

On the Fate of VOCs During the Spring 2020 COVID-19 Lockdowns in Europe: A Study Based on the AirBase Data



Key Points:

- Anthropogenic volatile organic compounds (VOCs) were impacted during the lockdowns in Europe in 2020: traffic-related VOCs the most, industrial were impacted the least
- The signal and magnitude of the change depended on the sources and the meteorological conditions
- Isoprene, a VOC originating from biogenic emissions, was not impacted by the lockdowns

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract The rapid spread of the SARS-CoV-2 virus led many European governments to issue stay-at-home orders for the sake of controlling its impacts on the health systems. The associated decrease in human activities and therefore emissions provided a unique opportunity for a real world laboratory for atmospheric scientists. The impact on primary emissions, that is, NO₂, has been vastly studied but its consequences on secondary pollutants, O₃ and secondary organic aerosol, have been reported to a lesser degree and the understanding is more limited. One reason is the chronic imbalance in the attention dedicated to volatile organic compounds. In the present study, we report on the evolution of volatile organic compounds under lockdown conditions in Europe by analyzing the concentrations relayed to the Airbase service of the European Environmental Agency. Subsetting was performed to account for human activity and the influence of meteorology. Traffic or urban stations exhibited the most important reduction in benzene and, more substantially, toluene concentrations. Xylenes, trimethylbenzenes, and ethylbenzene also decreased under lockdown conditions, though less when the synoptic conditions were associated with slow flows. Acyclic alkenes evidenced no change or increased slightly, whereas n-alkanes increased. The evolution of the relative importance of the sources was investigated by means of diagnostic ratios (toluene to benzene and benzene to toluene to ethylbenzene) and exhibited a shift from traffic toward biomass/biofuel/coal burning, indicating a possible increase in the domestic use of solvents.

Plain Language Summary Volatile organic compounds are a class of airborne toxics, which are fundamental to understand and control air pollution. In this paper, we investigate the effect of the stay-at-home orders (commonly known as lockdowns) issued during the COVID-19 pandemic of 2020 in Europe on the concentrations of volatile organic compounds. We found that the concentration of volatile organic compounds originating in human activities was reduced. More so for traffic-related compounds and less so for industry-related ones. Biogenic compounds were not impacted. The information given in this paper can be useful for policy-makers seeking to reduce or control air pollution locally.

1. Introduction

Volatile organic compounds (VOCs) are gas phase air pollutants that are emitted from a variety of sources, among which chemical products, transportation, and natural sources are particularly relevant in urban areas (Kamal et al., 2016; Kansal, 2009; McDonald et al., 2018). VOCs contribute to adverse human health impacts, although the direct impact on health is largely limited to a small number of the VOCs, such as formaldehyde, furans, benzene, or toluene, exerting carcinogenic or mutagenic effects as well as causing acute and chronic diseases (Laurent & Hauschild, 2014). The indirect human health impact comes from their role as an important precursor to secondary organic aerosols (SOA) and ozone (O₃) production (e.g., Ng et al., 2007; Taraborrelli et al., 2021). Ozone is formed photochemically via the oxidation of VOCs in the presence of nitrogen oxides (NO_x = NO₂ + NO) (Haagen-Smit & Fox, 1954; Sillman, 1999). Its adverse health effects are linked to respiratory and cardiovascular diseases, with 365,000 to 423,100 deaths globally attributed to O₃ in 2019, with the vast majority of the deaths occurring in urban and peri-urban areas (Jerrett et al., 2009; Malashock et al., 2022; Murray et al., 2020). As such, it is critical to reduce air pollution to adequately protect human health. One of the challenges for effective air quality regulation, specifically in the context of ozone and ozone precursors, is understanding how reductions of primary emissions will affect atmospheric concentrations given the complexity of ozone-NO_x-VOC sensitivity (Sillman, 1999).

The global COVID-19 pandemic that started at the end of 2019 led many countries to impose substantial restrictions on activities and mobility in an effort to contain the spread of the virus. As such, dramatic changes in

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emissions, especially those related to mobility and travel behavior, were observed over a very short time (e.g., Benita, 2021; Gkatzelis et al., 2021; Kraus & Koch, 2021). This has provided the scientific community a global real-world experiment to provide insight into dramatic emission reductions (Diffenbaugh et al., 2020; Kroll et al., 2020).

A growing number of studies have been published investigating the changes in emissions and their effect on atmospheric concentrations of air pollutants, such as NO₂, PM, and O₃ especially in urban areas (e.g., von Schneidmesser et al., 2021; Gkatzelis et al., 2021; Rodríguez-Urrego & Rodríguez-Urrego, 2020). Schneider et al. (2022) looked at ca. 50 cities in Europe, accounting for the influence of meteorology, emission trends, chemistry, and temporally variant lockdown period and showed that decreases were observed for NO₂, PM_{2.5}, and PM₁₀ starting in March 2020, which corresponds to the lockdown policy implementations across most European cities, and lesser increases in O₃ but especially at the end of April. Wijnands et al. (2022) used machine learning techniques to look at ca. 900 cities globally comparing their estimates to previous publications that also accounted for the influence of meteorology. They showed decreases in NO₂ and increases in O₃ for Spain, the UK, and Europe that were generally consistent with those from Petetin et al. (2020), Ropkins and Tate (2021), and Ordóñez et al. (2020), respectively. For example, Ordóñez et al. (2020) showed that NO₂ decreased by 5%–55% relative to previous years for 80% of the sites over the European continent. This was mostly attributed to emissions reductions. Although maximum daily 8-hr average (MDA8) O₃ showed decreases over Iberia and increases elsewhere, in particular over northwestern to central Europe ranging from 10% to 22%, the increases in O₃ were dominated by meteorology (Ordóñez et al., 2020).

A more limited number of studies have evaluated the effect of the COVID-19 restrictions on VOCs pollutant concentrations. The understudy of VOCs when compared to other air pollutants is well-known (e.g., von Schneidmesser et al., 2023). The majority of the studies that exist are focused on China (Li et al., 2020; T. Liu et al., 2020; Pei et al., 2022; Stavrakou et al., 2021; H. Wang et al., 2022; M. Wang et al., 2021; K. Zhang et al., 2022; Z. Zhang et al., 2022) and India (Korhale et al., 2021; Kuttippurath et al., 2022; Mor et al., 2021; Nabanita et al., 2022; Pakkattil et al., 2021; Rathod et al., 2021) with far fewer studies focused on Europe (Sbai et al., 2021; Viteri et al., 2021; C. Zhang & Stevenson, 2022), North America (Jiang et al., 2021) or other regions (Ibragimova et al., 2021; C. M. G. Salvador et al., 2022; Siciliano et al., 2020). Unfortunately, a large number of these studies simply compare concentrations pre-COVID (either in the months prior or to the same time period from the year(s) previous) to concentrations during the lockdowns and do not account for the influence of meteorology, which has been shown to have a substantial effect on the changes in concentration (e.g., Goldberg et al., 2020; Kroll et al., 2020; H. Wang et al., 2022).

Of those studies that evaluate changes in VOCs where meteorology has been taken into account in some form, the majority include modeling and are focused on China. T. Liu et al. (2020) evaluated changes in PM_{2.5}, MDA8 O₃, NO_x, and VOCs using observations and modeling for four major cities in the Yangtze River Delta (YRD) in China. They found that NO_x reductions contributed substantially to the increase in MDA8 O₃ (38%–59%), whereas the reductions in VOCs were too small to counter the resulting increase, contributing only between 5% and 9% to MDA8 O₃ reductions. Both meteorology (17%–49%) and emissions (29%–52%) contributed to the changes in MDA8 O₃. H. Wang et al. (2022) similarly evaluated the effects of the lockdown in the YRD with modeling an observations and found an 88% increase in O₃, and decreases of NO₂ and VOCs of 61% and 38%. They attributed ca. two-thirds of the O₃ increase to meteorological drivers. They also note that reduced O₃ production is overwhelmed by the weakened NO titration, resulting in the substantial increase in O₃, as well as an overall reduction in O_x. 81% of the reductions in O_x were attributed to VOC reductions. Speciated VOCs were not evaluated beyond the context of model evaluation. K. Zhang et al. (2022) also evaluated a city in the YRD and noted that the largest drop in concentrations was observed for aromatics, followed by oxygenated VOCs, and alkenes, which they attributed to the changes in industrial activities and traffic volumes, whereas formaldehyde (HCHO) and methanol increased, which was attributed to secondary formation. Stavrakou et al. (2021) evaluate the lockdown changes over China using satellite observations of NO₂, HCHO, glyoxal (CHOCHO), and peroxyacetyl nitrate (PAN) as well as a regional model. The model simulations and satellite observations show good agreement with ca. 40% decrease of NO₂ and 9%–20% decrease in HCHO, CHOCHO, and PAN observed during peak shutdown (February 2020) relative to 2019.

Two studies from China used positive matrix factorization to quantify the contribution of sources of the VOCs to observed changes (Pei et al., 2022; Z. Zhang et al., 2022). Both studies identified traffic sources as well as

industrial processes, solvent use, and cooking in the VOCs analysis. Pei et al. (2022) specifically noted the substantial reductions in industrial (−49%) and diesel exhaust (−42%) emissions relative to the lesser reductions in gasoline-related emissions (−8.8%).

Among the studies evaluating the effect of the lockdown on VOCs emissions in Europe, C. Zhang and Stevenson (2022) looked at the lockdown in the context of the longer term evolution of NO_x and VOCs concentrations and noted that O_3 formation in London was in the VOC-limited regime, attributing the increase in O_3 to the decrease in NO_x emission. Sbai et al. (2021) evaluated the changes in a suite of air pollutants, including VOCs, for Lyon, France, noting that VOCs levels remained relatively stable during the lockdown.

To have a complete picture regarding the effect of emission reductions and the effect on O_3 , not only changes in NO_x should be evaluated but also non-methane VOCs. Overall, no detailed evaluation of changes in measured speciated VOCs that also account for meteorological data, and consider speciated VOCs data for Europe, have been published to this point. We aim to fill this gap by evaluating changes in speciated VOCs from data from 25 countries across Europe as reported to Airbase.

2. Data and Methods

2.1. Synoptic Meteorological Patterns

Concentration levels of air pollutants in ambient air are highly conditioned not only by the intensity of the emissions from their sources and the atmospheric processing they undergo, but also by the atmospheric conditions produced under specific synoptic meteorological patterns (SMP), which influence the latter (Baudic et al., 2016; Debevec et al., 2021; Kim et al., 2019; Yuan et al., 2012). For this reason, the main SMPs over Europe along the study period were identified by means of a circulation classification procedure (Huth et al., 2008; Philipp et al., 2016). By comparing concentrations before and during the lockdown that occurred throughout the same SMP, we try and compartmentalize the analysis in terms of the influence of dispersion and stagnation/stability and, to some extent, photochemistry. Circulation classification techniques classify a high number of circulation patterns into a smaller group of SMPs according to their similarity and frequency of occurrence. Each one represents a specific circulation type of air masses over the region of study at the synoptic scale.

In this study, global daily fields of sea level pressure (at 12 UTC) were obtained from the National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Reanalysis data set (Kalnay et al., 1996) provided by NOAA/OAR/ESRL PSD, USA at a spatial resolution of 2.5° covering the timeframe between January 2016 and July 2020. Classifications of atmospheric circulations using sea level pressure daily fields have demonstrated to be very useful for discriminating SMP associated to the occurrence of high pollution episodes produced by natural (Russo et al., 2020) and anthropogenic sources (P. Salvador et al., 2021; Valverde et al., 2015).

Then, a cluster-based technique (nonhierarchical k -means cluster analysis) was carried out to classify each daily sea level pressure field in the domain 40°W – 30°E 20°N – 80°N into one specific SMP. A brief description on how the SMPs were identified is given in Text S11 in Supporting Information S1. More detail on the methodology followed can be found by Belis et al. (2019) and (P. Salvador et al., 2021, and references therein) whereas more detail on this particular application can be found in von Schneidemesser et al. (2021).

2.2. VOCs

Atmospheric concentrations of 18 VOC species measured at monitoring stations by networks reporting to the European Airbase system were retrieved for the years 2016–2020 on 29 April 2022. The VOC species present in the Airbase data set are: ethane, propane, butane, pentane, hexane, ethene, trans-2-butene, cis-2-butene, isoprene, ethyne (acetylene), benzene, toluene, m,p-xylene, o-xylene, 1,3,5-trimethylbenzene, 1,2,3-trimethylbenzene, 1,2,4-trimethylbenzene, and ethylbenzene. These 18 species are reported from a total 542 stations in 25 countries (Table 1).

