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Ozone levels in European and USA cities are increasing more than at rural sites, while peak values are decreasing

Elena Paoletti [a, *], Alessandra De Marco [b], David C.S. Beddows [c], Roy M. Harrison [c], William J. Manning [d]

[a] IPSP-CNR, Florence, Italy
[b] ENEA, Rome, Italy
[c] National Centre for Atmospheric Science, School of Geography, Earth & Environmental Sciences, University of Birmingham, B15 2TT, United Kingdom
[d] Dept Plant, Soil Insect Sciences, Univ. Massachusetts, Amherst, USA

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ABSTRACT

Ground-level ozone (O_3) levels are usually lower in urban centers than nearby rural sites. To compare trends in O_3 levels during the period 1990–2010, we obtained monitoring data from paired urban and rural sites from the European Environment Agency and the US Environmental Protection Agency. Ozone peaks decreased at both station types, with no significant differences between urban and rural stations. Ozone annual averages increased at both urban and rural sites, with a faster rate of increase for urban centers. The overall trend was for convergence between urban and rural O_3 data. Ozone levels exceeded the criteria established for the protection of human and vegetation health at both urban and rural sites.

1. Introduction

Ground-level ozone (O_3) is a global air pollutant and an important anthropogenic greenhouse gas (Fiore et al., 2002). The chemistry of O_3 formation requires photolysis and is favored by high temperatures. The majority of tropospheric O_3 comes from photochemical reactions of the so-called O_3 precursors, i.e. methane (CH_4), carbon monoxide (CO), volatile organic compounds (VOCs), and nitrogen oxides (NO_x), which are largely emitted from anthropogenic sources. In urban centers where NO_x levels are high, nitric oxide (NO) favors O_3 depletion and O_3 formation occurs by inputs of VOC, with such conditions referred to as VOC-limited (Royal Society, 2008). Ozone formation at rural sites is governed by levels of hydrocarbons, especially biogenic VOCs (BVOCs) emitted from forests and other vegetation (Calfapietra et al., 2013). In such rural areas, O_3 formation increases with increasing NO_x and is referred to as NO_x-limited. Because of higher NO_x levels and lower BVOC emission, O_3 levels are usually lower at urban centers than at nearby rural sites (EEA, 2013). Ozone formed in cities is also removed downwind to non-urban areas and moved by long-range transport.

Ozone effects on human health include lung inflammation, reduced lung function, degenerative diseases, age related disorders and eventually cancer (Kampa and Castanas, 2008). Recent studies estimated considerably larger effects than previously thought, i.e. 0.7 M/yr respiratory mortalities worldwide, corresponding to 1.1% of all mortalities (Anenberg et al., 2010). Ozone is the second most important air pollutant, after particulate matter, in causing human mortality and morbidity impacts. Effects on vegetation are complex and include reduced photosynthesis and carbon sequestration, reduced stomatal conductance and control on water losses, biodiversity changes, occurrence of leaf visible injury (Paoletti, 2007). The global impact of ozone on crop yield losses is estimated to be 14–16 billion US$ (Van Dingenen et al., 2009). Forest ecosystem services are also altered by ozone (Ainsworth et al., 2012).

Background O_3 concentrations have risen from ~10 ppb before the industrial revolution (Volz and Kley, 1988) to daytime summer concentrations exceeding 40 ppb in many parts of the Northern Hemisphere (Vingarzan, 2004). Chemistry transport models suggest recent reductions in O_3 peaks in North America and Europe (Dentener et al., 2010), which are likely due to effective control measures on NO_x and VOCs in response to the Clean Air Act in the

* Corresponding author.
E-mail address: elena.paoletti@cnr.it (E. Paoletti).

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USA and the Thematic Strategy on Air Pollution in Europe. Future trends depend upon climate change and O_3 precursor emissions. Legislation-enforced control of NO_x emission, however, is likely to translate into higher O_3 concentrations in the VOC-limited urban atmospheres. In North America, primary standards for the protection of human health are available, and guidelines to also protect ecosystems are discussed (EPA, 2008; CCME, 2000). In Europe, a number of O_3 standards to protect both human and ecosystem health have been established (Paoletti and Manning, 2007).

