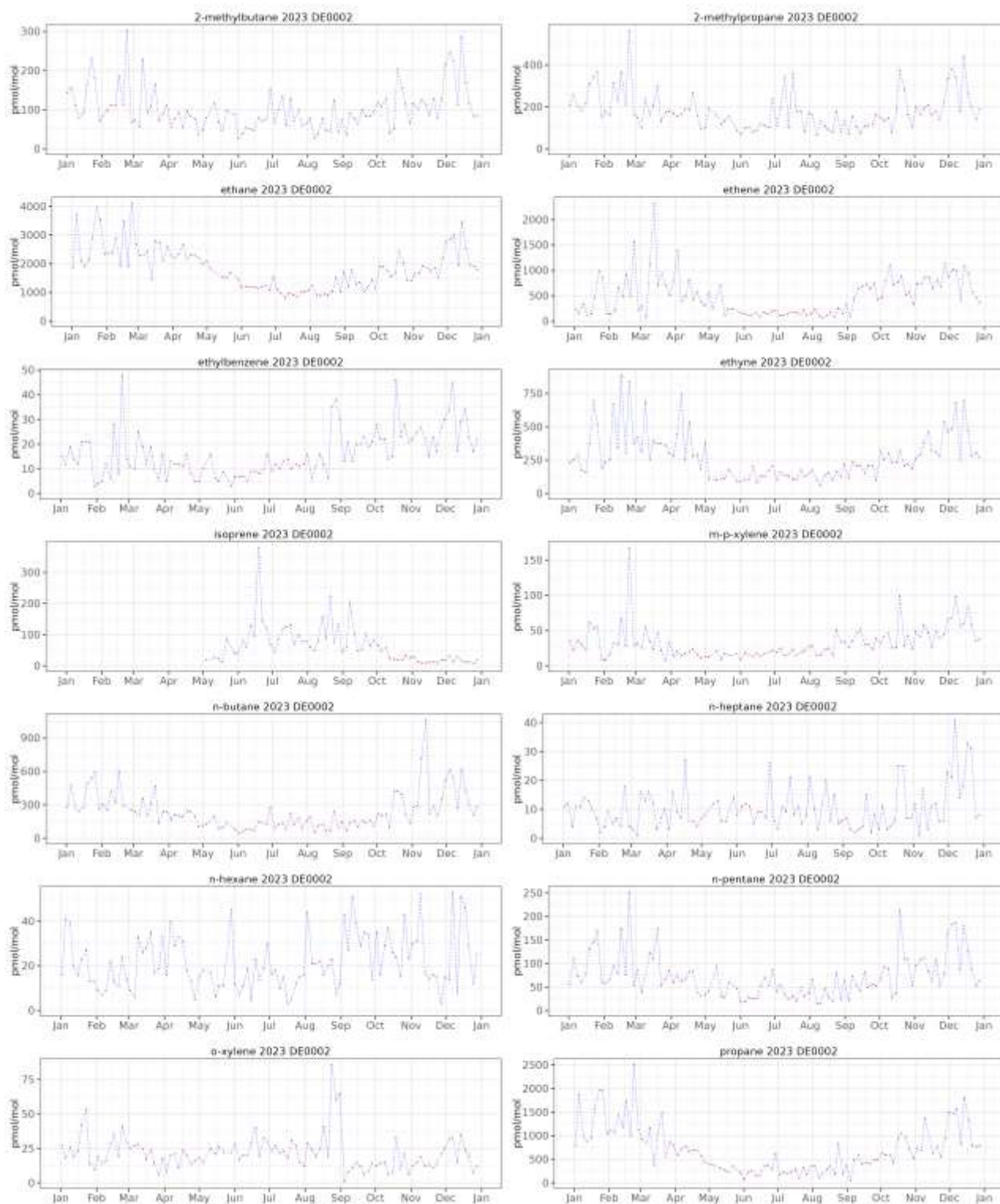




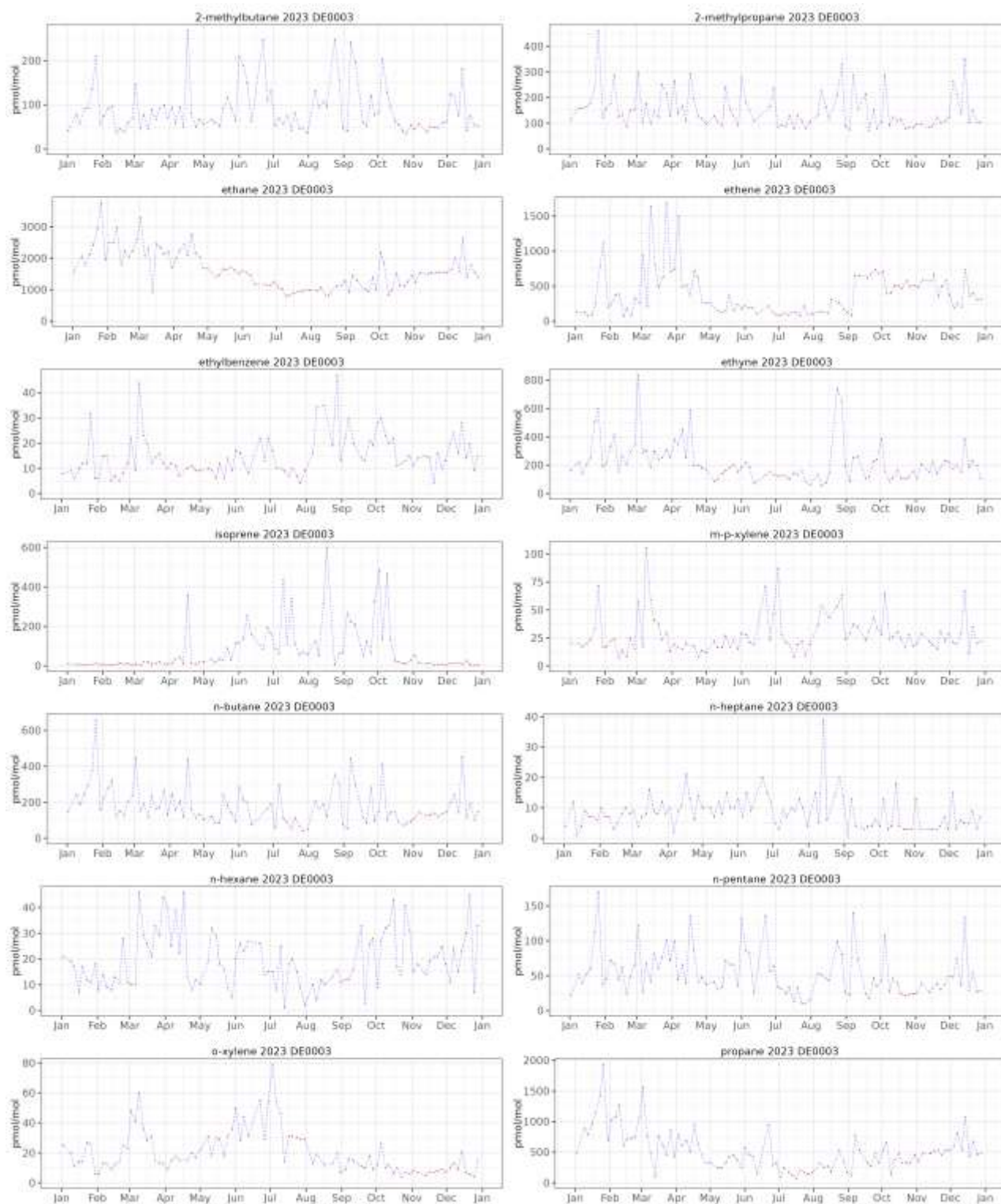




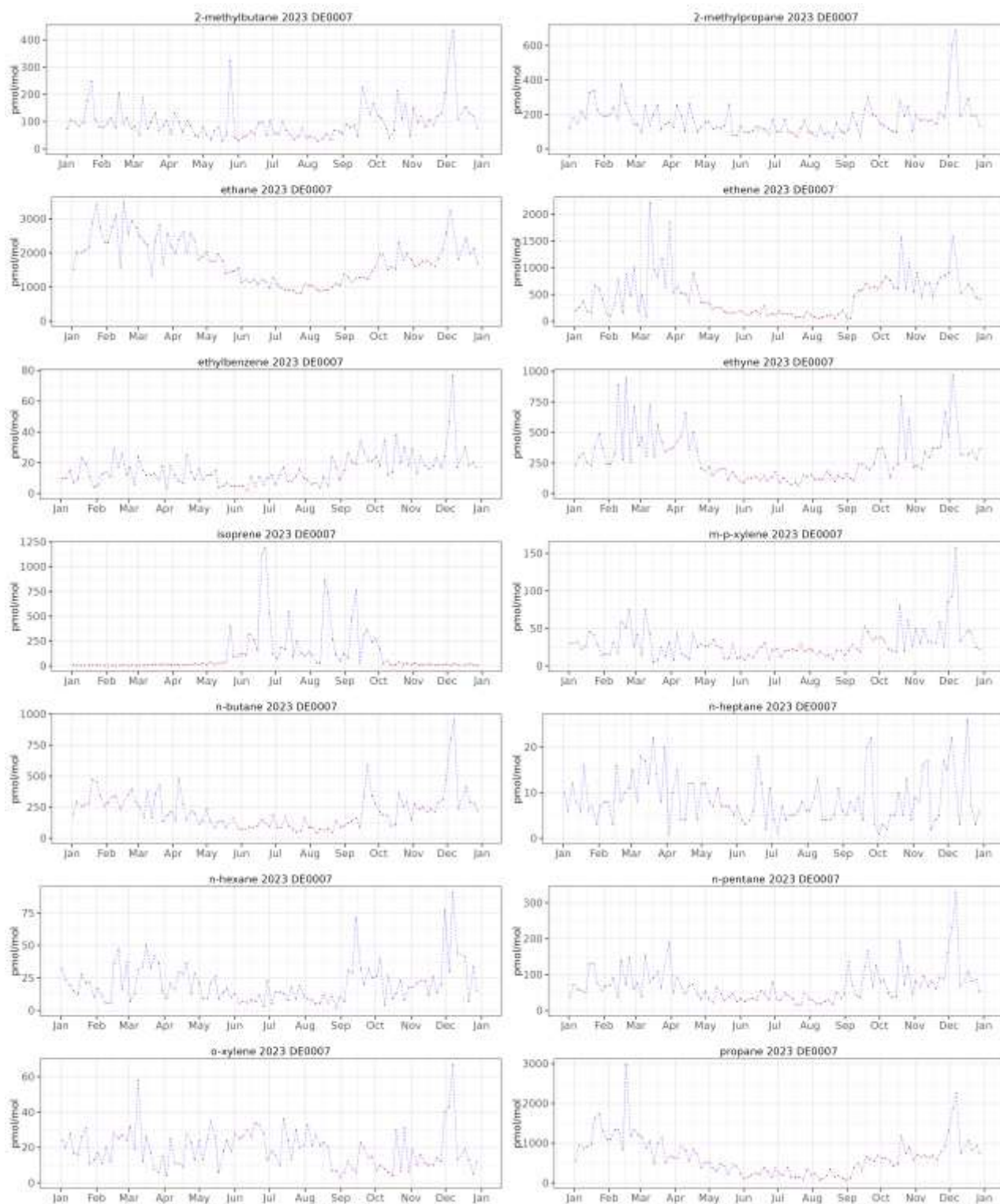