All the VOCs concentrations used in the present study were validated and verified by the national agencies before being reported to Airbase and are made available as the E1a data set. The quality assurance/quality control (QA/QC) steps conducted by the individual contributing country's agencies are assumed as appropriate and valid and not further investigated. Additional QA/QC steps were embedded in the subsetting and are described below and in

Table 1
Spring 2020 Lockdown Dates, Number of Monitoring Stations, Number of Species and Number of Valid Data Points for Each Country Included in the Study

Country	Lockdown start	Lockdown end	Number of stations	Number of species	Number of valid data points
Austria	16.03.2020	13.04.2020	4	1	35,976
Belgium	18.03.2020	04.05.2020	23	13	322,965
Bulgaria	13.03.2020	13.04.2020	18	6	652,072
Switzerland	17.03.2020	27.04.2020	2	2	155,112
Cyprus	24.03.2020	13.04.2020	1	1	40,057
Czechia	16.03.2020	12.04.2020	8	1	12,895
Germany	16.03.2020	11.04.2020	30	12	1,932,854
Denmark	12.03.2020	13.04.2020	1	10	7,363
Estonia	11.03.2020	11.04.2020	4	1	6,576
Spain	14.03.2020	09.05.2020	79	15	2,678,265
France	17.03.2020	11.05.2020	8	1	285,173
United Kingdom	23.03.2020	04.07.2020	4	17	1,824,079
Greece	23.03.2020	04.05.2020	6	1	168,914
Croatia	18.03.2020	11.05.2020	3	1	96,283
Hungary	28.03.2020	10.04.2020	10	5	59,194
Ireland	12.03.2020	18.05.2020	2	5	10,511
Italy	09.03.2020	18.05.2020	193	1	6,653,668
Lithuania	16.03.2020	18.06.2020	1	1	25,703
Luxembourg	16.03.2020	20.04.2020	1	2	68,095
Latvia	13.03.2020	14.04.2020	4	2	237,257
Netherlands	15.03.2020	06.04.2020	4	2	293,801
Poland	13.03.2020	11.04.2020	27	18	1,391,973
Portugal	18.03.2020	02.05.2020	1	1	25,219
Romania	25.03.2020	12.04.2020	98	1	1,703,984
Slovakia	12.03.2020	22.04.2020	11	1	428,466

the Supplement Information (SI). In the present work, only verified data with the observation verification flag = 1 (*Data has gone through full quality assurance and quality control by data provider/owner. Data is considered verified and can be formally used for its reporting purposes.* <https://dd.eionet.europa.eu/vocabulary/aq/observationverification>) were used. Furthermore, the data marked as validated with the validity flag = 1, 2, or 3 <https://dd.eionet.europa.eu/vocabulary/aq/observationvalidity> was selected. A validity flag value of 1 refers to *primary data considered valid by the data provider for its related reporting purposes*, whereas values of 2 refer to *data considered valid by the data provider for its related reporting purposes. However, the value has been measured below the detection limit and the value reported is the detection limit* and of 3 to *data considered valid by the data provider for its related reporting purposes. However, the value has been measured below the detection limit and the value reported is half of this detection limit*. In the case of data identified with a validity flag = 2, the reported value, that is, the detection limit, has been replaced by half its value before the analysis conducted for the present work. In this way, all concentrations reported as measured below the detection limit were replaced by half the detection limit value. The distribution of the fraction of data points measured below the limit of detection among the stations for each VOC species is presented in the SI.

Only measurements with an averaging time of 1 day or 1 hour were kept. Here, caution must be observed when interpreting the results for species which are measured with different time resolutions at different and/or the same stations, since correlations between VOCs may change with changing time resolutions (Salameh et al., 2019). Such species are toluene (31 stations with hourly measurements and 24 stations with daily measurements), m,p-xylene (7 stations with hourly measurements and 26 stations with daily measurements), and benzene (289 stations

Table 2
Evolution of the Data Set and Its Subsettings

Operation	Number of data points	Number of stations	Number of species	Number of countries	Detail
Original downloaded data set	19,116,455	542	18	25	Averaging time equal to “day” or “hour,” validated and verified data
Subset for the dates of interest	3,446,286	540	18	25	Lockdown (2020) and the corresponding periods in 2016–2019
Subset for the time resolution	3,404,054	540	18	25	Reduce time resolution to daily for station/species pairs which have both “day” and “hour” averaging time
Subset for data coverage	2,225,014	344	13	19	Keep only station/species pairs for which the data coverage is above 70%
Subset to remove negative values	2,202,092	340	13	19	Keep only station/species pairs for which there are no negative data points
Subset to remove low variability	2,200,742	340	13	19	Keep only station/species pairs for which the relative standard deviation is >1%

with hourly measurements, 37 stations with daily measurements, and 15 stations with daily and hourly measurements).

The data set thus downloaded consisted of 19,116,455 data points. Quality assurance reports are provided by the reporting national agencies under https://discomap.eea.europa.eu/App/AQViewer/index.html?fqn=Airquality_Dissem.b2g.Measurements#.

The data were subset within each country for the respective lockdown dates in 2020 (Table 1) and the corresponding periods in 2016–2019. For some station/species pairs, the averaging time changed within the course of the period of interest. For those station/species pairs, the averaging time was reduced to one day by averaging the hourly concentrations and keeping the daily ones. The comparison of the VOCs concentrations between lockdown conditions in 2020 (LD) and no-lockdown conditions in 2016–2019 (NL) was conducted at monitoring stations for which the data coverage during the lockdown period and during the same periods in 2016–2019 was above 0.7 in both cases. The data coverage condition removed 5 species completely from the data set: 1,2,3-trimethylbenzene, ethane, ethene, propane, and acetylene. Despite the considerations on the LOD (see above), some time series still presented negative values at this stage. Those station/species pairs were removed from the data set. A few time series presented the same value for all (or almost all) the timestamps. As such, station/species pairs with a time series of a relative standard deviation within a same year below or equal to 1% were removed from the data set.

Figures S1 to S18 in Supporting Information S1 show the number of stations as a function of the data coverage. The plots show that there was a discrepancy in the distribution of the stations with less stations in total reporting for 2020 but more stations reporting with data coverage closer to 1. Due to this discrepancy, we opted to apply the threshold to both periods individually rather than to the 5 years in bulk.

The subsetting procedure described above and summarized in Table 2 resulted in a data set with 2,200,742 data points from 344 monitoring stations located in 19 countries: Austria, Belgium, Bulgaria, Switzerland, Cyprus, Germany, Estonia, Spain, France, Greece, Croatia, Hungary, Ireland, Italy, Latvia, the Netherlands, Poland, Romania, and Slovakia. The data points of the final data set refer to the following 13 species: butane, pentane, hexane, trans-2-butene, cis-2-butene, isoprene, benzene, toluene, m,p-xylene, o-xylene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, and ethylbenzene.

Benzene was the most measured species: 338 monitoring stations from 19 countries, corresponding to 1,922,702 data points (Tables 3 and 4). The most measured species following benzene were: toluene (163,092 data points, 54 stations, 9 countries), m,p-xylene (43,933 data points, 33 stations, 5 countries), and ethylbenzene (23,333 data points, 30 stations, 4 countries).

Monitoring stations in the Airbase data set can be classified in terms of *area* or *type*. Station *types* represent a differentiation between *traffic*, *industrial*, and *background* stations, whereas station *areas* correspond to a

Table 3
Data Coverage for the VOC Species in the Data Set Considered for the Analysis

Species	Number of			Type			Area			Coverage 2016–2019		Coverage 2020	
	Countries	Data points	Stations	Tr.	Ind.	Bgd.	Urb.	Sub.	Rur.	Min	Max	Min	Max
1,2,4-Trimethylbenzene	1	2,093	14	0	2	12	1	8	5	0.84	1	0.96	1
1,3,5-Trimethylbenzene	1	1,761	11	0	2	9	1	8	2	0.84	1	1	1
Hexane	1	2,970	18	1	2	15	2	11	5	0.84	1	0.96	1
Ethylbenzene	4	23,333	30	5	2	23	12	13	5	0.71	1	0.84	1
Toluene	9	163,092	54	18	6	30	31	14	9	0.71	1	0.79	1
Benzene	19	1,922,702	338	128	62	148	242	68	28	0.7	1.05	0.7	1.03
Isoprene	1	809	7	0	1	6	1	3	3	0.84	1	0.96	1
cis-2-Butene	1	687	6	0	2	4	1	3	2	0.84	1	0.96	1
Pentane	1	3,003	18	1	2	15	2	11	5	0.84	1	0.96	1
Butane	1	1,128	10	0	2	8	1	4	5	0.84	1	0.96	1
m,p-Xylene	5	43,933	33	8	2	23	14	13	6	0.71	1	0.86	1
o-Xylene	3	35,990	30	6	2	22	11	14	5	0.71	1	0.85	1
trans-2-Butene	1	916	8	0	2	6	1	4	3	0.84	1	0.96	1

Note. Station types: Traffic (Tr.), Industrial (Ind.) and Background (Bgd.). Station areas: Urban (Urb.), Suburban (Sub.), Rural (Rur.).

differentiation between *urban*, *suburban*, and *rural* stations. Some rural stations are discerned between *rural-near city*, *rural-regional*, and *rural-remote*. In the present analysis, all are considered *rural*. Table 3 gives an overview of the number of stations, differentiated by type and area, for each pollutant in the data set used for analysis.

Table 4
Number of Stations for Each Time Resolution of the VOC Measurements in the Data Set Considered for the Analysis

Species	Time resolution			Periodicity daily (days)						
	Hourly	Daily	Hourly and daily	1	2	3	4	5	6	7
1,2,4-Trimethylbenzene	0	14	0	4	10	0	0	0	0	0
1,3,5-Trimethylbenzene	0	11	0	4	7	0	0	0	0	0
Hexane	0	18	0	8	10	0	0	0	0	0
Ethylbenzene	3	27	0	17	10	0	0	0	0	0
Toluene	31	23	0	13	10	0	0	0	0	0
Benzene	286	37	15	23	10	1	1	1	1	0
Isoprene	0	7	0	0	7	0	0	0	0	0
cis-2-Butene	0	6	0	0	6	0	0	0	0	0
Pentane	0	18	0	8	10	0	0	0	0	0
Butane	0	10	0	0	10	0	0	0	0	0
m,p-Xylene	7	26	0	16	10	0	0	0	0	0
o-Xylene	5	25	0	15	10	0	0	0	0	0
trans-2-Butene	0	8	0	0	8	0	0	0	0	0

Note. Not all stations measured every day. The periodicity indicates how often a species was monitored, for example, 3 means once every third day.

When distinguishing individual station data for each of the 8 SMPs and for 2 days types (weekdays and weekends), the data set for a single station can be subdivided into 16 subsets, each consisting of hourly or daily concentrations during the 2020 lockdown and for the same dates in the 4 years prior. The following analysis focuses on the ratio of the means and medians (2020 vs. 2016–2019), and on the comparison between the concentrations distributions by means of the Wilcoxon-Mann-Whitney *U*-Test's *p*-value for the different subsets, and further differentiating for station types. The *p*-values are interpreted not as categorical but rather as continuous measures of the strength of evidence.

The ratio of the means (RoA, Ratio of the Averages), the ratio of the medians (RoM), and the *p*-value were thus retrieved 16 times each for a given VOC species measured at a given station. The objective is to answer the primary research question: whether the concentration during the lockdown is different from the concentration before the lockdown. By subsetting, within a station, for SMP and day type, we intend to isolate the uniqueness of the lockdown concentrations relative to the concentrations in the years prior from confounders. The results in terms of RoA and RoM were interpreted by means of box and whisker plots and cumulative distribution functions (CDF) differentiated by SMP. The CDF is represented on a plot where the abscissa is the ratio of concentrations ($\frac{\text{lockdown}}{\text{no lockdown}}$) and the ordinate is the cumulative quantile. The quantile at which the CDF crosses the line representing equal average (median) concentrations under lockdown and no lockdown conditions ($\frac{\text{lockdown}}{\text{no lockdown}} = 1$) indicates the fraction of stations for which there was a decrease in average (median) concentration. Furthermore, the ratio at which the CDF crosses the *quantile* = 0.5 indicates the median ratio of concentrations through all the stations.

3. Results

3.1. Circulation Classification Results

Eight SMPs were finally identified, which were consistent with synoptic patterns found in the literature (von Schneidemesser et al., 2021). Composite sea level pressure maps representing the mean SMP resulting from each cluster are showed in the Figure S19 in Supporting Information S1. Figure S20 in Supporting Information S1 shows the frequency of the SMPs during the period April–March for 2016–2020. The main general characteristics of each SMP can be summarized as follows:

- SMP-1 Strong baric gradient across western, central, and northern Europe. Fast W flows. It has occurred mainly in winter.
- SMP-2 High pressures in central and southwestern Europe. Highly stable atmospheric conditions over southern and central Europe and moderate SW flows over northern Europe. It has occurred mainly in winter and autumn.
- SMP-3 High pressures in western Europe extended across France and Germany. Highly stable atmospheric conditions over southern and central Europe and moderate W flows over northern Europe. It has occurred mainly in winter and autumn.
- SMP-4 Weak baric gradient over central and eastern Europe. Slow flows all over the continent. It has occurred mainly in summer.
- SMP-5 High pressures in northern Europe. Moderate N-NE flows over northern and central Europe and slow NE-E flows over southern Europe. It has occurred mainly in spring and autumn.
- SMP-6 High pressures in northern Europe extended toward the E. Moderate E flows. It has occurred mainly in spring and autumn.
- SMP-7 Weak baric gradient over central and eastern Europe. Moderate W flows over western Europe and highly stable atmospheric conditions over central, northern, and eastern Europe. It has occurred mainly in spring and summer.
- SMP-8 Strong baric gradient across N Europe. Fast NW-N flows over central, northern, and eastern Europe and moderate NW-N flows over western Europe. It has occurred mainly in spring and autumn.