On the basis of the two-decadal O_3 monitoring records available in Europe and the USA, our aim was to assess O_3 risk to plants and population by comparing trends in O_3 levels during the period 1990–2010 and testing exceedances of air quality guidelines. The analysis was carried out at paired urban-rural sites in order to ensure that the sites were influenced by the same regional air mass.

2. Materials and methods

Quality assured data from urban and rural stations were obtained from the European Environment Agency (EEA) and the US Environmental Protection Agency (EPA) databases. To avoid artifacts due to local pollution effects and insufficient data availability, only background stations with an annual coverage >80% over the period from 1990 to 2010 were selected. However, most stations in the USA have O_3 monitoring data available for download since 1993, while the 2010 records of all the European stations were not completed at the time the data were downloaded. Therefore, stations were selected when covering the period 1993–2010 in the USA and 1990–2009 in Europe. Stations were considered as a pair when the distance between the urban and rural station at each city did not exceed 50 km. This 50-km threshold would reasonably ensure that the paired sites were influenced by the same regional air mass. The urban designation implies that the site was more likely to be exposed to fresh NO_x emissions and thus NO titration of O_3. The rural designation implies that the site was less likely to be subject to fresh NO_x emissions and NO titration. Assuming that pollution dynamics are affected by the city size, only large cities (≥0.5 M inhabitants) were selected. Such strict selection criteria resulted in eight US and seven EU paired stations (Table 1).

Table 1
Characteristics of the paired urban (U) and rural (R) stations, selected in Europe and the USA.

City, State	Inhabitants [Million]	Station	Altitude [m a.s.l.]	Longitude [degrees]	Latitude [degrees]
Sacramento, California	2.149	U	0	−121.491	38.558
		R	6	−121.422	38.301
Las Vegas, Nevada	1.951	U	0	−115.266	36.224
		R	0	−114.906	36.390
Tucson, Arizona	0.980	U	756	−110.974	32.222
		R	937	−110.774	32.047
Albuquerque, New Mexico	0.887	U	1592	−106.585	35.134
		R	1806	−106.508	35.185
Oklahoma city, Oklahoma	1.253	U	397	−97.494	35.477
		R	353	−97.475	35.614
Dallas, Texas	6.426	U	125	−96.860	32.819
		R	0	−96.871	32.676
Houston, Texas	5.920	U	0	−95.503	29.724
		R	55	−95.673	30.039
Philadelphia, Pennsylvania	5.965	U	22	−75.097	40.008
		R	28	−75.011	40.076
London, UK	8.308	U	20	−0.120	51.517
		R	125	0.181	50.794
Paris, France	2.211	U	60	2.346	48.891
		R	152	1.883	48.581
Bruxelles, Belgium	1.119	U	100	4.358	50.796
		R	20	4.225	50.768
Berlin, Germany	3.502	U	35	13.349	52.542
		R	50	13.225	52.473
Wien, Austria	1.731	U	207	16.357	48.249
		R	286	16.206	47.959
Varsavia, Poland	1.720	U	98	20.962	52.280
		R	166	19.517	51.291
Helsinki, Finland	0.596	U	21	24.950	60.187
		R	64	24.689	60.316

Many ozone-exposure metrics are available to summarize risk to people and vegetation (Paoletti et al., 2007). Among those metrics, we selected the most used for research and regulatory purposes to protect vegetation and human health in North America and Europe, i.e.: M24, annual mean of hourly concentrations; Max: maximum value of hourly concentration in a year; AOT40: annual sum of the excess of hourly concentrations over the cut-off of 40 ppb during daytime hours (8 am–8 pm), calculated from April to September for the protection of forest trees (AOT40F, ICP, 2010) or from May to July for any other kind of vegetation (AOT40dir, 2008/50/CE Directive) in Europe, HHdir: for the protection of human health in Europe, calculated as cases of excess of daily maximum 8-h means over the cut-off of 60 ppb in a year (2008/50/CE Directive); NAst: primary standard in North America for the protection of human health, calculated as annual fourth highest value in the array of the highest daily maximum 8-h averages (EPA, 2008); and W126: discussed as secondary standard for the protection of ecosystems in the USA (EPA, 2008), calculated as sum of the hourly concentrations from 8 am to 8 pm from May to September in a year, where each concentration c_i is weighted by a sigmoidal function $w_i = 1/(1 + 4403 \exp -0.126\ c_i)$, to assign greater emphasis to the higher concentrations.