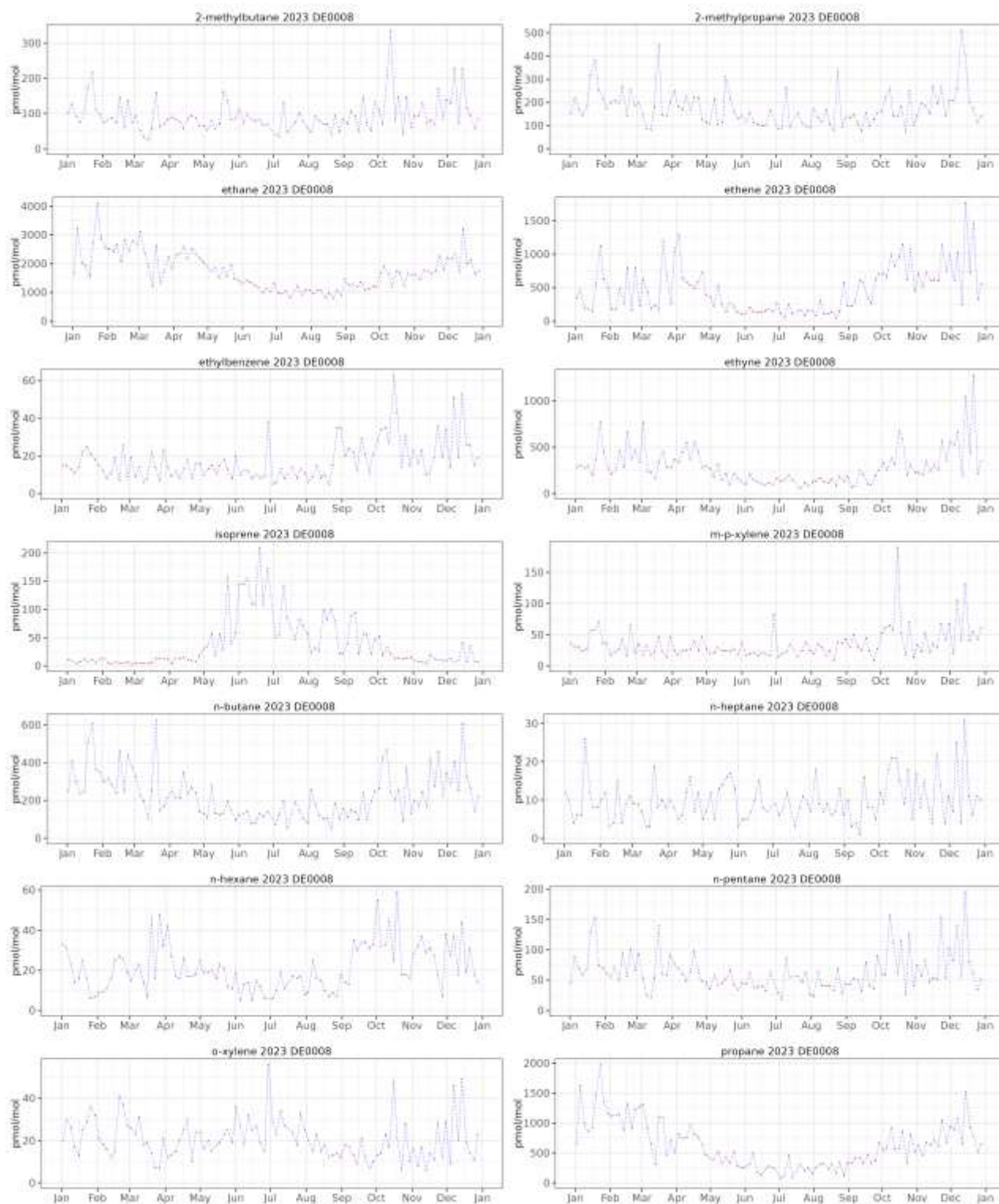




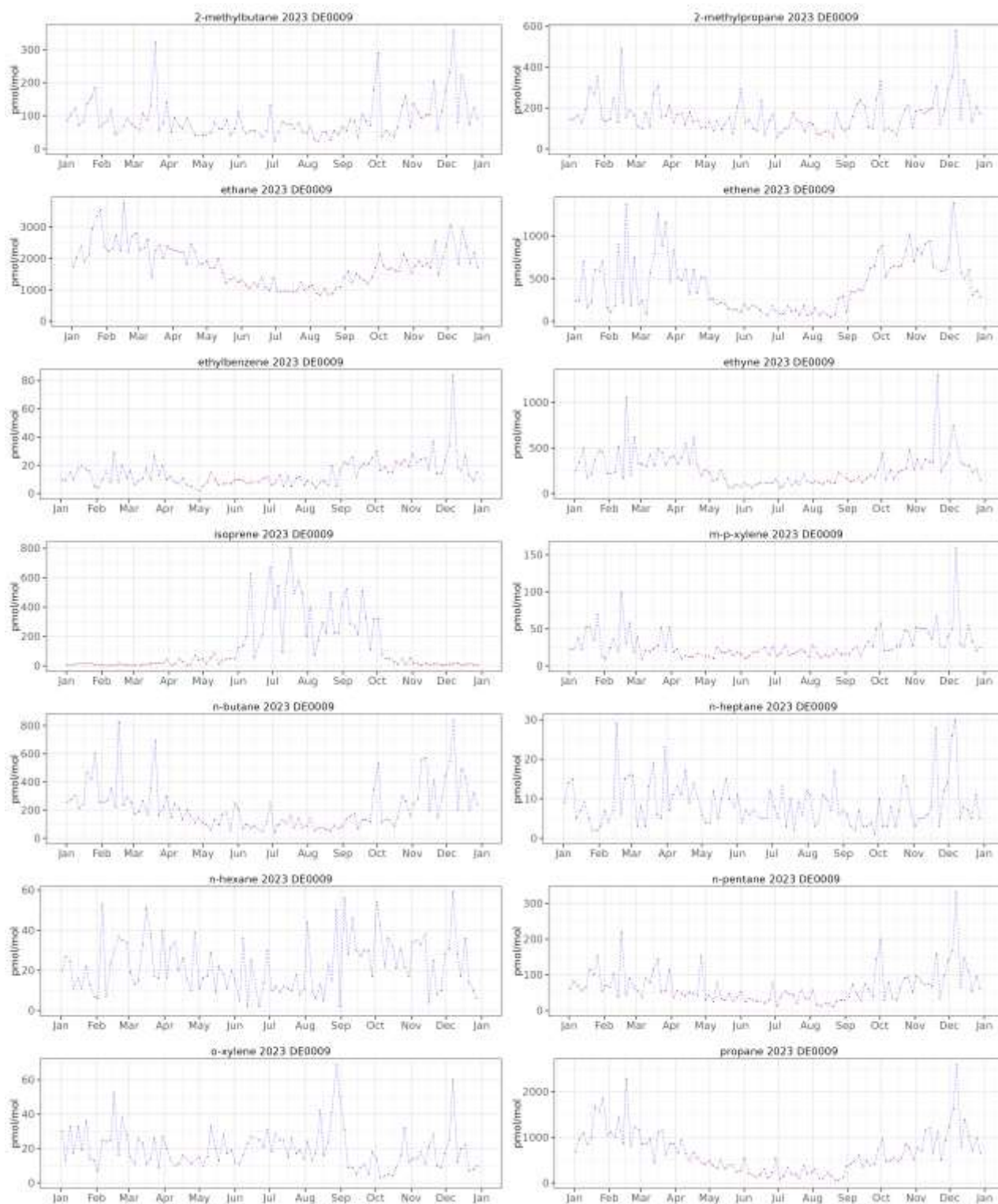