3.2. Benzene

Benzene is a known carcinogenic present in gasoline, as additive, which presence in solvents has been strongly regulated (Bravo et al., 2002; Perry & Gee, 1995; Sekar et al., 2019). It is an important contributor to the source profile of combustion sources, such as wood burning (residential wood burning and wildfires, among others) and

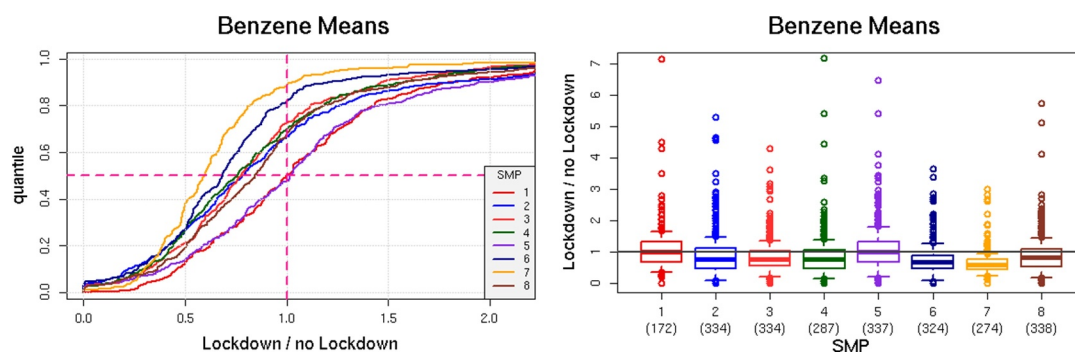


Figure 1. Cumulative distribution functions (left) and box and whisker plots (right) of the RoA on weekdays differentiated for the eight different SMPs. In the cumulative distribution plot, the horizontal dashed line represents the 50th percentile, the vertical dashed line represents RoA equal to unity, and the *x*-axis scale extends up to the 95th percentile. In the box and whisker plots, the box and whiskers represent, from bottom to top, the 5th, 25th, 50th, 75th, and 95th percentiles, the individual points are either below the 5th or above the 95th percentiles. The number in parenthesis denotes the number of subsets in the sample.

fossil fuels burning (motor vehicle exhaust, the burning of coal and oil), and of evaporative sources which has an atmospheric lifetime of a few days (e.g., Baudic et al., 2016; Borbon et al., 2018; Fraser et al., 1998; Gentner et al., 2009; Sauvage et al., 2009).

Benzene was the most measured VOC in the Airbase data set during the lockdown periods in 2020 and the corresponding periods in 2016–2019. It was measured at 338 stations in 19 countries, of which 128 are traffic stations, 62 are industrial stations, and 148 are background stations. When distinguishing individual station data for each of the 8 SMPs and for 2 days types (weekdays and weekends), the overall 1,922,702 benzene datapoints were subdivided into 4,412 valid subsets. As explained in Section 2, a subset refers to the data corresponding, for a given station, of the concentrations from a same SMP and day type before (2016–2019) and during the lockdown (2020).

Figure S53 in Supporting Information S1 shows the Benzene RoA by station over Europe. The RoA (RoM) was 1.0 ± 1.9 (1.1 ± 1.6) on average, and 0.78 (0.83) on median, through all the subsets. For a majority of subsets, the mean and the median of the concentrations were lower during the lockdown than in the 4 years prior for the same monitoring station, SMP and day type, as confirmed by the *p*-values which exhibited an average of 1.1×10^{-4} among the medians of all the subsets. The values listed above include weekday and weekend subsets.

When subsetting for SMP and day type, the RoA were, on average (median), 0.66–1.1 (0.60–1.0) on weekdays and 0.71–2.0 (0.52–1.1) on weekends. The distribution of the RoA across the SMPs on weekdays is shown in Figure 1. The median of the RoA on weekdays was above unity under no SMP, being equal to 1.0 for SMPs 1 and 5. A similar analysis for the RoM shows that the average (median) was 0.69–1.3 (0.58–1.1) on weekdays and 0.68–1.9 (0.52–1.1) on weekends for the different SMPs.

The considerations above show that there was a statistically significant (median *p*-values across the SMPs between 1.7×10^{-13} and 7.7×10^{-4} on weekdays and between 4.0×10^{-7} and 3.4×10^{-2} on weekends) reduction in benzene average concentrations during the lockdowns for a majority of monitoring stations with a median reduction in benzene average concentrations on weekdays of up to 34% across the SMPs.

The SMPs under which average benzene concentrations were the lowest during the lockdown relative to no-lockdown conditions were SMPs 6 and 7 and are related to moderate continental (E or S) flows (see Table 5). For these SMPs, the RoA cumulative distribution function (CDF) crosses the RoA = 1 (vertical dashed line in the plots) line at percentiles 82 and 89, respectively (weekdays). The CDF crosses the median line (horizontal dashed line in the plots) at a ratio of 0.68 and 0.60, translating into a median reduction of 32% and 40% in the average benzene concentrations across all stations under lockdown conditions for SMP 6 and 7, respectively. Benzene concentrations under no-lockdown conditions during days corresponding to SMPs 6–7 were 0.86 ± 0.67 and $0.85 \pm 0.73 \mu\text{g m}^{-3}$ (average \pm standard deviation throughout the station averages), respectively. During the lockdown, average concentrations were down to 0.62 ± 0.80 and $0.46 \pm 0.39 \mu\text{g m}^{-3}$.

Table 5

Benzene Concentrations ($\mu\text{g m}^{-3}$, Average \pm Standard Deviation of the Averages Throughout All Stations) During the 2020 Lockdowns and in the Same Period of the Four Previous Years, Average and Median Reduction $\left(\frac{\text{lockdown} - \text{no lockdown}}{\text{no lockdown}}\right)$ Differentiated by SMP (Weekdays Only)

		SMP1	SMP2	SMP3	SMP4	SMP5	SMP6	SMP7	SMP8
All stations	$[C_6H_6]_{\text{lockdown}}$	1 ± 0.7	0.8 ± 0.7	0.83 ± 0.93	0.55 ± 0.55	0.78 ± 0.61	0.62 ± 0.8	0.46 ± 0.39	0.76 ± 0.67
	$[C_6H_6]_{\text{no lockdown}}$	1.03 ± 0.84	1.13 ± 1.41	1.01 ± 0.85	0.79 ± 0.72	0.82 ± 0.74	0.86 ± 0.67	0.85 ± 0.73	0.92 ± 0.76
	median reduction	0	0.22	0.23	0.24	-0.02	0.32	0.4	0.16
	average reduction	-0.14	-0.04	0.11	0.12	-0.12	0.21	0.34	0.08
Traffic stations	$[C_6H_6]_{\text{lockdown}}$	0.97 ± 0.56	0.75 ± 0.64	0.69 ± 0.55	0.53 ± 0.59	0.71 ± 0.61	0.61 ± 1.07	0.45 ± 0.36	0.68 ± 0.56
	$[C_6H_6]_{\text{no lockdown}}$	1.14 ± 0.87	1.14 ± 0.9	1.07 ± 0.86	0.89 ± 0.79	0.88 ± 0.77	0.98 ± 0.78	0.95 ± 0.81	0.99 ± 0.75
	median reduction	0.08	0.29	0.33	0.4	0.16	0.44	0.49	0.3
	average reduction	0.03	-0.03	0.28	0.32	0.05	0.41	0.46	0.21
Industrial stations	$[C_6H_6]_{\text{lockdown}}$	1.12 ± 0.99	0.92 ± 0.88	1.04 ± 1.49	0.81 ± 0.75	0.9 ± 0.63	0.78 ± 0.83	0.66 ± 0.54	0.91 ± 0.9
	$[C_6H_6]_{\text{no lockdown}}$	1.01 ± 0.73	1.2 ± 1.55	0.97 ± 0.67	0.75 ± 0.51	0.8 ± 0.58	0.83 ± 0.54	0.81 ± 0.54	0.94 ± 0.65
	median reduction	-0.14	0.17	0.13	0.05	-0.1	0.21	0.25	0.12
	average reduction	-0.25	-0.14	-0.09	-0.21	-0.24	0.08	0.12	0
Background stations	$[C_6H_6]_{\text{lockdown}}$	0.99 ± 0.72	0.8 ± 0.65	0.87 ± 0.87	0.45 ± 0.29	0.8 ± 0.6	0.56 ± 0.42	0.36 ± 0.25	0.76 ± 0.65
	$[C_6H_6]_{\text{no lockdown}}$	0.96 ± 0.86	1.09 ± 1.69	0.99 ± 0.9	0.72 ± 0.72	0.77 ± 0.76	0.76 ± 0.6	0.78 ± 0.72	0.86 ± 0.81
	median reduction	-0.03	0.12	0.13	0.11	-0.05	0.26	0.4	0.05
	average reduction	-0.28	-0.02	0.05	0.06	-0.23	0.09	0.33	-0.01
Urban stations	$[C_6H_6]_{\text{lockdown}}$	1 ± 0.67	0.81 ± 0.69	0.82 ± 0.79	0.5 ± 0.45	0.79 ± 0.65	0.61 ± 0.86	0.42 ± 0.31	0.76 ± 0.66
	$[C_6H_6]_{\text{no lockdown}}$	1.14 ± 0.93	1.21 ± 1.46	1.09 ± 0.94	0.87 ± 0.79	0.88 ± 0.82	0.92 ± 0.74	0.93 ± 0.81	1 ± 0.83
	median reduction	0.03	0.28	0.29	0.37	0.08	0.41	0.48	0.23
	average reduction	-0.09	0.02	0.19	0.26	-0.06	0.26	0.41	0.13
Suburban stations	$[C_6H_6]_{\text{lockdown}}$	1.13 ± 0.9	0.91 ± 0.8	1.03 ± 1.43	0.77 ± 0.8	0.85 ± 0.52	0.75 ± 0.74	0.62 ± 0.55	0.85 ± 0.79
	$[C_6H_6]_{\text{no lockdown}}$	0.87 ± 0.53	1.08 ± 1.47	0.9 ± 0.54	0.67 ± 0.43	0.73 ± 0.47	0.79 ± 0.44	0.71 ± 0.41	0.85 ± 0.56
	median reduction	-0.15	0.09	0.07	0.05	-0.13	0.19	0.3	0.09
	average reduction	-0.3	-0.27	-0.17	-0.28	-0.32	0.09	0.15	-0.09
Rural stations	$[C_6H_6]_{\text{lockdown}}$	0.75 ± 0.41	0.54 ± 0.33	0.52 ± 0.33	0.43 ± 0.27	0.55 ± 0.32	0.41 ± 0.21	0.33 ± 0.2	0.48 ± 0.27
	$[C_6H_6]_{\text{no lockdown}}$	0.56 ± 0.34	0.59 ± 0.4	0.59 ± 0.36	0.42 ± 0.24	0.47 ± 0.23	0.5 ± 0.25	0.49 ± 0.26	0.49 ± 0.27
	median reduction	-0.26	0.09	0.09	-0.03	-0.16	0.19	0.38	0.07
	average reduction	-0.22	-0.07	0.1	-0.09	-0.2	0.13	0.31	0.01

The percentile value where the RoA CDF crosses the RoA = 1 line were at lowest for SMPs 1 and 5: 51% and 48%, respectively (weekdays). SMPs 1 (fast W flows) and 5 (moderate N-NE flows) represent conditions which favor dispersion and/or bring cleaner maritime air. Such conditions diminish the relative importance of local sources and indeed the average reduction in average benzene concentrations was -13% and -12% for SMP1 and SMP5, respectively, whereas the median reduction across the stations was 0% and -2% for SMP1 and SMP5, respectively, indicating a light increase for a majority of stations. Under these meteorological conditions, average benzene concentrations were (no-lockdown) 1.0 ± 0.84 and $0.82 \pm 0.74 \mu\text{g m}^{-3}$ for SMPs 1 and 5, respectively. Under lockdown conditions, those were 1.0 ± 0.70 and $0.78 \pm 0.61 \mu\text{g m}^{-3}$.

The RoA CDF crosses the RoA=1 line at intermediate percentile values for SMPs 2, 3, 4 and 8: percentiles 67–73. Under SMPs 2, 3 and 4, associated with slow flows (SE-SW, NW and W, respectively) which do not favor dispersion, the average (median) reductions in the average benzene concentrations across stations and under lockdown conditions were -4%–12% (22%–24%). SMP8 matches dispersion favoring conditions (fast NW-N flows) but its CDF does not cluster with SMPs 1 and 5 but with SMPs 2, 3, and 4 instead. The average (median) reductions in the average benzene concentrations across stations on weekdays under SMP8 was 8% (16%).

Because of the atmospheric reactivity of benzene and of the distance from the emission sources, the identified general behavior may be distinct when considering stations of different types and/or areas. At traffic stations (1,761 subsets from 128 stations), the RoA indicates a clear decrease in benzene concentrations under lockdown conditions, exhibiting the 75th percentile below or equal to unity for all SMPs except for SMPs 1 and 5 (Figure S57 in Supporting Information S1). Even for those two SMPs, the 50th percentile of the RoA was <1. On weekdays, the average (median) reduction in benzene average concentrations at traffic sites was 22% (32%). Moving from traffic-type environments toward industrial (839 subsets from 62 stations) or background (1,826 subsets from 148 stations) stations diminishes the decrease in benzene average concentrations: average (median) weekdays reductions of −8% and 0% (10% and 12%), respectively. A negative reduction indicates an increase in concentration.

Analyzing the RoA distribution subsetting the monitoring stations by *area* (Figure S58 in Supporting Information S1) allows for a clearer trend recognition. At urban stations (3,158 subsets from 242 stations), the distribution of the RoA is very similar to that at traffic stations with reductions in benzene average concentrations (albeit very diminished for SMPs 1 and 5) for a majority of stations. Reductions of Benzene weekdays average concentrations were 14% (28%) in average (median). Moving away from the city, into suburban (909 subsets from 68 stations) and then rural (359 subsets from 28 stations) areas, shifts the distribution of the RoA toward larger values (CDF curves move toward the right) with the proportions of the distribution above unity increasing with the distance from the city as well as the median reduction values. In average (median), the reduction in benzene average concentrations was −14% and 0% (8% and 6%) at suburban and rural stations. There was an increase in average benzene concentrations at a majority of suburban and rural stations under SMPs 1, 4, and 5.

A decrease of ~10% in average benzene concentration (against the concentrations in periods immediately after and before) at a urban traffic site in the Pearl River Delta (PRD) was reported by Pei et al. (2022), which is in-line, though of a lesser magnitude, than the data presented here (weighted average reduction of 14% at urban stations). In an important Romanian center for petroleum refining and associated product manufacturing, Sanda et al. (2023) found benzene concentrations ($2.67 \mu\text{g m}^{-3}$) during the lockdown lower than the annual averages of 2019–2021 ($3.41\text{--}3.63 \mu\text{g m}^{-3}$).