Normality of data distribution was tested by the Kolmogorov–Smirnov test. Student *t*-test was applied for comparing the effect of station type or continent. Given the great site-specific and year-by-year variability, percent deviations from the two-decadal (1990–2010) average were calculated at any station and a linear regression over time was applied. Non-parametric correlation (Spearman R) was applied when data were not normally distributed i.e. when Max was tested. The effects of station type were tested between lines for colinearity by the homogeneity of regression lines test. All statistics were applied by the STATISTICA 7.0 software (StatSoft).

3. Results

All the metrics were significantly higher at rural sites than in the cities, except Max in both continents and HHdir in Europe where no difference between rural and urban centers was recorded (Fig. 1). All of indices were higher in the USA than in Europe. The criteria to protect human and vegetation health were frequently exceeded at both urban and rural sites (Table 2). Exceedances at the rural sites, however, were ~10–20% more frequent than at the urban sites. The exceedances were ~35–40% more frequent in the USA than in Europe. All the indices were significantly correlated to each other, with the exception of Max (Table 3). Therefore, annual trends are shown only for M24 and Max.

M24 deviations from the two-decadal average showed significant increases over time in both continents and for both station types (Fig. 2). In detail, the annual increase was 7% at rural stations in both continents, and 12% and 17% at urban stations in USA and Europe, respectively. The differences in the regression slopes of urban and rural stations were significant in Europe and not significant in the USA. The coefficients of determination show that 10–15% and 25–42% of the variance in M24 deviations was explained by the year at urban and rural stations, respectively. Max deviations from the 1990–2010 average showed significant decreases over time in both continents and for both station types (Fig. 3). In detail, the annual decrease at urban and rural stations was 14 and 18% in Europe and 10 and 12% in the USA, respectively. The regression slopes at urban and rural stations did not differ significantly, and 11–13% and 26–32% of the variance in Max deviations was explained by the year at European and US stations, respectively.

4. Discussion

Ozone pollution continues to be a serious issue for public health and the environment. Europe and USA have significantly cut domestic emission of O_3 precursors in recent times (EEA, 2013; EPA, 2013). Summer emissions of VOCs and NOx in the USA decreased 20 and 30%, respectively, during the period 1997 to 2006, and NAst was 9% lower in 2006 than in 1990 (EPA, 2008). Emissions of NOx, non-methane VOC, CO and CH_4 decreased by 27%, 28%, 32% and 15%, respectively, in the European Union between 2002 and 2011,