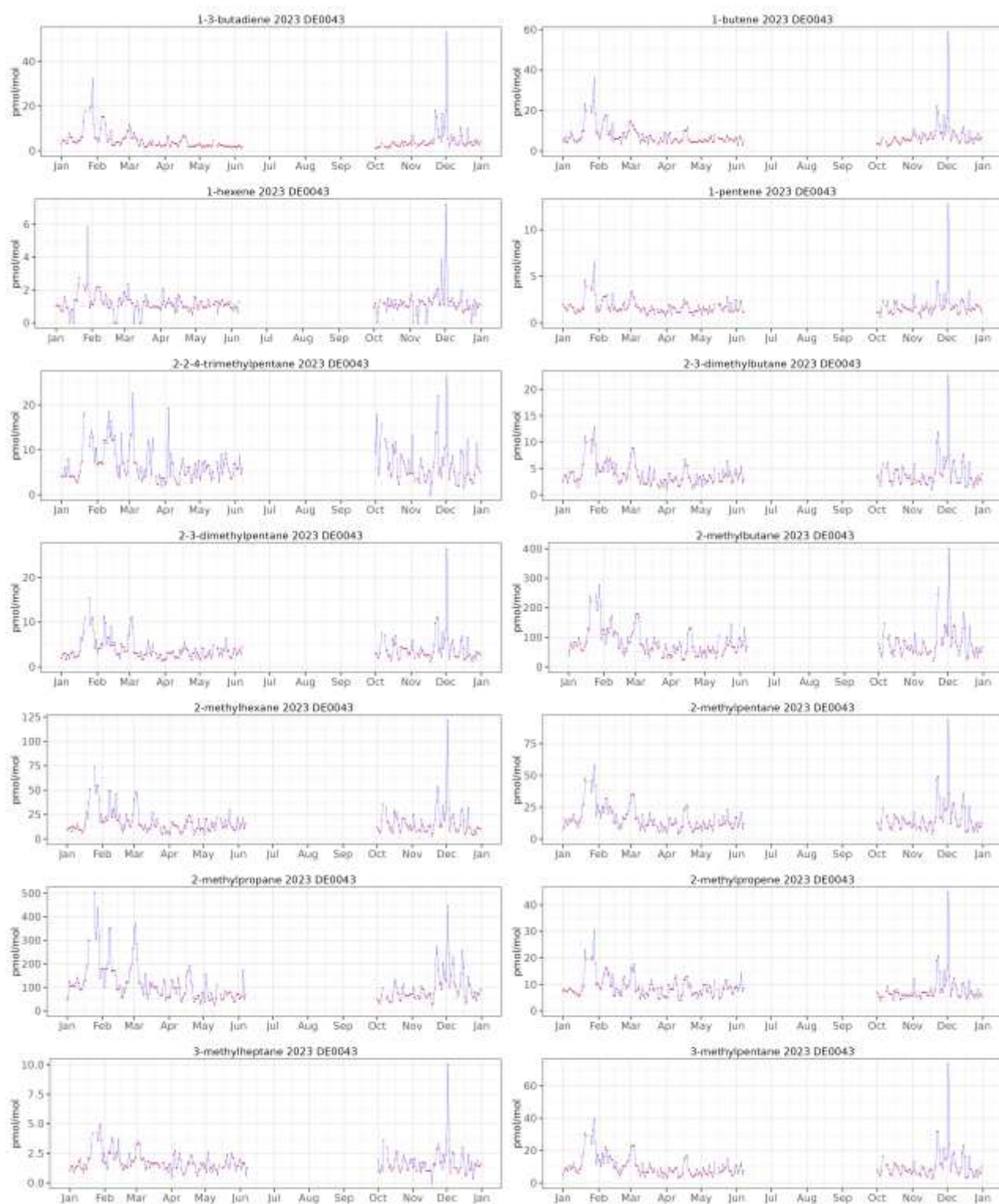


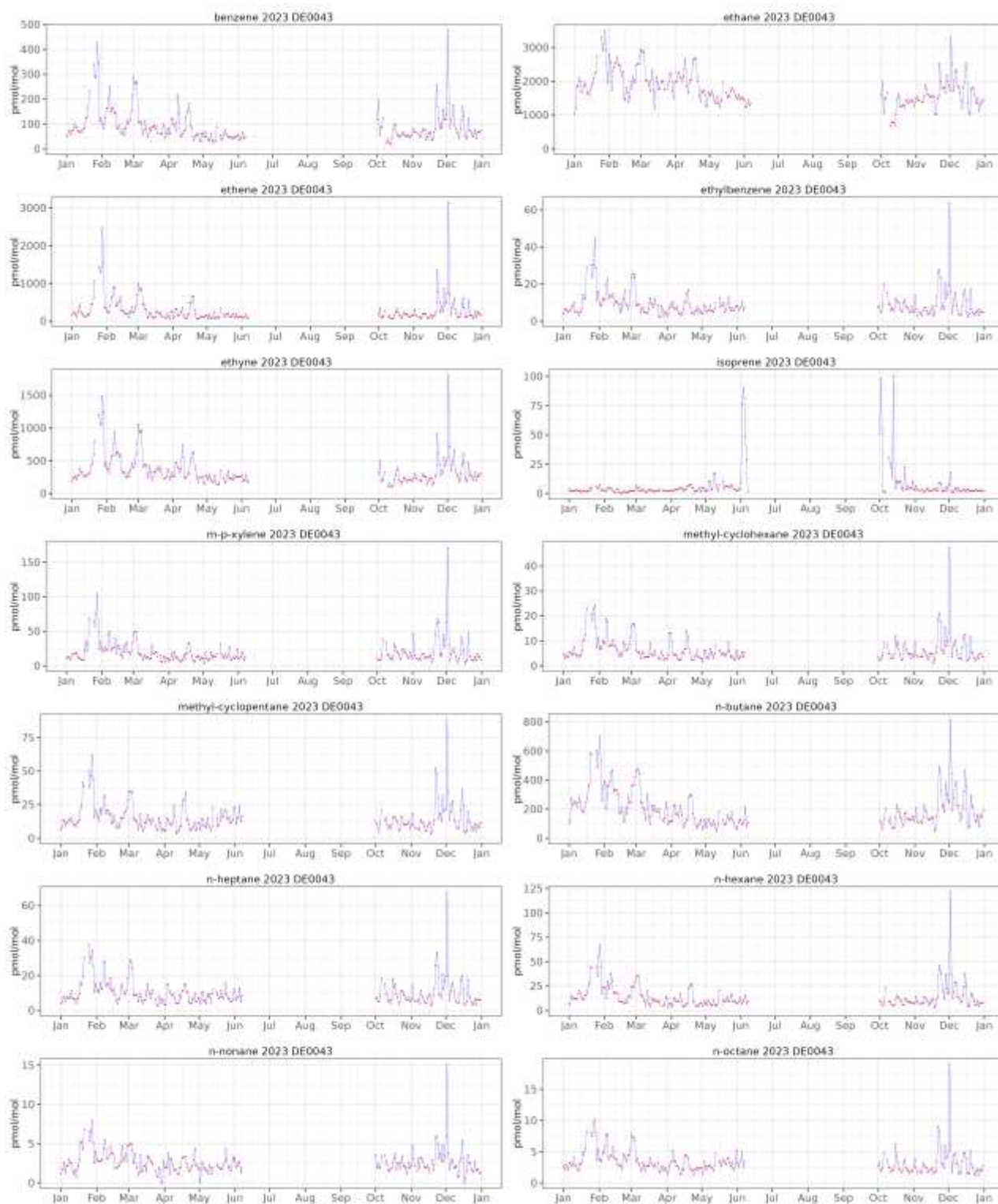


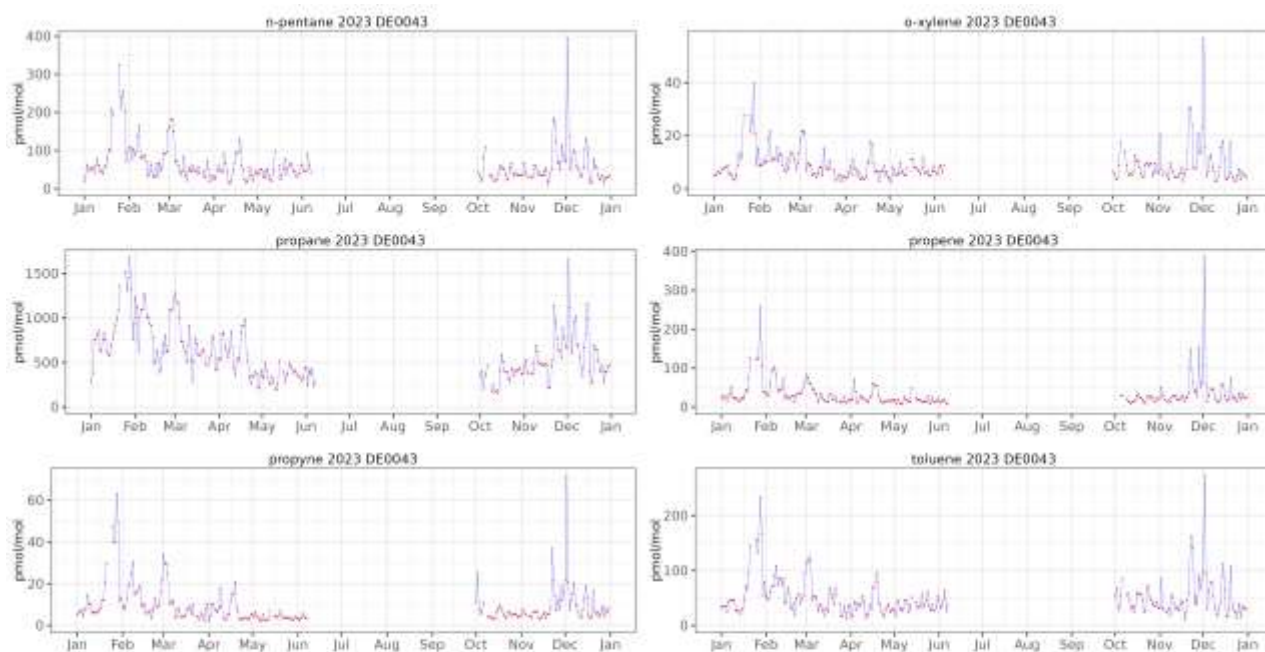