3.3. Toluene

The concentrations of toluene (typical atmospheric lifetime of about 2 days), a strong marker for traffic exhaust but also an additive to solvents in paints, coatings, fragrances and cleaning products, among others (Barletta et al., 2008; Baudic et al., 2016; Buzcu & Fraser, 2006; Gentner et al., 2009; Sauvage et al., 2009), from 54 stations throughout 9 countries were reported to Airbase for the 2020 lockdown periods and its corresponding dates in the 4 years prior with a collection efficiency larger than the threshold and passing our QA/QC procedure. There were 580 subsets resulting from the combinations between SMPs and day types.

Figure S54 in Supporting Information S1 shows the toluene RoA by station over Europe. The distribution of the RoA across the SMPs on weekdays is shown in Figure 2. When subsetting for SMP and day type (Table 6), but leaving SMP 1 out of this analysis for the low number of subsets (2), the RoA were, on average (median), 0.45–1.2 (0.43–0.87) on weekdays. On weekends, leaving SMPs 1 and 3 out (0 and 12 subsets, respectively), the RoA average (median) were 0.49–1.0 (0.44–0.84). No subset exhibited a median RoA equal or above 1.

At monitoring stations of *type* Traffic (206 subsets from 18 stations) median RoA were in the range 0.44–0.83 (0.43–0.87 when all station *types* considered) on weekdays, and the CDFs for the different SMPs crossed the RoA = 1 line at percentiles 79–88 (67–99 when all station *types* considered), evidencing an almost-ubiquitous reduction in toluene concentrations (average and median reduction of 33% and 36%, respectively, at traffic sites and 16% and 32% at all sites). Across SMPs, a trend consistent throughout *types* and *areas* emerges (Figure 2): reductions were more important under SMP 6 (moderate E flows) less important under SMPs 7 and 8 (moderate S flows and fast NW-N flows, respectively) and intermediate under SMPs 2–4 and 5 (slow flows and moderate N-NE flows, respectively). Despite the consistency, there is no similarity between SMPs that are analogous in terms of dispersion conditions. However, the low number of stations and subsets for some station *types* or *areas* limits the trend analysis: 18 (206), 6 (64), and 30 (310) stations (subsets) for stations of *types* traffic, industrial, and background, respectively, and 31 (327), 14 (158), and 9 (95) for stations of *areas* urban, suburban, and rural, respectively.

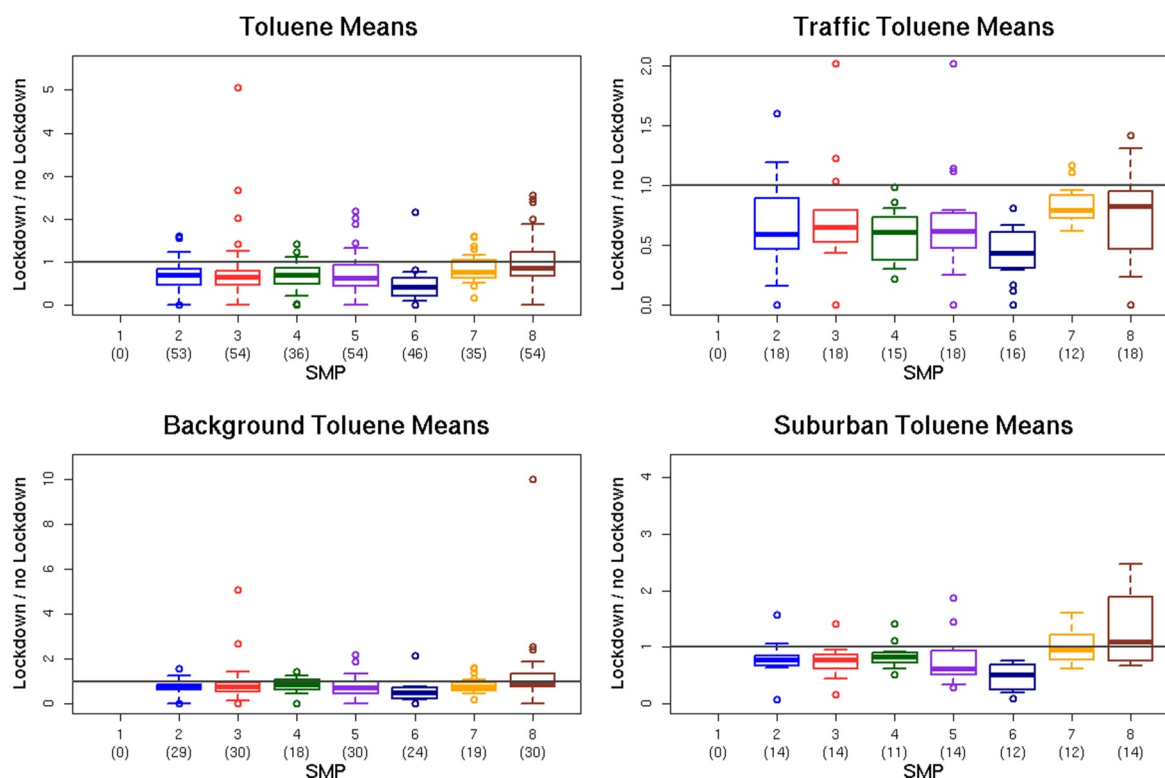


Figure 2. Box and whisker plots of the RoA for Toluene on weekdays differentiated for the eight SMPs for all monitoring stations (upper left) for Traffic and Background stations *types* (upper right and lower left, respectively, and for station *area* Suburban). The box and whiskers represent, from bottom to top, the 5th, 25th, 50th, 75th, and 95th percentiles, the individual points are either below the 5th or above the 95th percentiles. Station *type* Industrial and station *area* Rural are not shown because of the low number (<10 for all combinations) of subsets. Station *area* Urban is not shown for being very similar to station *type* Traffic.

Low *p*-values of the Wilcoxon-Mann-Whitney *U*-tests (medians in the range 4.2×10^{-8} – 2.2×10^{-1} through the SMPs on weekdays and 6.8×10^{-5} – 1.3×10^{-1} on weekends) indicate that the reductions were statistically significant for a majority of stations.

In the PRD, average toluene concentration decreased (against the concentrations in periods immediately after and before) by ~35% (Pei et al., 2022), a value larger to that experienced in Europe (this study, weighted average: 17% at weekdays). Here, the comparison is to the average; however, we have generally focused on medians across stations and the median change is 33% (weekdays).

3.4. Xylenes

m,p-Xylenes and o-xylene were measured in 33 and 30 monitoring stations from 5 and 3 countries, respectively. The RoA for both species are mapped in Figures S55 and S56 in Supporting Information S1. These species have been associated with traffic tailpipe emissions, other combustion sources, unburnt gasoline (misfiring or malfunction), or solvent use and have a typical atmospheric lifetime of about one day (e.g., Abeleira et al., 2017; Baudic et al., 2016; Buzcu & Fraser, 2006; Gentner et al., 2009; Nelson & Quigley, 1984; Sauvage et al., 2009).

In total, there were 349 subsets for m,p-Xylenes. m,p-Xylenes was measured more at stations of *type* Background (241 subsets) and less at stations of *type* Traffic (86 subsets) and Industrial (22 subsets). The *area* of the stations were mostly Urban or Suburban (138 and 145 subsets, respectively) and less rural (66 subsets).

In total, there were 327 subsets for o-xylene. o-Xylene was measured more at stations of *type* Background (243 subsets) and less at stations of *type* Traffic and Industrial (62 and 22 subsets, respectively). The *area* of the stations were mostly Suburban (158 subsets) and less urban and rural (114 and 55 subset, respectively).

Figure 3 shows the RoA for xylenes throughout all stations. The distribution of the RoA shows that reductions were more important for m,p-Xylenes than for o-xylene (median reductions through the SMPs of –4%–57% and

Table 6

Toluene Concentrations ($\mu\text{g m}^{-3}$, Average \pm Standard Deviation of the Averages Throughout All Stations) During the 2020 Lockdowns and in the Same Period of the Four Previous Years, Average and Median Reduction $\left(\frac{\text{lockdown} - \text{no lockdown}}{\text{no lockdown}}\right)$ Differentiated by SMP (Weekdays Only)

		SMP1	SMP2	SMP3	SMP4	SMP5	SMP6	SMP7	SMP8
All stations	$[C_6H_5 - CH_3]_{\text{lockdown}}$	0.02 ± 0.02	1.78 ± 2.15	1.72 ± 2.19	1.62 ± 2.39	1.4 ± 2.62	1.32 ± 3.64	1.07 ± 0.72	1.7 ± 2.21
	$[C_6H_5 - CH_3]_{\text{no lockdown}}$	1.91 ± 2.76	2.85 ± 3.32	2.45 ± 3.2	2 ± 2.83	2.04 ± 2.9	1.89 ± 2.66	1.98 ± 2.64	2.04 ± 2.81
	median reduction	0.8	0.3	0.36	0.31	0.37	0.59	0.23	0.13
	average reduction	0.8	0.13	-0.15	0.37	0.31	0.57	0.19	-0.07
Traffic stations	$[C_6H_5 - CH_3]_{\text{lockdown}}$	-	2.79 ± 2.39	2.52 ± 2.57	2.24 ± 2.76	1.94 ± 2.5	1.54 ± 2.06	1.74 ± 0.41	2.53 ± 2.55
	$[C_6H_5 - CH_3]_{\text{no lockdown}}$	3.2 ± 3.35	4.37 ± 3.79	3.62 ± 3.76	3.36 ± 3.08	3.03 ± 3.52	3.15 ± 3.51	3.13 ± 3.14	3.44 ± 3.35
	median reduction	-	0.4	0.35	0.39	0.38	0.56	0.2	0.17
	average reduction	-	0.33	0.27	0.42	0.32	0.57	0.16	0.22
Industrial stations	$[C_6H_5 - CH_3]_{\text{lockdown}}$	-	0.79 ± 0.93	0.69 ± 0.55	0.76 ± 0.19	0.62 ± 0.53	0.21 ± 0.18	1.18 ± 0.47	1.41 ± 1.69
	$[C_6H_5 - CH_3]_{\text{no lockdown}}$	1.04 ± 0.26	2.39 ± 1.22	2.38 ± 1.95	0.98 ± 0.46	1.09 ± 1.12	1.07 ± 0.66	1.04 ± 0.25	1.06 ± 0.52
	median reduction	-	0.57	0.7	0.23	0.5	0.8	-0.06	0.22
	average reduction	-	0.59	0.67	0.29	0.42	0.79	-0.08	-0.01
Background stations	$[C_6H_5 - CH_3]_{\text{lockdown}}$	0.02 ± 0.02	1.39 ± 1.99	1.47 ± 2.05	1.26 ± 2.22	1.23 ± 2.91	1.44 ± 4.72	0.64 ± 0.59	1.27 ± 2.01
	$[C_6H_5 - CH_3]_{\text{no lockdown}}$	1.32 ± 2.41	2.03 ± 3.05	1.79 ± 2.91	1.4 ± 2.72	1.65 ± 2.65	1.35 ± 2.12	1.49 ± 2.4	1.41 ± 2.45
	median reduction	0.8	0.25	0.25	0.18	0.36	0.52	0.27	0.13
	average reduction	0.8	-0.09	-0.55	0.34	0.27	0.53	0.26	-0.25
Urban stations	$[C_6H_5 - CH_3]_{\text{lockdown}}$	0 ± 0	2.52 ± 2.54	2.46 ± 2.62	2.36 ± 2.96	2.1 ± 3.33	2.22 ± 4.84	1.45 ± 0.59	2.32 ± 2.66
	$[C_6H_5 - CH_3]_{\text{no lockdown}}$	2.91 ± 3.34	4.15 ± 3.89	3.46 ± 3.84	3.04 ± 3.42	3.02 ± 3.54	2.79 ± 3.29	2.99 ± 3.16	3.02 ± 3.43
	median reduction	1	0.4	0.36	0.39	0.4	0.59	0.24	0.17
	average reduction	1	0.05	-0.42	0.4	0.32	0.53	0.19	-0.12
Suburban stations	$[C_6H_5 - CH_3]_{\text{lockdown}}$	0.03 ± 0	1.21 ± 0.87	1.14 ± 0.81	0.8 ± 0.44	0.65 ± 0.38	0.41 ± 0.26	0.98 ± 0.66	1.3 ± 1.11
	$[C_6H_5 - CH_3]_{\text{no lockdown}}$	0.82 ± 0.48	1.71 ± 1.2	1.59 ± 1.42	0.91 ± 0.45	1 ± 0.73	0.97 ± 0.55	0.88 ± 0.39	1 ± 0.47
	median reduction	0.61	0.21	0.22	0.17	0.4	0.54	0.1	-0.09
	average reduction	0.61	0.16	-0.02	0.35	0.29	0.61	0.11	-0.2
Rural stations	$[C_6H_5 - CH_3]_{\text{lockdown}}$	-	0.21 ± 0.2	0.16 ± 0.13	0.16 ± 0.12	0.22 ± 0.17	0.11 ± 0.08	0.17 ± 0.08	0.21 ± 0.16
	$[C_6H_5 - CH_3]_{\text{no lockdown}}$	0.25 ± 0.12	0.41 ± 0.44	0.42 ± 0.27	0.21 ± 0.08	0.37 ± 0.28	0.41 ± 0.35	0.33 ± 0.19	0.4 ± 0.35
	median reduction	-	0.27	0.62	0.07	0.15	0.61	0.4	0.15
	average reduction	-	0.32	0.59	0.26	0.28	0.65	0.36	0.34

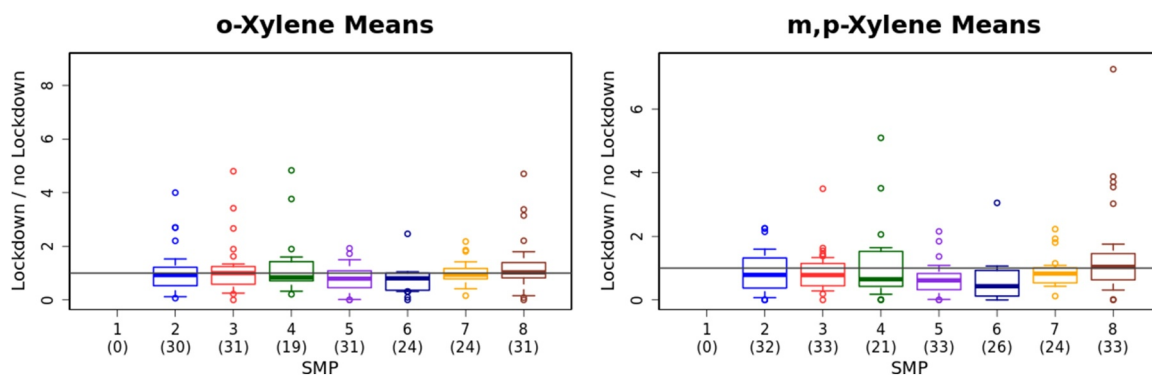


Figure 3. Box and whisker plots of the RoA for Xylenes on weekdays differentiated for the eight SMPs for all monitoring stations (left: o-Xylene, right: m,p-Xylenes). The box and whiskers represent, from bottom to top, the 5th, 25th, 50th, 75th, and 95th percentiles, the individual points are either below the 5th or above the 95th percentiles.

Table 7

Xylenes Concentrations ($\mu\text{g m}^{-3}$, Average \pm Standard Deviation of the Averages Throughout All Stations) During the 2020 Lockdowns and in the Same Period of the Four Previous Years, Average and Median Reduction $\left(\frac{\text{lockdown} - \text{no lockdown}}{\text{no lockdown}}\right)$ Differentiated by SMP (Weekdays Only)

		SMP1	SMP2	SMP3	SMP4	SMP5	SMP6	SMP7	SMP8
All stations	$[o - C_6H_4 - (CH_3)_2]_{\text{lockdown}}$	0 ± 0	0.27 ± 0.25	0.27 ± 0.22	0.27 ± 0.32	0.22 ± 0.5	0.15 ± 0.13	0.23 ± 0.2	0.3 ± 0.38
	$[o - C_6H_4 - (CH_3)_2]_{\text{no lockdown}}$	0.22 ± 0.18	0.4 ± 0.37	0.36 ± 0.37	0.26 ± 0.26	0.27 ± 0.27	0.25 ± 0.22	0.27 ± 0.2	0.27 ± 0.25
	median reduction	1	0.07	0	0.16	0.21	0.19	0.06	-0.03
	average reduction	1	-0.06	-0.13	-0.27	-0.17	0.27	-0.01	-0.69
All stations	$[m,p - C_6H_4 - (CH_3)_2]_{\text{lockdown}}$	0 ± 0	0.92 ± 1.05	0.82 ± 0.78	0.69 ± 0.84	0.6 ± 1.24	0.33 ± 0.37	0.64 ± 0.67	0.85 ± 0.95
	$[m,p - C_6H_4 - (CH_3)_2]_{\text{no lockdown}}$	0.72 ± 0.79	1.37 ± 1.39	1.11 ± 1.01	0.81 ± 0.79	1.02 ± 1.26	0.79 ± 0.63	0.92 ± 0.83	0.83 ± 0.8
	median reduction	1	0.21	0.22	0.35	0.38	0.57	0.17	-0.04
	average reduction	1	-0.08	0.11	-0.14	0.01	0.39	0.11	-0.42

-3%–21% on weekdays, Table 7). Xylene concentrations reductions during lockdowns relative to no lockdown conditions were more pronounced under SMPs 5 (moderate N-NE flows) and 6 (moderate E flows) with average (median) weekdays reductions of 1/–17% (38/21%) and 39/27% (57/19%), respectively (m,p-Xylenes/o-xylene). SMPs associated with slow flows (2, 3, and 4) exhibited a RoA distribution centered around 1 (average (median) weekdays reductions between -14% and 11% (21% and 35%) and -27% and -6% (0% and 16%) for m,p-Xylenes and o-xylene, respectively). SMP 8 (fast NW-N flows) followed a behavior close to that of the slow flow SMPs (average weekdays (median) reduction of -42% (-4%) and -69% (-3%) for m,p-Xylenes and o-xylene, respectively) and evidenced neither a clear reduction nor a clear increase. RoA distribution under SMP 7 (moderate W flows, high stability) displayed some degree of reduction (11% (17%) and -1% (6%) as average (median) on weekdays for m,p-Xylenes and o-xylene, respectively) although not as pronounced as for SMPs 5 and 6. The general trend is replicated for stations of *type* Background (Figure S60 in Supporting Information S1) and *area* Urban and Suburban (Figure S61 in Supporting Information S1) (Table 8).

Despite the changes in averages and medians, those must be regarded with caution as large *p*-values of the Wilcoxon-Mann-Whiney *U*-tests (medians in the range 0.1–0.5 for both VOCs) indicate that H_0 (the distribution of concentrations through the SMPs and day types were equal before and during the lockdowns) cannot be rejected in a majority of stations.

Pei et al. (2022) reported a ~50% reduction in xylenes at traffic sites in the PRD. For the present study, the change (weighted average) for this study was 6% and 22% increase for m,p-Xylenes and o-xylene, respectively with a

Table 8

Ethylbenzene and Trimethylbenzenes Concentrations ($\mu\text{g m}^{-3}$, Average \pm Standard Deviation of the Averages Throughout All Stations) During the 2020 Lockdowns and in the Same Period of the Four Previous Years, Average and Median Reduction $\left(\frac{\text{lockdown} - \text{no lockdown}}{\text{no lockdown}}\right)$ Differentiated by SMP (Weekdays Only)

		SMP1	SMP2	SMP3	SMP4	SMP5	SMP6	SMP7	SMP8
All stations	$[1,2,4 - C_6H_3(CH_3)_3]_{\text{lockdown}}$	-	0.18 ± 0.12	0.18 ± 0.14	0.16 ± 0.08	0.37 ± 0.99	0.12 ± 0.04	0.16 ± 0.12	0.27 ± 0.48
	$[1,2,4 - C_6H_3(CH_3)_3]_{\text{no lockdown}}$	0.15 ± 0.09	0.35 ± 0.49	0.25 ± 0.2	0.16 ± 0.07	0.2 ± 0.14	0.24 ± 0.22	0.19 ± 0.12	0.17 ± 0.09
	median reduction	-	0	0	0	0.3	0.3	0.18	0
	average reduction	-	-0.12	0.14	0	-0.25	0.31	0.07	-0.26
All stations	$[1,3,5 - C_6H_3(CH_3)_3]_{\text{lockdown}}$	-	0.09 ± 0.01	0.09 ± 0.03	0.08 ± 0.02	0.14 ± 0.25	0.07 ± 0.03	0.08 ± 0.03	0.11 ± 0.11
	$[1,3,5 - C_6H_3(CH_3)_3]_{\text{no lockdown}}$	0.08 ± 0.03	0.11 ± 0.12	0.09 ± 0.05	0.08 ± 0.03	0.08 ± 0.03	0.09 ± 0.05	0.08 ± 0.03	0.08 ± 0.02
	median reduction	-	0	-0.09	0	0.03	0.01	0	0
	average reduction	-	-0.37	-0.07	-0.25	-0.4	0.14	0.03	-0.2
All stations	$[C_6H_5 - C_2H_5]_{\text{lockdown}}$	0 ± 0	0.29 ± 0.32	0.27 ± 0.21	0.29 ± 0.33	0.22 ± 0.5	0.16 ± 0.13	0.24 ± 0.24	0.28 ± 0.33
	$[C_6H_5 - C_2H_5]_{\text{no lockdown}}$	0.24 ± 0.25	0.47 ± 0.45	0.41 ± 0.41	0.27 ± 0.27	0.3 ± 0.27	0.25 ± 0.23	0.29 ± 0.24	0.28 ± 0.23
	median reduction	1	0	0.08	0.03	0.3	0.19	0.07	-0.01
	average reduction	1	0.03	0.06	-0.18	0	0.21	0.06	-0.15

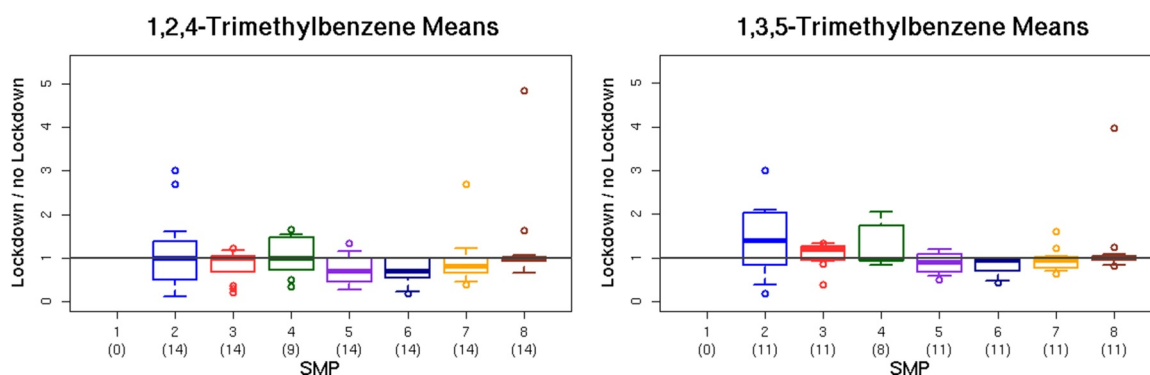


Figure 4. Box and whisker plots of the RoA for 1,2,4-Trimethylbenzene and 1,3,5-Trimethylbenzene on weekdays differentiated for the eight SMPs for all monitoring stations. The box and whiskers represent, from bottom to top, the 5th, 25th, 50th, 75th, and 95th percentiles, the individual points are either below the 5th or above the 95th percentiles.

large variability through the subsets: between 50% increase and 100% decrease for m,p-Xylenes and between 77% increase and 100% decrease for o-xylene.

3.5. Trimethylbenzenes and Ethylbenzene

Ethylbenzene is emitted by road transport (e.g., evaporation of diesel) but can also be associated with combustion or industrial emissions (Borbon et al., 2018; Ding et al., 2020; Kumar et al., 2020) and has an atmospheric lifetime in the order of several hours to a few days (Monod et al., 2001), whereas C9 aromatics are associated to industrial activities or traffic (An et al., 2014; Languille et al., 2020).

1,2,4-trimethylbenzene and 1,3,5-trimethylbenzene from 14 and 11 stations, respectively, in a single country were reported to Airbase for the periods of interest and considered for this study. 1,2,4-trimethylbenzene (162 subsets, Figure 4 and Table 8) and 1,3,5-trimethylbenzene (129 subsets) exhibit a similar behavior, throughout the SMPs, as xylenes (Figure 6): more significant reductions under moderate N-NE and E flows (SMPs 5 and 6), no clear reductions under slow or fast NW-N flows (SMPs 2, 3, 4, and 8) and intermediate reductions under high stability conditions (SMP 7).

Another VOC exhibiting a similar pattern through the SMPs, albeit with higher dispersion of RoA values, as xylenes is ethylbenzene (Figure 6). Ethylbenzene concentrations from 30 monitoring stations (4 countries, 306 subsets) were reported to Airbase for the periods of interest and with the sufficient data coverage to be considered for this study. Most data came from background stations (235 subsets, 23 stations).

The general trend followed by ethylbenzene shows a decrease during the lockdown (Figure 5): in average the change for all stations corresponded to a decrease of 1% (range between -18% and 21% reduction on weekdays for the different SMPs with more than one subset), although the median of the changes indicates a decrease for most stations (10% average of medians across SMPs, range between -1% and 30%, on weekdays for the different SMPs with more than one subset). A decrease in ethylbenzene concentrations (~45%) was also reported by Pei et al. (2022) for the PRD (traffic site) under lockdown conditions compared to periods immediately after and before.

Although changes were identified, the distributions of the concentration of 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene, and ethylbenzene during the lockdowns were not statistically different from the distribution of the concentration before the lockdowns (medians of the *p*-values of the Wilcoxon-Mann-Whiney *U*-tests in the ranges 0.2–0.6, 0.2–0.8 and 0.1–0.7, respectively) for a majority of stations.

3.6. *n*-Alkanes

Butane, pentane, and hexane were measured at 10, 18, and 18 monitoring stations (110, 214, and 214 subsets), respectively, from a single country. Atmospheric light alkanes (C2–C4) are representative of oil and natural gas operations, whereas heavier compounds (C4–C6) are important contributors in the evaporation products of gasoline or liquid petroleum gas and solvents (e.g., Baudic et al., 2016; Gilman et al., 2013; Pétron et al., 2012;

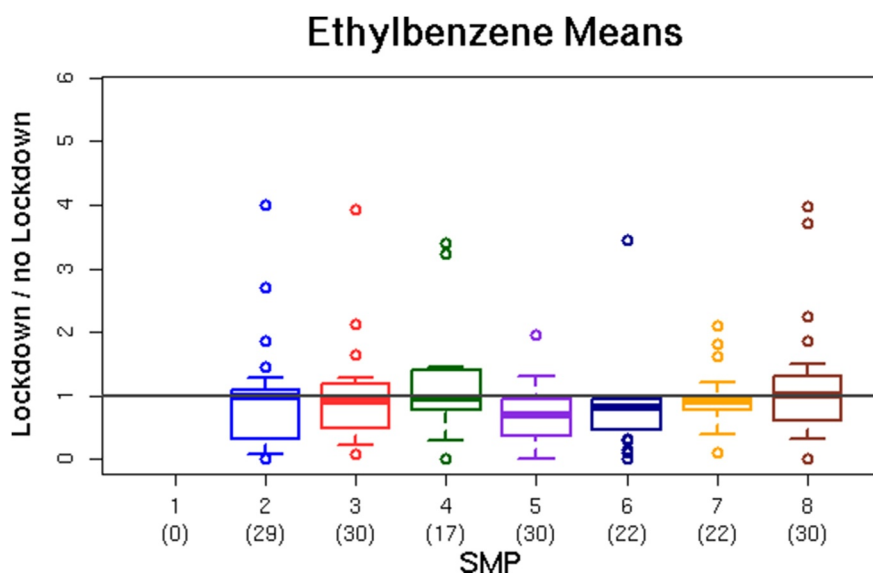


Figure 5. Box and whisker plots of the RoA for Ethylbenzene on weekdays differentiated for the eight SMPs for all monitoring stations. The box and whiskers represent, from bottom to top, the 5th, 25th, 50th, 75th, and 95th percentiles, the individual points are either below the 5th or above the 95th percentiles.