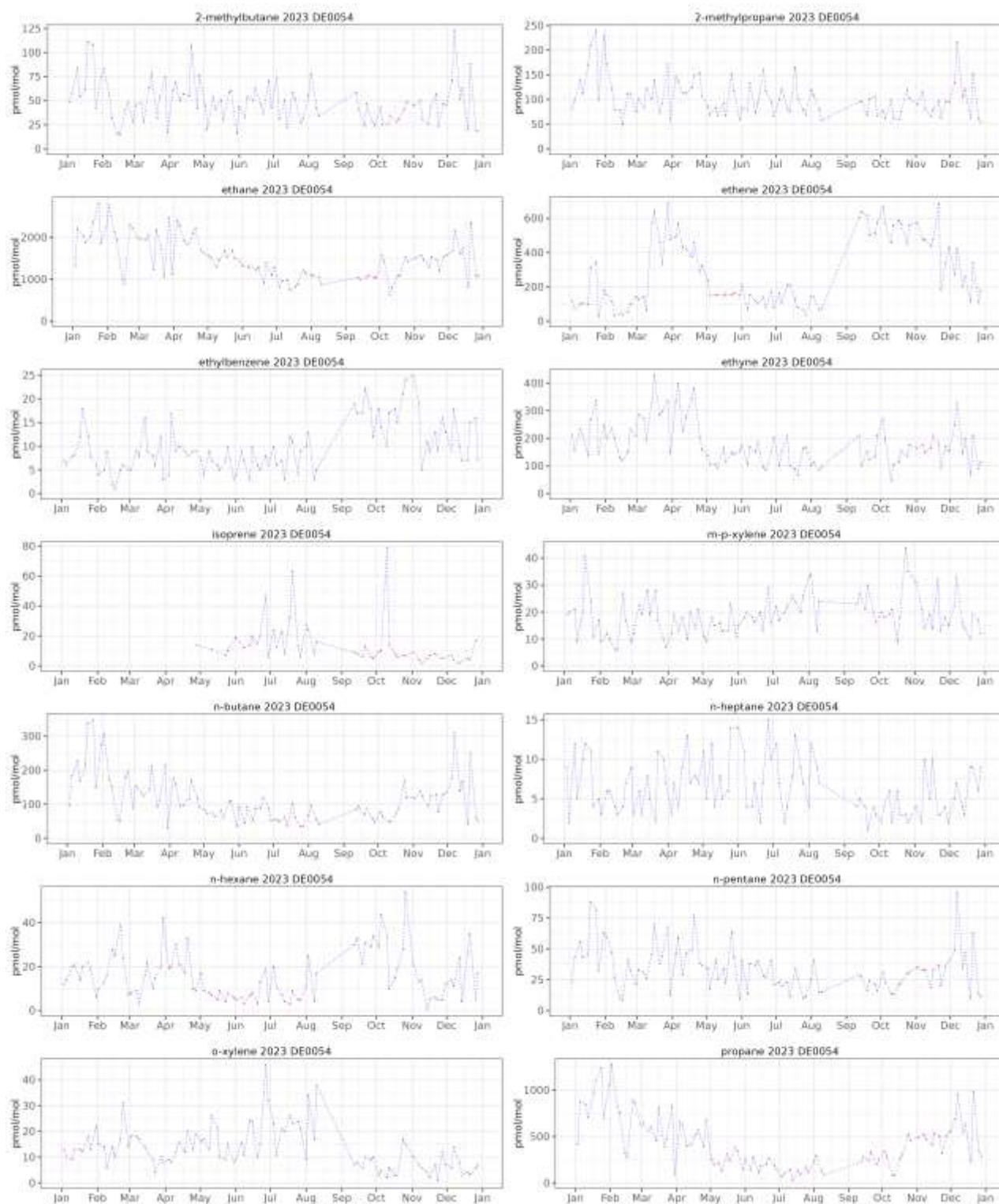




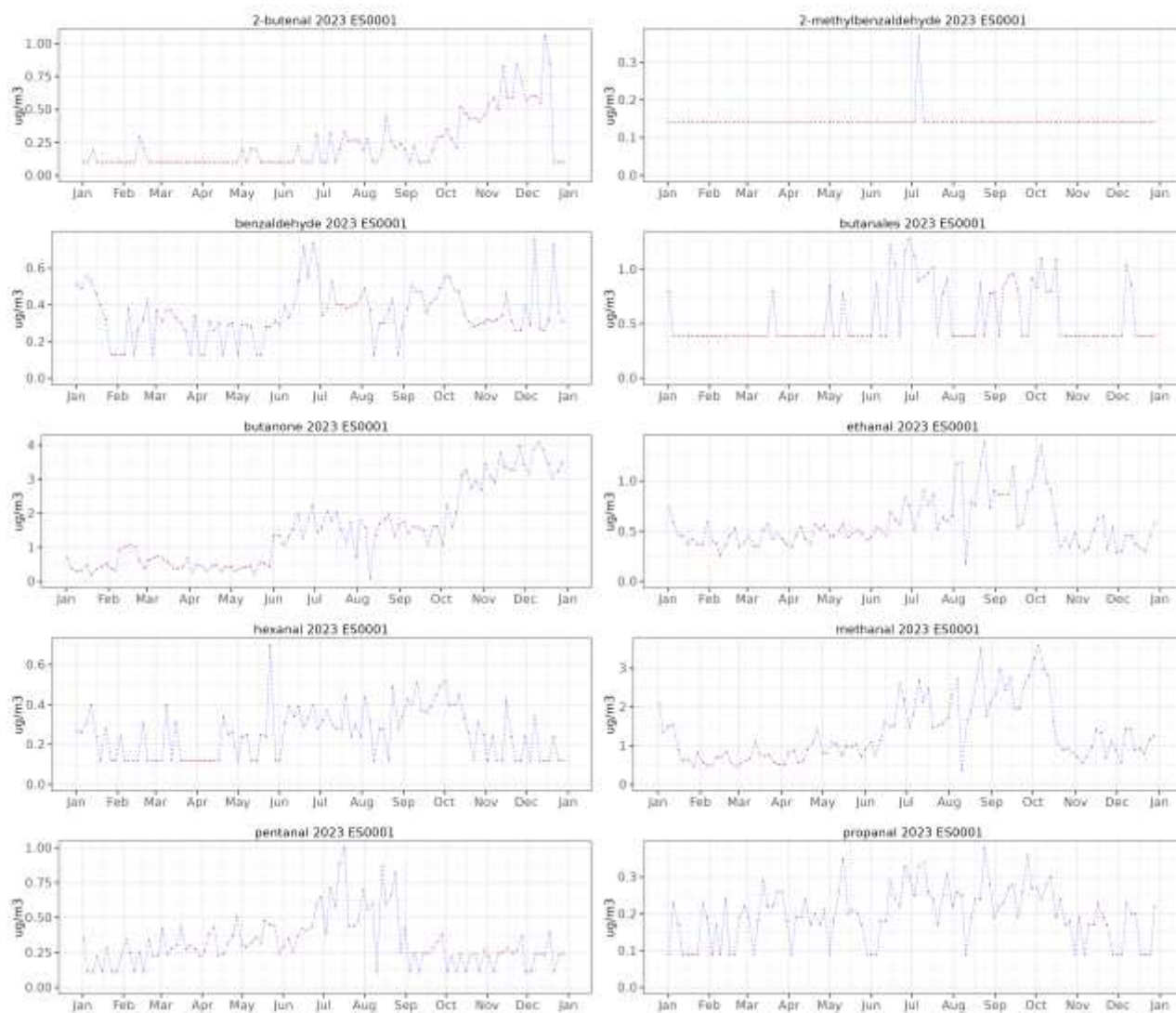


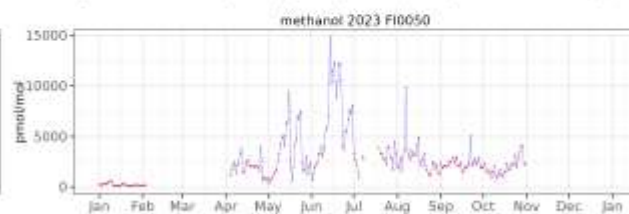
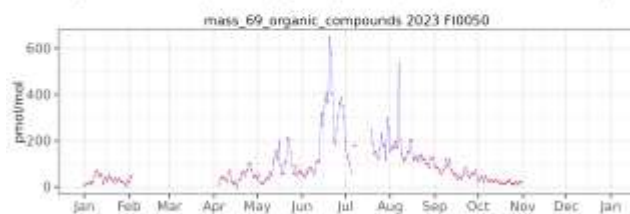
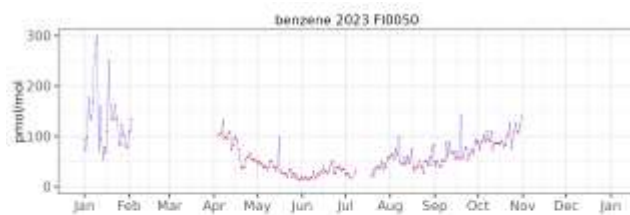