Rossabi et al., 2021). These long-lived compounds have not been associated with combustion sources, although pentane is emitted by gasoline combustion in very small amounts (e.g., Abeleira et al., 2017; Barletta et al., 2005; Yuan et al., 2012).

Median and average reductions for the three are shown in Table 9.

Butane average concentrations were lowered during the lockdowns by 7% in average (13% median) on weekdays. At the SMP level and on weekdays, SMPs 2, 3, 6 and 7 exhibited a clear decrease (average reduction in the range 5%–35%, median 18%–34%), whereas average butane concentrations increased under SMPs 4 and 8 (average increases of 19% and 16%, respectively, median of 13% and 8%). Under SMP 5, the change was less conclusive: 2% increase in average, 9% decrease in median on weekdays.

For hexane, the average concentration was reduced for a majority of stations under all SMPs except SMP4 (median reductions in the range 10%–19% on weekdays). The reduction was the lowest for SMP8 (10% median,

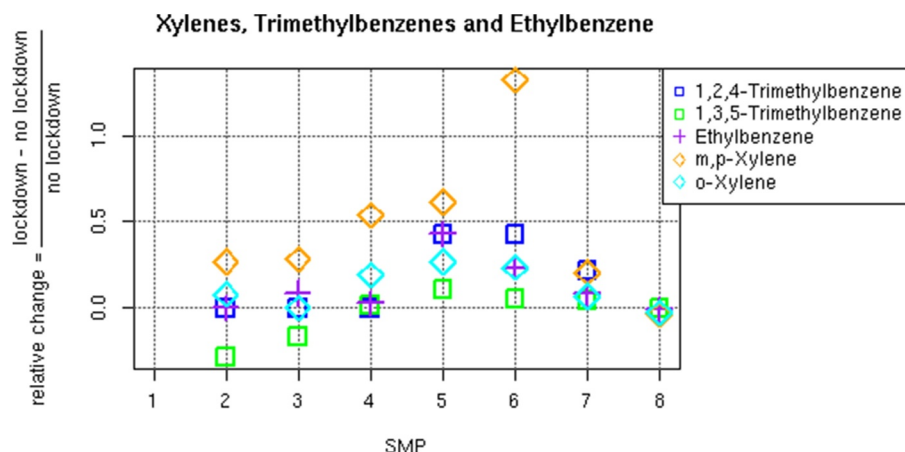


Figure 6. Median reduction through the SMPs for Xylenes, Trimethylbenzenes, and Ethylbenzene on weekdays. SMP 1 is left out for having, at most, only one subset.

Table 9

n-Alkanes Concentrations ($\mu\text{g m}^{-3}$, Average \pm Standard Deviation of the Averages Throughout All Stations) During the 2020 Lockdowns and in the Same Period of the Four Previous Years, Average and Median Reduction $\left(\frac{\text{lockdown} - \text{no lockdown}}{\text{no lockdown}}\right)$ Differentiated by SMP (Weekdays Only)

	SMP1	SMP2	SMP3	SMP4	SMP5	SMP6	SMP7	SMP8
All stations $[\text{CH}_3 - \text{CH}_2 - \text{CH}_2 - \text{CH}_3]_{\text{lockdown}}$	–	0.91 ± 0.65	1.32 ± 1.25	1 ± 0.45	1.48 ± 1.88	1.19 ± 1.65	0.72 ± 0.59	1.47 ± 1.22
$[\text{CH}_3 - \text{CH}_2 - \text{CH}_2 - \text{CH}_3]_{\text{notlockdown}}$	1.06 ± 1.28	1.15 ± 1.04	1.51 ± 1.31	0.82 ± 0.51	1.28 ± 1.1	1.3 ± 1.47	1.24 ± 0.93	1.27 ± 1.07
median reduction	–	0.18	0.2	–0.13	0.09	0.21	0.34	–0.08
average reduction	–	0.05	0.13	–0.19	–0.02	0.17	0.35	–0.16
All stations $[\text{CH}_3 - (\text{CH}_2)_3 - \text{CH}_3]_{\text{lockdown}}$	–	0.87 ± 1	1.1 ± 1.37	1.38 ± 1.56	1.02 ± 1.17	0.86 ± 1.3	0.83 ± 1.25	0.96 ± 1.11
$[\text{CH}_3 - (\text{CH}_2)_3 - \text{CH}_3]_{\text{notlockdown}}$	0.43 ± 0.39	0.63 ± 0.65	0.59 ± 0.43	0.43 ± 0.38	0.57 ± 0.43	0.7 ± 0.71	0.58 ± 0.55	0.56 ± 0.43
median reduction	–	0.1	–0.04	–0.98	–0.1	0.23	0.08	–0.1
average reduction	–	–0.44	–0.8	–1.41	–0.69	–0.19	–0.2	–0.78
All stations $[\text{C}_6\text{H}_{14}]_{\text{lockdown}}$	–	0.37 ± 0.36	0.33 ± 0.26	1.15 ± 2.78	0.42 ± 0.64	0.52 ± 1.13	0.63 ± 1.52	0.48 ± 0.64
$[\text{C}_6\text{H}_{14}]_{\text{notlockdown}}$	0.28 ± 0.29	0.44 ± 0.39	0.38 ± 0.36	0.37 ± 0.45	0.33 ± 0.26	0.56 ± 1.05	0.46 ± 0.63	0.38 ± 0.4
median reduction	–	0.18	0.16	–0.03	0.17	0.19	0.12	0.1
average reduction	–	–0.15	–0.18	–0.77	–0.39	–0.04	–0.08	–0.58

weekdays) and there was an increase under SMP4 (3% median, weekdays), the SMPs with a clear increase in butane.

Pentane showed similarities with butane and hexane (Figure 7): clear increases under SMPs 4, 8, and 5 (weekdays medians 10%–98%) and decreases for SMPs 2, 6, and 7 (weekdays medians 8%–23%). SMP3 was less conclusive: median increase of 4%.

The median of the *p*-values of the Wilcoxon-Mann-Whiney *U*-tests, in the range 0.4–1.0 (butane and pentane) and 0.2–1.0 (hexane) through the combinations of SMPs and day types, indicates that the changes were not statistically significant for a majority of stations.

Pei et al. (2022) reported a decrease of 13% in short-chain ($C < 6$) alkanes during lockdown in the Pearl River Delta (traffic site) when compared with periods immediately prior or after. However, due the disparity in the sources and emissions, in data aggregation and methodologies, a direct, quantitative, comparison with the data presented here is not meaningful. Nevertheless, specific disparities may be identified between Europe and the

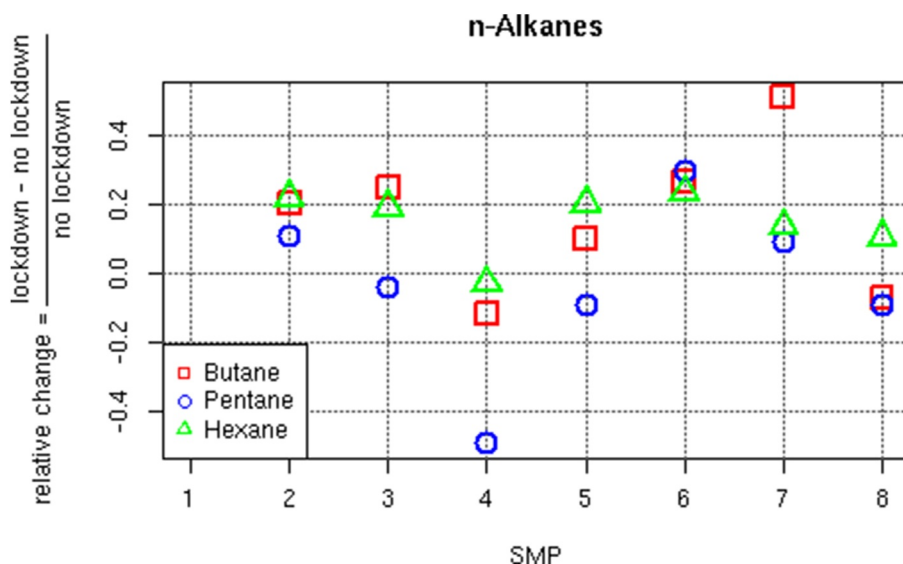


Figure 7. Median reduction through the SMPs for the *n*-Alkanes Butane, Pentane, and Hexane on weekdays. SMP 1 is left out for having, at most, only one subset.

Table 10

Acyclic Alkenes Concentrations ($\mu\text{g m}^{-3}$, Average \pm Standard Deviation of the Averages Throughout All Stations) During the 2020 Lockdowns and in the Same Period of the Four Previous Years, Average and Median Reduction ($\frac{\text{lockdown} - \text{no lockdown}}{\text{no lockdown}}$) Differentiated by SMP (Weekdays Only)

		SMP1	SMP2	SMP3	SMP4	SMP5	SMP6	SMP7	SMP8
All stations	$[\text{CH}_2 = \text{CH} - \text{C}(\text{CH}_3) = \text{CH}_2]_{\text{lockdown}}$	–	0.09 \pm 0	0.09 \pm 0.01	0.09 \pm 0	0.1 \pm 0.04	0.1 \pm 0.03	0.09 \pm 0	0.1 \pm 0.01
	$[\text{CH}_2 = \text{CH} - \text{C}(\text{CH}_3) = \text{CH}_2]_{\text{no lockdown}}$	0.09 \pm 0.02	0.08 \pm 0.02	0.09 \pm 0.01	0.09 \pm 0	0.09 \pm 0.01	0.1 \pm 0.03	0.09 \pm 0.01	0.09 \pm 0.01
	median reduction	–	0	0	0	0	0.01	0	0
	average reduction	–	–0.17	–0.04	0.01	–0.1	0.03	0	–0.03
All stations	$[\text{cis} - \text{CH}_3 - \text{CH} = \text{CH} - \text{CH}_3]_{\text{lockdown}}$	–	0.09 \pm 0	0.09 \pm 0.01	0.09 \pm 0	0.1 \pm 0.03	0.09 \pm 0.01	0.09 \pm 0	0.09 \pm 0
	$[\text{cis} - \text{CH}_3 - \text{CH} = \text{CH} - \text{CH}_3]_{\text{no lockdown}}$	0.1 \pm 0.03	0.09 \pm 0.03	0.09 \pm 0.02	0.09 \pm 0	0.09 \pm 0.01	0.1 \pm 0.04	0.09 \pm 0.01	0.1 \pm 0.01
	median reduction	–	0	0	0	0	0	0	0
	average reduction	–	–0.14	0	0.01	–0.05	0.07	0.01	0.03
All stations	$[\text{trans} - \text{CH}_3 - \text{CH} = \text{CH} - \text{CH}_3]_{\text{lockdown}}$	–	0.09 \pm 0	0.1 \pm 0.02	0.09 \pm 0	0.1 \pm 0.04	0.09 \pm 0.01	0.09 \pm 0	0.1 \pm 0.01
	$[\text{trans} - \text{CH}_3 - \text{CH} = \text{CH} - \text{CH}_3]_{\text{no lockdown}}$	0.11 \pm 0.08	0.1 \pm 0.03	0.1 \pm 0.02	0.09 \pm 0	0.1 \pm 0.02	0.11 \pm 0.07	0.09 \pm 0.01	0.1 \pm 0.03
	median reduction	–	0	0.02	0	0	0	0	0
	average reduction	–	–0.02	0.03	0.02	–0.02	0.07	0.03	0.04

PRD: butane and hexane decreased for a majority of stations in Europe (median reduction of 13%) and pentane increased (median increase of 8%). The concentrations of all three species decreased in the PRD ($\sim 15\%$ – $\sim 40\%$).

3.7. Acyclic Alkenes

Isoprene, a chemical marker of biogenic emissions (e.g., Abeleira et al., 2017), *cis*-2-butene and *trans*-2-butene, markers of industrial activities (e.g., used to produce synthetic rubber) with long lifetimes and typically associated to the regional background (Abeleira et al., 2017; Yang et al., 2018), were measured at 6, 8, and 7 monitoring stations, respectively, in a single country, representing 66, 110, and 77 subsets. Median and average reductions are shown in Table 10. In all combinations of SMP and day type, the median reduction was 0 for the three VOCs (average reductions of –0.02, 0.02, and –0.07 for *cis*-2-butene, *trans*-2-butene, and isoprene), evidencing only minor changes during the lockdowns.

Our data show stagnation in the concentration of alkenes, which is confirmed by the indication of no change given by the high *p*-values of the Wilcoxon-Mann-Whiney *U*-tests for the three species (means and medians between 0.21 and 1.0 through the combinations of SMPs and day types). This is in accordance with the long-lived characteristic and/or the biogenic origin of these species but in contradiction with the findings of Pei et al. (2022) who found a reduction of 24.8% at a traffic station in the Pearl River Delta (lockdown compared to periods immediately after and before): a contradiction that highlights the importance of considering meteorology when analyzing changes in atmospheric concentrations.