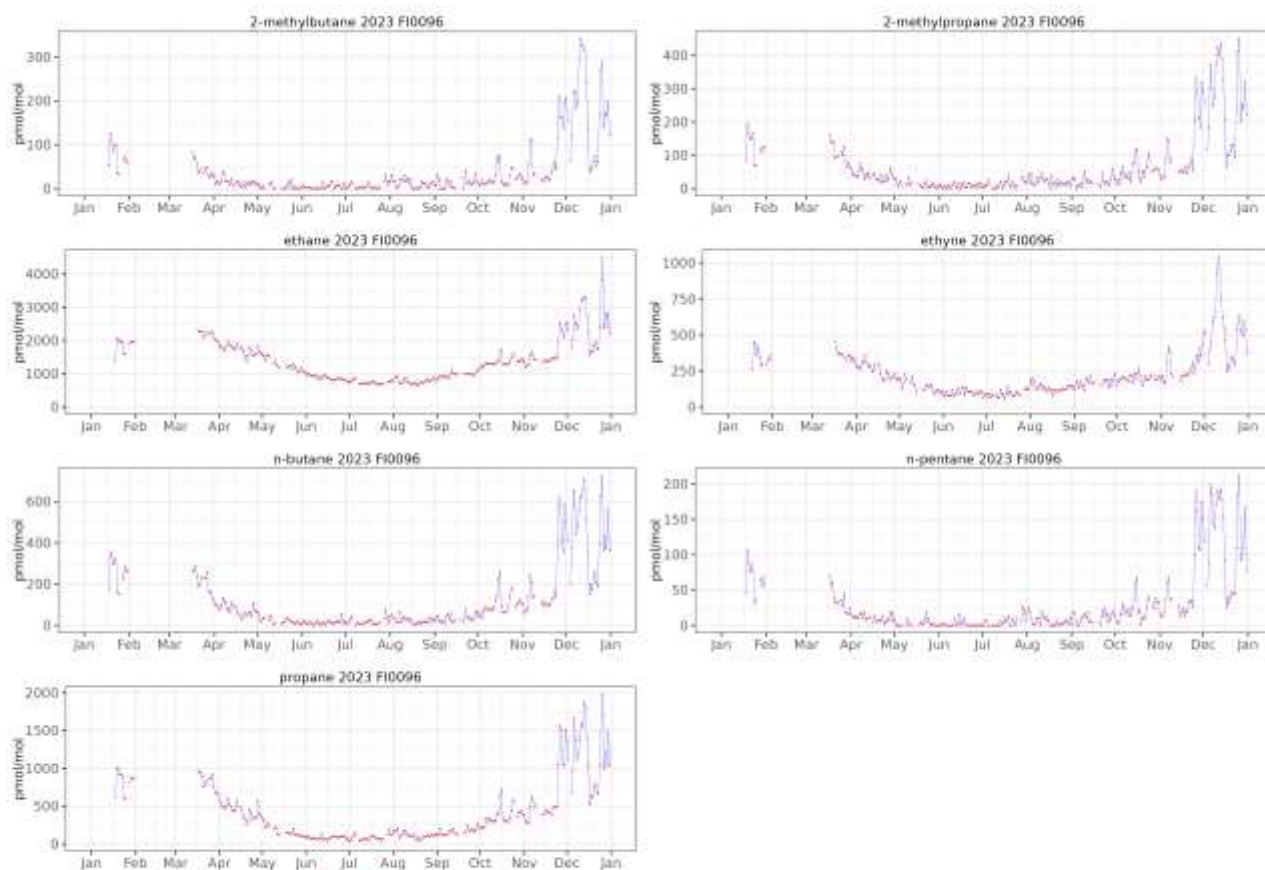


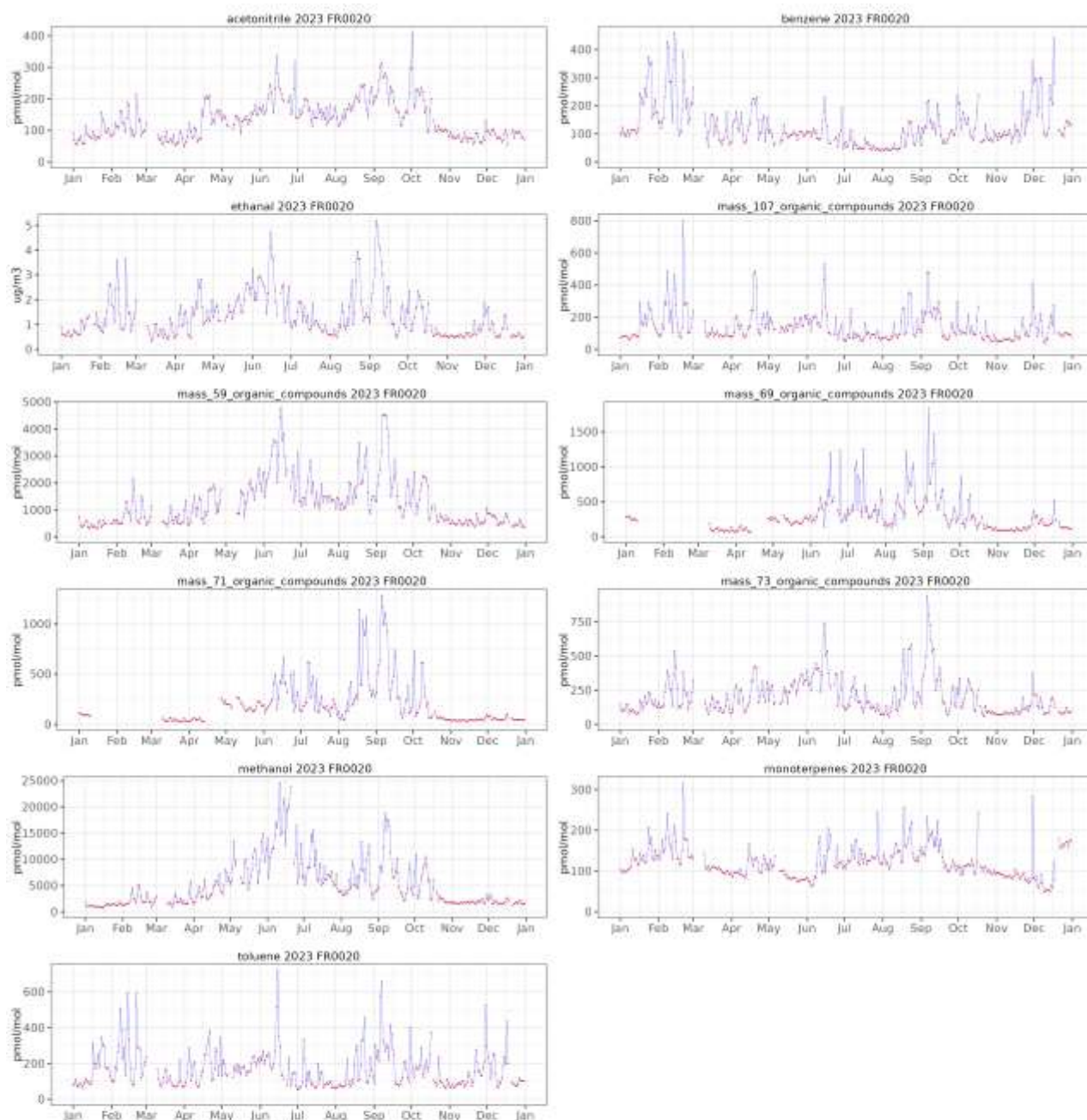


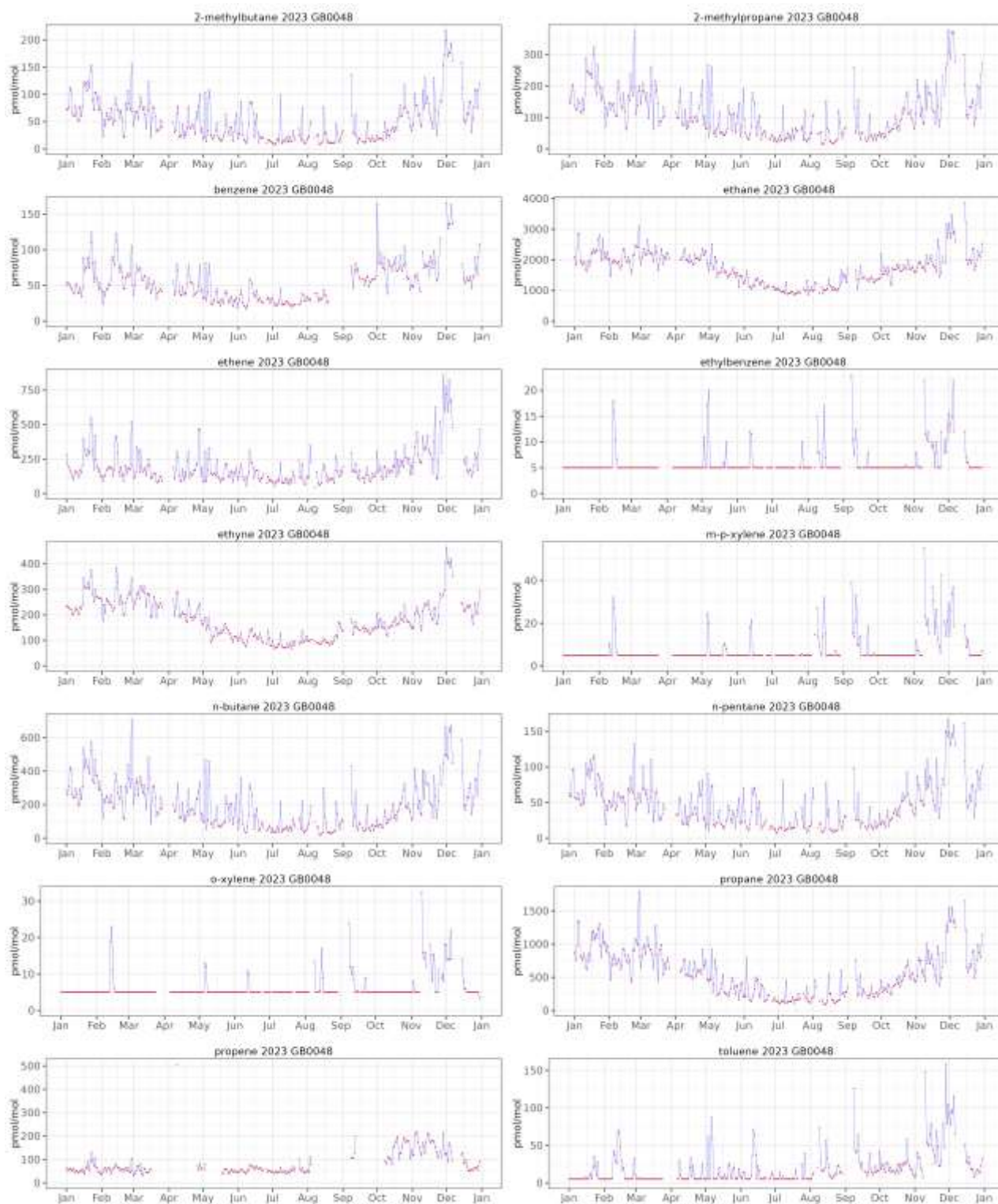


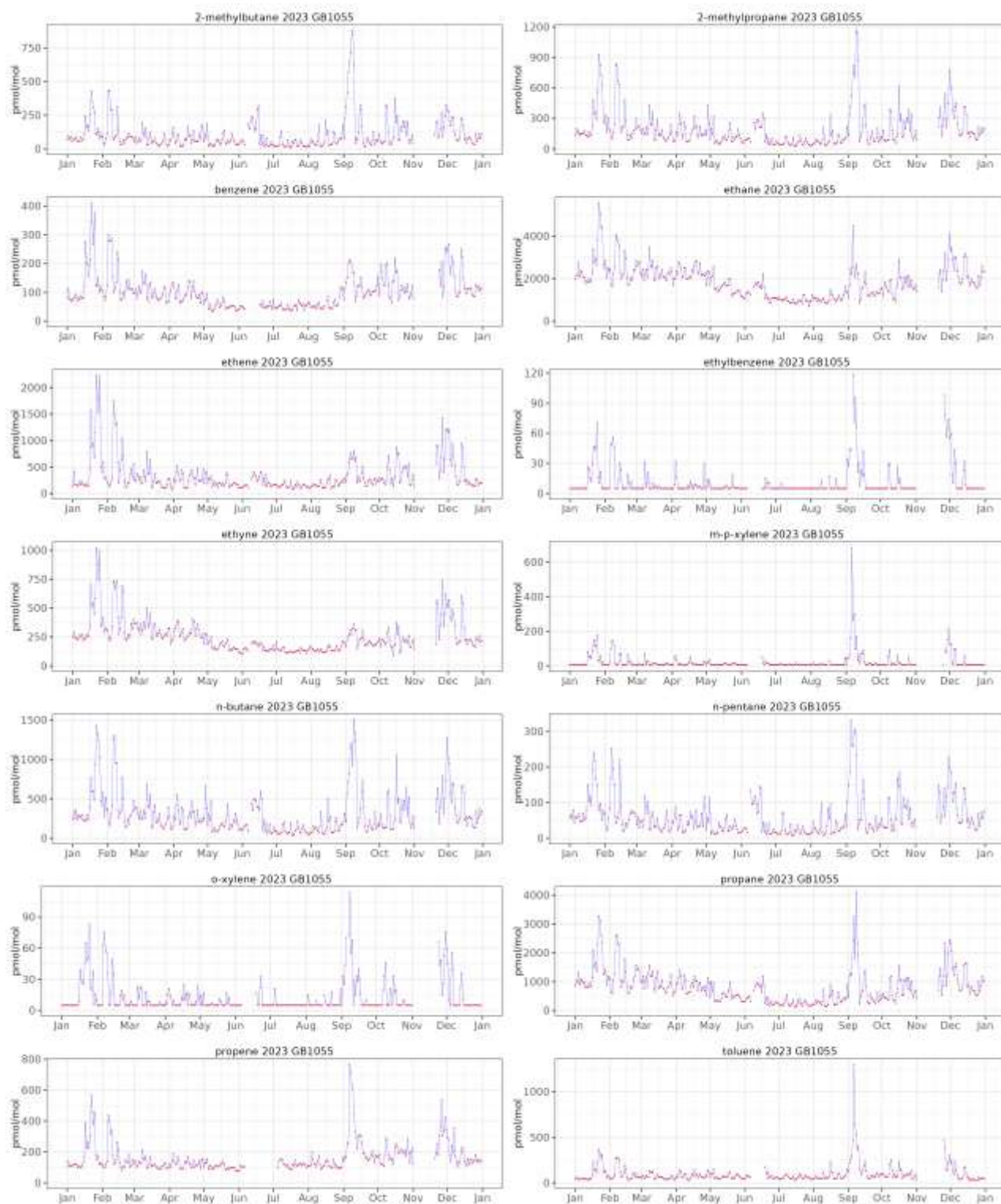


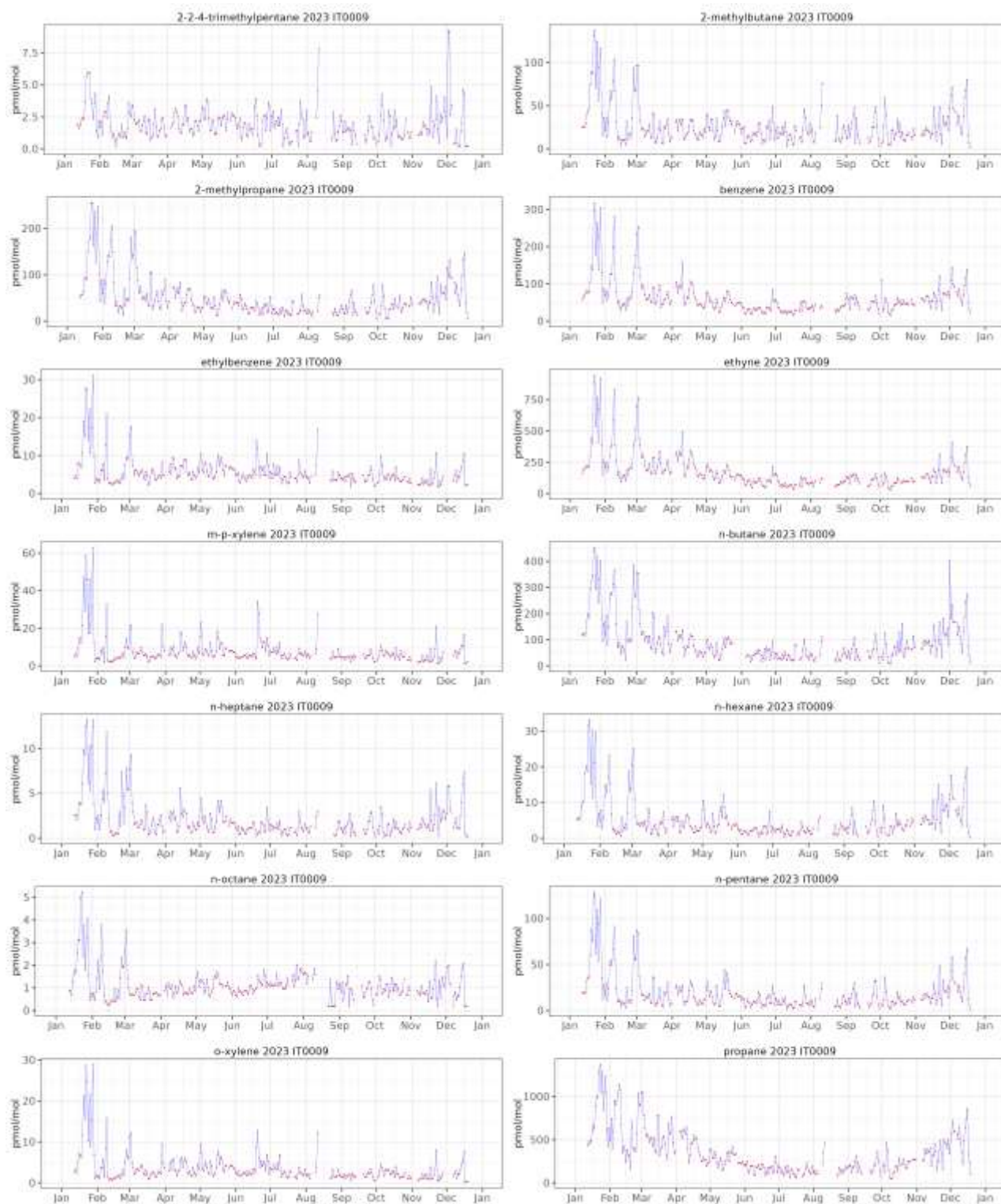


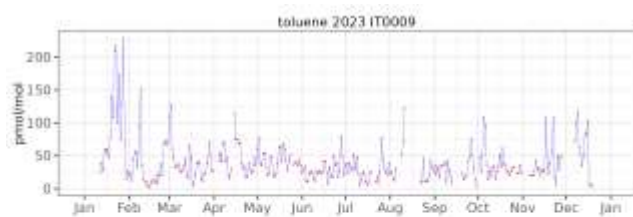