3.8. Sources of VOCs

The relative abundance of individual VOCs have been used as indicators of their dominant sources (e.g., Barletta et al., 2005; Borbon et al., 2018; Nelson & Quigley, 1984). Under lockdown conditions traffic was reduced, domestic burning may have increased due to the higher amount of time that people spent at home, and fossil fuel burning for energy generation may also have changed (e.g., Guevara et al., 2021). Therefore, a change in the relative contribution of the different sources to the ambient VOCs concentrations may appear in the data collected over Europe.

Sources of individual VOCs overlap in part, therefore investigating the variations in source strengths based on individual VOCs concentrations is difficult. On the other hand, the relationship between individual VOCs concentrations may give some information about sources (Andreae & Merlet, 2001).

3.8.1. Toluene to Benzene Ratio

Since toluene is present both in vehicular fuels and solvents, but benzene was banned from solvents, the toluene to benzene ratio (T/B) can be used as an indication of the contribution of traffic as a source of aromatic VOCs (Gelencsér et al., 1997; Ding et al., 2020; Ibragimova et al., 2021; Mor et al., 2021; Pakkattil et al., 2021; Pei et al., 2022; C. M. G. Salvador et al., 2022) and will be used here as a first approach at VOCs source apportionment. Several threshold values for that ratio have been presented both for fresh emissions or for particular environments with different degrees of detail. Z. Zhang et al. (2016) sampled near sources in China and gathered typical values of <1, 1–10 and >10 for biomass/coal burning, vehicular emissions, and industrial solvent use, respectively. Others reported typical values for traffic-dominated samples in the range 1–3, or higher in the case of freshly emitted vehicular emissions (e.g., Barletta et al., 2005; Borbon et al., 2018; Bruno et al., 2006; Miller et al., 2011; Monod et al., 2001; Salameh et al., 2019; Steinbacher et al., 2005; Wei et al., 2019). Samples for which industrial solvent use were identified as the main contribution exhibited a T/B value above 4.3 (Barletta et al., 2008; Miller et al., 2012; Tiwari et al., 2010). Away from sources, that is, in rural areas, T/B values are lower (e.g., Suthawaree et al., 2010) due to the much faster kinetics of toluene than benzene with the hydroxyl radical (Gelencsér et al., 1997). Other typical values used to apportion the influence of sources are 0.6 or <0.8 for biomass burning (Borbon et al., 2018; Gilman et al., 2015; Y. Liu et al., 2008) and 0.7 for coal combustion (Shi et al., 2020; M. Wang et al., 2013). In summary, in this paper, T/B ratio values around 0.6 are considered to correspond to biomass burning, 0.7 to coal combustion, 1–3 to vehicular traffic and > 4 to solvent use.

The T/B ratio was computed for 532 subsets from 50 stations. Figure 8 compares the average T/B ratio under no lockdown and lockdown conditions for each station. At stations of *type* background, the T/B ratio is clustered between the typical value for biomass burning and that for vehicular emission. With the exception of one station which lies in the area of values typically attributed to solvent use, both under lockdown and no lockdown conditions, the T/B ratio at stations of *type* traffic (blue in Figure 8) clusters around the values of 1.5–3.5. Such a distribution was expected from reported T/B from, for example, Paris (Baudic et al., 2016; Salameh et al., 2019) or northern Italy (Steinbacher et al., 2005). The T/B value for traffic stations clusters around the 1:1 line, showing little change between Lockdown and no Lockdown conditions, with one exception where the T/B ratio was lowered during the lockdown toward values more typical to biomass burning.

Figure S59 in Supporting Information S1 offers more detail and shows the T/B distribution across the stations on weekdays, differentiated by SMP, station *type* and *area*. At traffic sites, pre-lockdown T/B median values were above 2 under all SMPs but below 4, indicating traffic as the dominant source (Baudic et al., 2016; Salameh et al., 2019; Steinbacher et al., 2005). During the lockdowns, median values were lowered (except under SMPs 7 and 8), above the threshold of 1 but between the values indicated by Yang et al. (2018) and Ding et al. (2020) for traffic (1.6) and coal/biomass combustion (0.6). Under SMP7, there was a slight increase, whereas SMP8 exhibits minimal change. The same pattern occurred at stations of *area* urban or even suburban. For the latter, the lockdown exhibited larger reductions. The median T/B ratios at stations of *type* background exhibited a similar pattern, albeit with lower lockdown and no lockdown values, closer to the typical coal/biomass burning value. At industrial stations, T/B range includes higher values as might be expected based on previous work. The effect of the lockdown at industrial stations, where before the lockdowns the influence of industrial solvent use translated into distributions including higher values than at stations of other *types*, was to shift the T/B medians toward values more typical of vehicle exhaust and coal/biomass burning. This could potentially indicate a reduction in industrial activity. T/B at stations of *area* rural showed distributions centered around values typical of coal/biomass burning with the lockdown having the effect of shifting those distributions further to lower values.

For all station types or areas, the effect of the lockdown was to shift the T/B distribution toward lower values, which we interpret as a decrease in the emissions from traffic and industry. However, SMPs 7 (weak baric gradient over central and eastern Europe with moderate S flows at the 850 mb level) and 8 (strong baric gradient across N Europe accompanied by fast NW-N flows) exhibited an increase (SMP7) or no change (SMP8) in the median T/B.

3.8.2. Benzene to Toluene to Ethylbenzene Ratio

The benzene to toluene to ethylbenzene (B:T:E) ratio may be used to investigate the relative contributions of the sources of atmospheric aromatics. Figures 9 and 10 show ternary plots for the B:T:E ratio under no lockdown and lockdown conditions, and the differences in relative importance, for the different station *types* (Figure 9) and

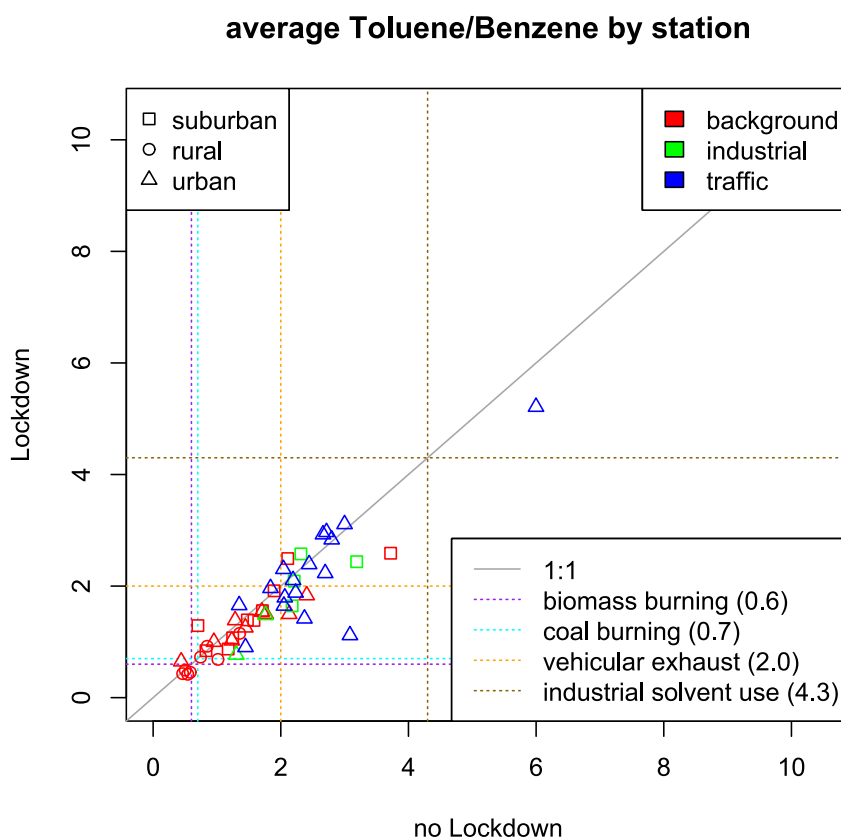


Figure 8. Average Toluene to Benzene ratio under no lockdown and lockdown conditions for stations. Station *type* and *area* are indicated. Vertical and horizontal lines indicate the typical T/B ratio for different sources as reported in the literature.

station *areas* (Figure 10). Z. Zhang et al. (2016), reported typical B:T:E ternary ratios for three different sources. Those are indicated in the ternary plots (colored circles): 0.69:0.27:0.04 for Biomass/biofuel/coal burning emissions (blue, lower right), 0.31:0.59:0.10 for traffic emissions (red, upper right), and 0.06:0.59:0.35 for industrial/solvent use emissions (green, upper left). The average B:T:E partial ratios are given in Table S1 in Supporting Information S1 differentiated by SMP and station *type*.

Because ethylbenzene measurements only yielded 306 subsets (from 30 stations), against 4,412 and 580 for benzene and toluene, respectively, ethylbenzene is the limiting component when analyzing the B:T:E ratio. It was possible to compute the B:T:E ratio at 26 stations from 4 countries (14 stations in Belgium, 8 stations in Hungary, 1 station in Ireland, all of which with a daily time resolution, and 3 stations in Germany, with hourly time resolution) and a majority of them (20) of the *type* Background, 4 of the *type* traffic and 2 of the *type* industrial. As a consequence, the interpretation of the data set has a limited reach.

The differential plots (right column in Figure 9) for sites of *type* traffic exhibit a decrease in areas around typical traffic values toward, mostly, the upper vertex, representing a large dominance of toluene over benzene while maintaining ethylbenzene relatively constant. A possible explanation, in terms of sources, could be an increased relative importance of solvent use against traffic. The behavior at traffic sites is replicated at sites of *area* urban. Not as important as the shift toward the upper vertex of the triangle, but still discernible, is an increase toward the lower right corner (more evident at urban than traffic sites) evidencing an increase in the relative importance of biomass burning. The latter trend is more important at sites of *area* suburban and/or *type* background. Industrial (*type*) and/or rural (*area*) show some of both tendencies although with much less data points.

Figure 11 shows the distribution of B:T:E differentiated by SMP, SMPs 1 (no data during the lockdowns) and 2 (52 data points during the lockdowns and results similar to SMP3) are not shown due to limited data and for simplicity.

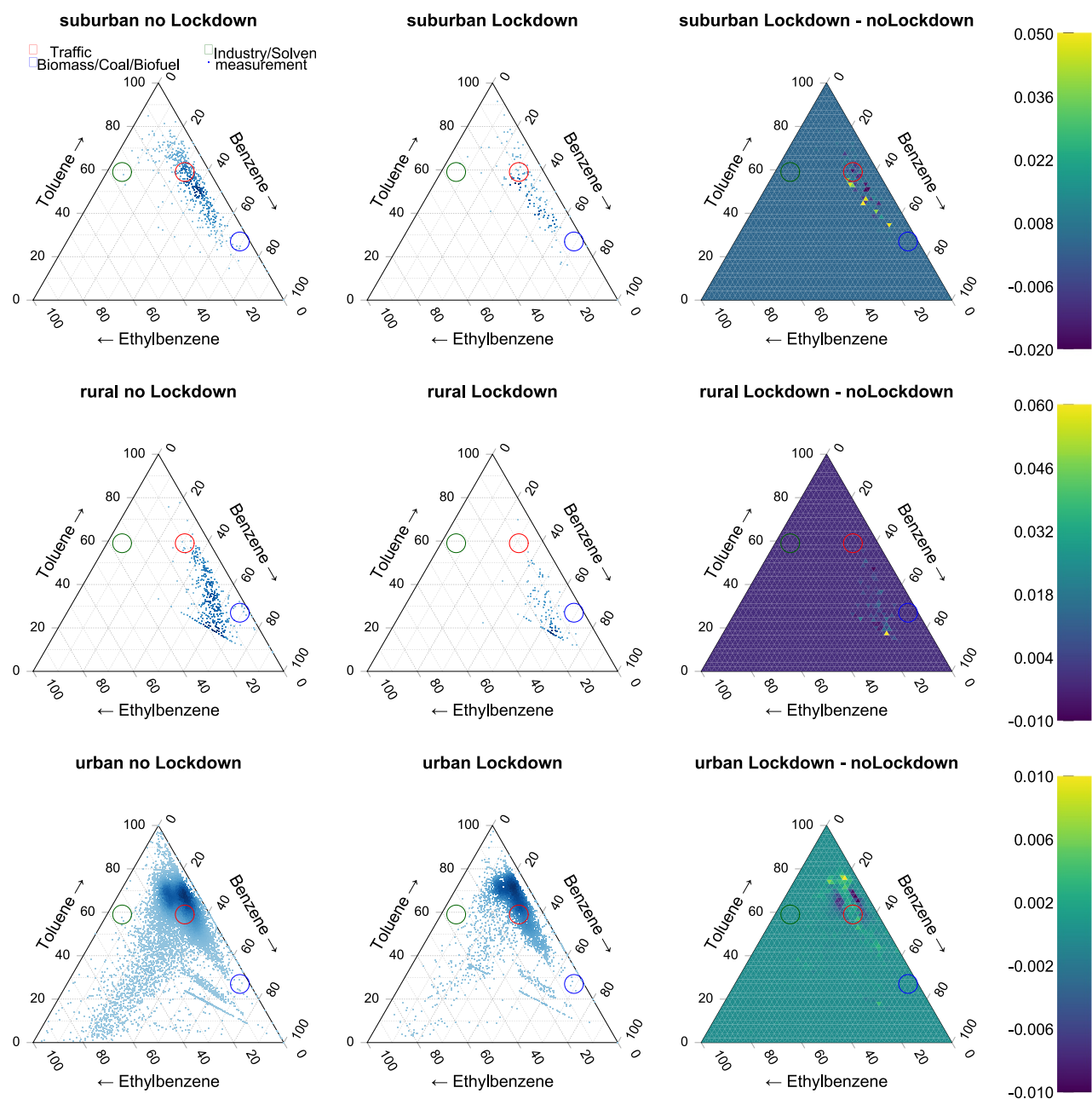


Figure 9. Ternary plots for the Benzene to Toluene to Ethylbenzene ratio, differentiated by station *type*, under no-Lockdown (left) and Lockdown (middle) conditions, lighter shades mean lower point density, darker shades mean higher point density, and for the difference in the relative contribution (right). The relative contribution was computed as the number of measurements at a given point in the tridimensional space divided by the total number of measurements. Areas with yellow shades gained whereas areas with purple shades lost relative importance with the lockdowns.

SMPs 3, 4, 7, and 8 exhibit a trend similar to that of traffic/urban sites: strong increase in areas more typical of solvent use and moderate increase toward areas typical of biomass burning. For SMP6 (moderate E flows), and even more for SMP5 (moderate N-NE flows), the increase toward areas typical of biomass burning becomes preponderant.

We hypothesize a general increase in the use of solvents and in the burning of biomass relative to traffic. The latter is preponderant under colder conditions (moderate north-eastern or eastern winds generally bring colder air) and in suburban/background areas, whereas the former is more important in traffic/urban areas and under

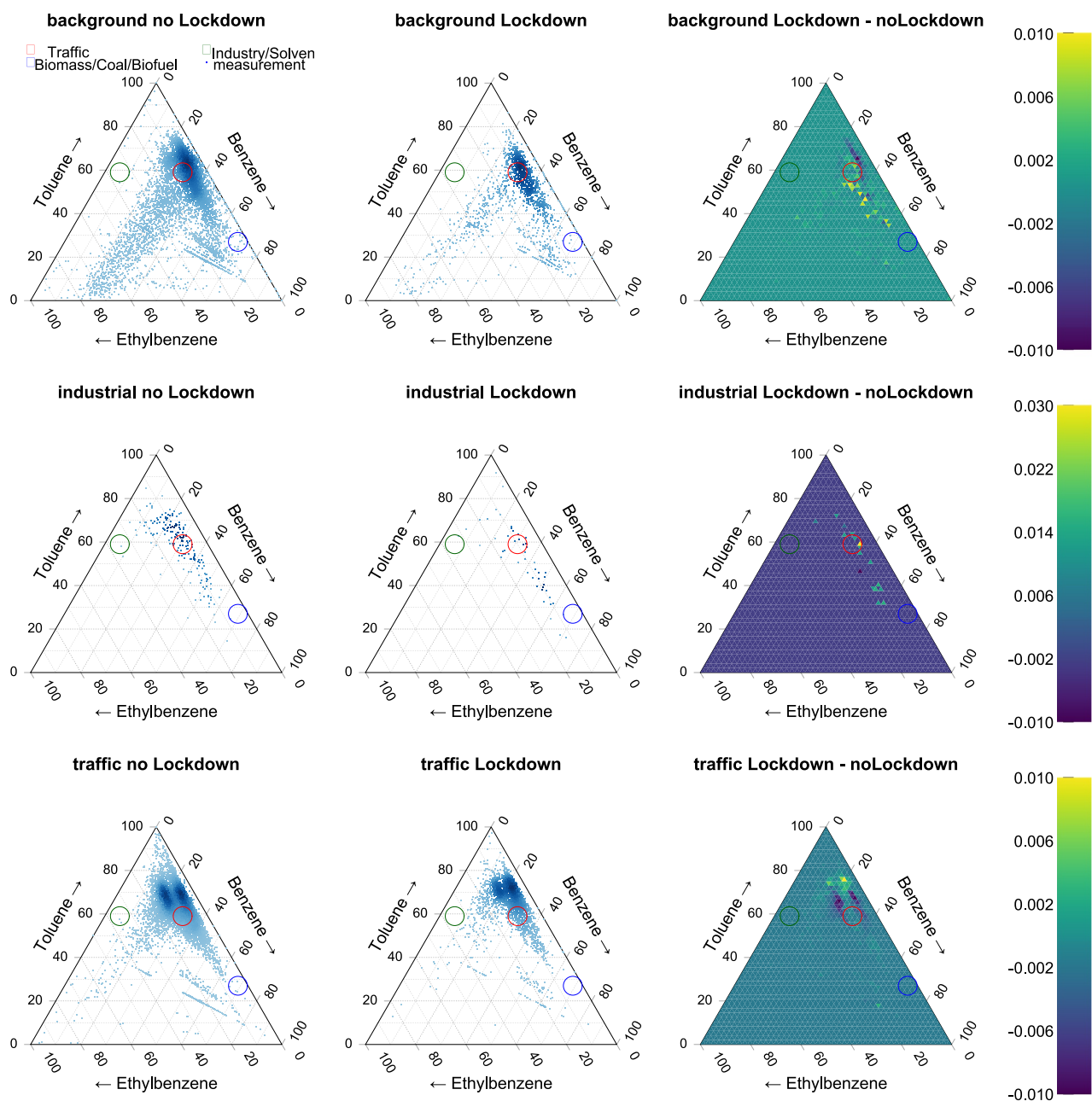


Figure 10. Ternary plots for the Benzene to Toluene to Ethylbenzene ratio, differentiated by station *area*, under no-Lockdown (left) and Lockdown (middle) conditions, lighter shades mean lower point density, darker shades mean higher point density, and for the difference in the relative contribution (right). The relative contribution was computed as the number of measurements at a given point in the tridimensional space divided by the total number of measurements. Areas with yellow shades gained whereas areas with purple shades lost relative importance with the lockdowns.

meteorological conditions of slow flows and stagnation (SMPs 3 and 4) or moderate flows bringing cleaner/maritime air (SMPs 7 and 8).

4. Conclusions

In early 2020, governments throughout Europe took measures to limit human contact in order to slow down the circulation of the novel coronavirus SARS-CoV-2. Those measures, commonly referred to as lockdowns, considerably reduced human activity and thus atmospheric emissions. In the present work, we gathered measured

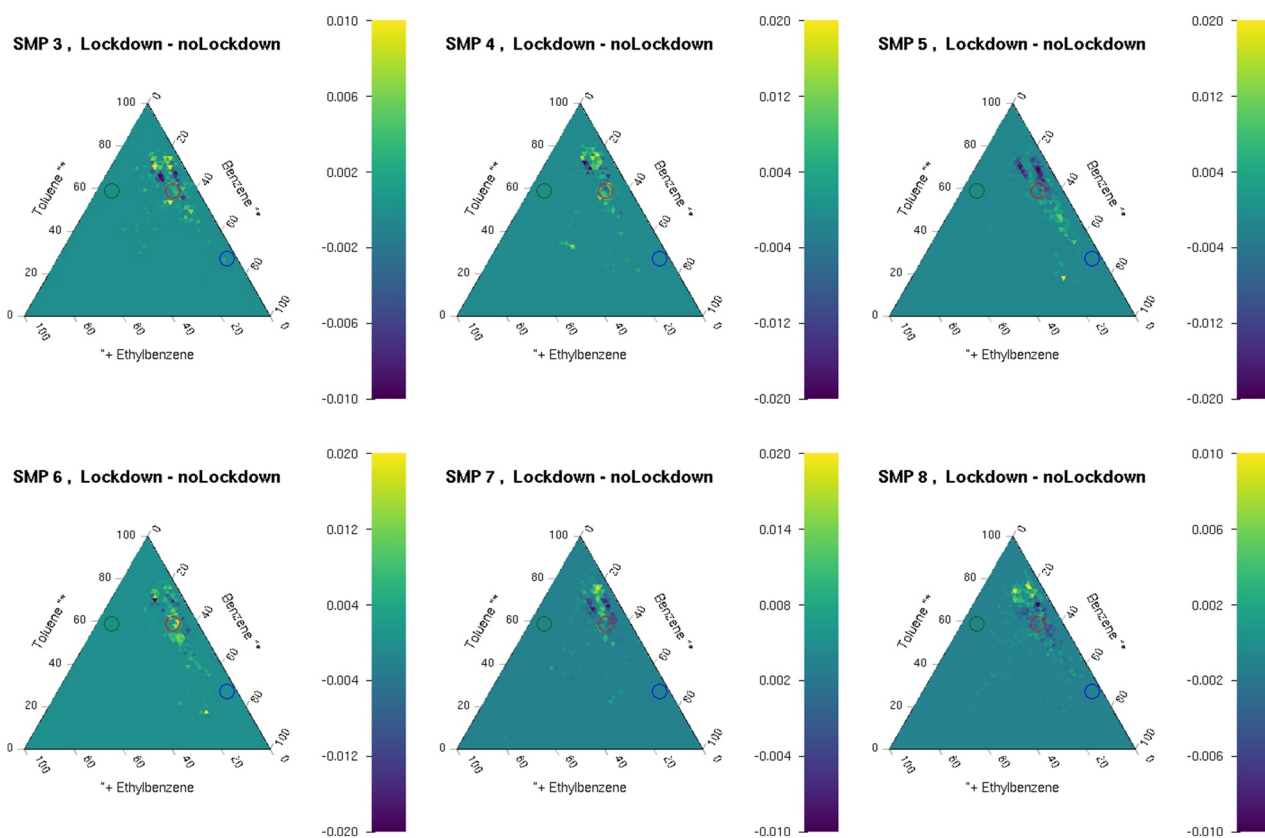


Figure 11. Ternary plots for the Benzene to Toluene to Ethylbenzene ratio, differentiated by SMP (SMPs 1 and 2 were left out for lack of data and/or clarity) for the difference in the relative contribution. The relative contribution for the lockdown and the no lockdown periods was computed as the number of measurements at a given point in the tridimensional space divided by the total number of measurements. Areas with yellow shades gained whereas areas with purple shades lost relative importance with the lockdowns.

VOCs concentrations from the European Environment Agency repository for monitoring data, AirBase. Each concentration was geolocated to a monitoring station within a country. Only those data within the period of the country 2020 lockdown and the corresponding period of the 4 years prior (2016–2019) were considered in order to study the VOCs concentration evolution under lockdown conditions. The data were classified in terms of weekday or weekend to account for human activities and in terms of synoptic meteorological patterns to account for the influence of meteorology and, to some extent, chemistry.

Concentrations from a large number of stations in many countries located in geographical regions with different characteristics have been analyzed. For this reason, it is difficult to determine general conclusions for the whole continent and/or for all the VOCs under study. More focused studies should be conducted to derive conclusions with finer granularity. Nevertheless, some general trends were identifiable. Benzene and its methylated (toluene, xylenes, and trimethylbenzenes) and ethylated (ethylbenzene) derivatives generally follow a similar trend, which shows a reduction during the lockdowns for a majority of stations. Only benzene and toluene were reported from a sufficient number of stations to allow for a differentiation between station *types* or *areas*. Traffic or urban stations exhibited the most important reduction in benzene and toluene concentrations with a more significant decrease for toluene. SMP6 was the most affected by the decrease for both benzene and toluene. For benzene, the SMPs less affected by the lockdown were SMPs 1 and 5, whereas for toluene those were SMPs 7 and 8. Here, we find that, besides for dispersion, SMPs must be relevant for controlling sources, because no similarities are found between SMPs which are analogous in terms of dispersion. For xylenes, trimethylbenzenes, and ethylbenzene, the SMPs more affected by the lockdown were SMPs 5 and 6, those less affected were the SMPs related to slower flows (2, 3, and 4). Regarding *n*-Alkanes, hexane exhibited a general increase under lockdown, except for SMP 4, butane

decreased under SMPs 4 and 8, whereas pentane decreased under SMPs 3, 4, 5, and 8. Acyclic alkenes (butenes and isoprene) evidenced no change in their concentration under lockdown.

The differentiation by SMPs generates more interpretable results in cases where VOCs are emitted from anthropogenic sources and in areas with a high density of sources (i.e., urban areas). For example, the RoA and RoM for benzene were close to 1 under intense atmospheric dispersion conditions (SMPs 1 and 5), whereas under stable spring conditions the ratios are less than 1, for example, decreased benzene concentrations under SMPs 2–4. On the other hand, for VOCs generated by a wide variety of sources, including biogenic ones, the effect of the lockdown on their concentrations is less significant, regardless of the SMP, because of the importance of other meteorological factors such as temperature, solar radiation, or humidity for emissions, which depend more on the geographical region than on the SMP. Isoprene is an extreme of such cases.

The evolution of source strengths under lockdown conditions was investigated by means of diagnostic ratios (toluene to benzene and benzene to toluene to ethylbenzene) commonly explored in the literature. The diagnostic ratios showed a decrease in the relative source strength of traffic in favor of biomass/coal/biofuel burning and solvent use, and it was possible to identify an increase in the use of domestic solvent.

The effect of the lockdown on VOCs will impact secondary pollutants (O_3 and secondary organic aerosol, SOA). For O_3 , the extent of the impact will depend not only on the absolute change in VOCs or NO_x concentrations but also on the change in the VOC/ NO_x ratio (i.e., on the chemical regime which, in European cities, is generally VOC-limited). We found that traffic dominated the observed reduction in VOC concentrations in the early spring 2020 lockdowns in Europe. This traffic reduction meant a reduction in the NO_x emissions and concentrations (e.g., von Schneidmesser et al., 2021), probably leading to reduced NO titration and possibly a change in chemical regime, favoring an increase in O_3 concentrations. Control policies should focus on sources that have a high VOC/ NO_x emission ratio as also emphasized by Kroll et al. (2020), H. Wang et al. (2022), and K. Zhang et al. (2022). The data and the results presented here have the potential to be used in an effort to estimate and investigate the effect of the lockdowns on secondary pollutants such as O_3 and SOA as well as in the design of policies aimed at reducing atmospheric pollution levels.

Data Availability Statement

The VOCs concentrations data can be downloaded from the AirBase repository: <https://www.eea.europa.eu/data-and-maps/data/aqreporting-9> (European Environmental Agency, n.d.). The meteorological fields for the synoptic classification were obtained from NOAA: <https://psl.noaa.gov/data/reanalysis/reanalysis.shtml> (Kalnay et al., 1996).

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