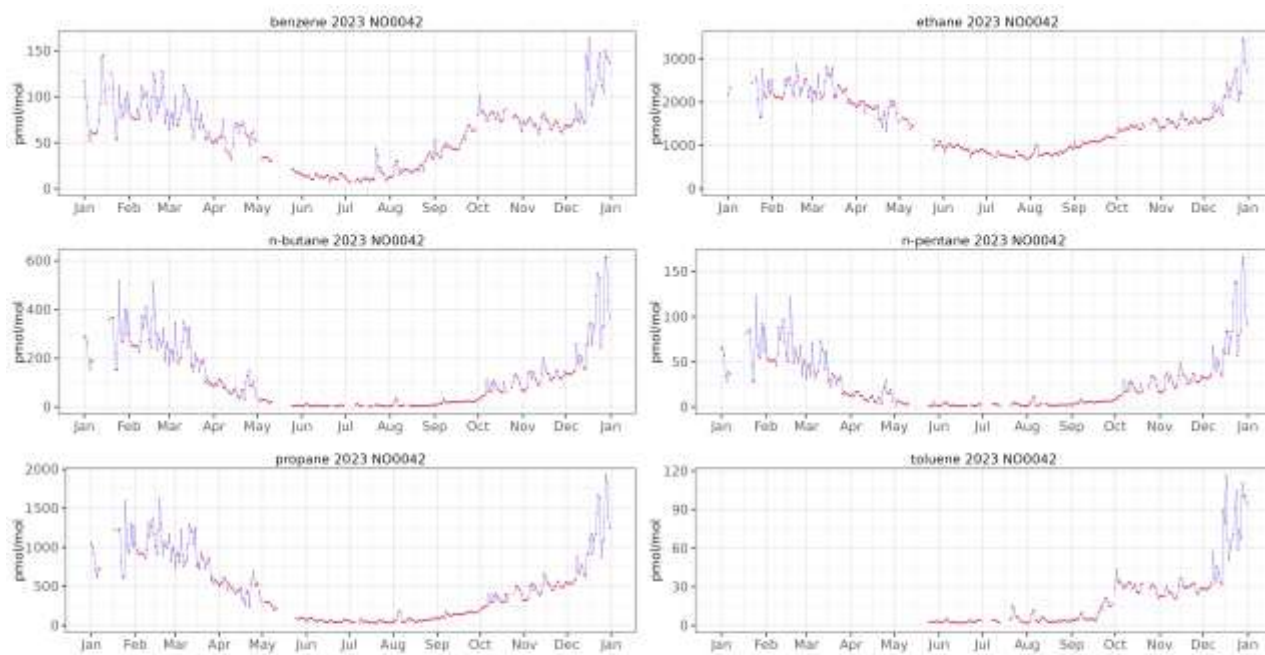












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