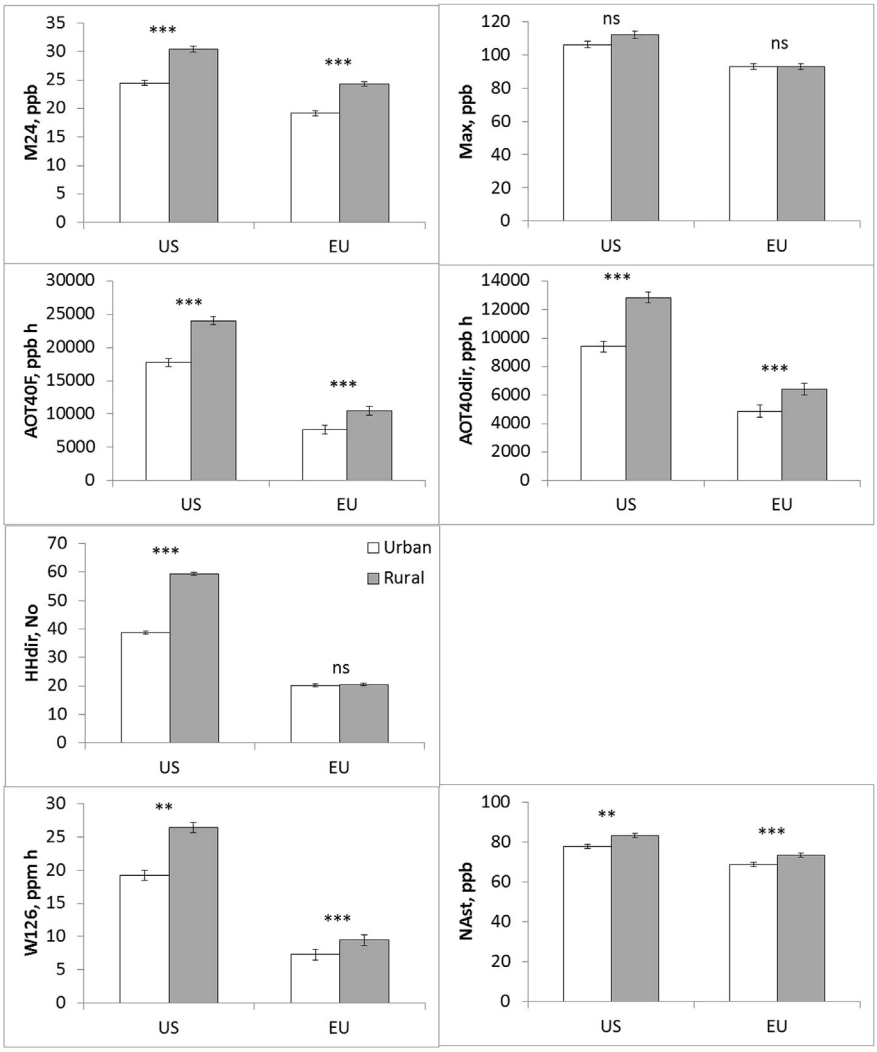

Fig. 1. Annual averages (±S.E.) of M24 (annual mean of hourly concentrations), Max (annual hourly peak), AOT40F (European guideline for the protection of forests), AOT40dir (European standard for the protection of vegetation), HHdir (European standard for the protection of human health), W126 (secondary standard for the protection of ecosystems in the USA), and Nast (primary standard for the protection of human health in North America), at urban (white bars) and rural (gray bars) stations in Europe (EU) and the United States (US) over the period 1990–2010. Asterisks show significant differences between urban and rural stations in each continent (***, $p < 0.001$; **, $p < 0.01$; ns, $p > 0.1$). Differences between continents were always significant ($p < 0.001$).

but there was no clear trend for HHdir over the same period in 80% of the monitoring stations and no clear effect of the station type (EEA, 2013). Over a twice-longer period, we compared a selection of paired urban-rural stations and calculated the percent deviations relative to the two-decadal average, thus removing the effects of site and annual variability, respectively. The results suggest a clear and significant reduction of annual peaks at both urban and rural stations in Europe and the USA. The station type did not significantly affect this response. In contrast, background O_3 levels as summarized by annual averages were increasing, with a faster rate for urban centers. As O_3 levels are traditionally lower in urban centers than at nearby rural sites (Gregg et al., 2003; Paoletti, 2009; Sicard et al., 2013), our results suggest a convergence between urban and rural O_3 pollution in the long term. These results are consistent with an analysis of Mediterranean European stations over the period 2000–2010, where Max decreased both at rural (−1.21%/y) and urban (−1.25%/y) stations, and M24 decreased at rural sites (−0.43%/y) and increased at urban centers (+0.64%/y) (Sicard et al., 2013). Eleven remote sites in Europe, North America and Asia doubled M24 from 1950 to 2000 (Parrish et al., 2012). Significant daytime increases (0.19–0.50 ppb/yr) occurred at several rural sites in the western USA from 1987 to 2004 (Jaffe and Ray, 2007). A strong increase in springtime O_3 mixing ratios during 1995–2008 occurred in the free troposphere over western North America, with a rate of increase of around 0.63 ppb/yr (33rd, 50th and 67th percentiles), while rates for the 95th and 5th percentiles were not significant (Cooper et al., 2010). Significant increases in O_3 mean concentrations during the 1990s were recorded at Northern Hemispheric rural sites (Vingarzan, 2004). Most rural sites of

Table 2
Percentage of stations exceeding the annual thresholds for protection (in parenthesis). See Fig. 1 for acronyms.

Standard	US Urban	US Rural	EU Urban	EU Rural
AOT40F (5000 ppb h)	98	100	63	85
AOT40dir (9000 ppb h)	45	72	15	24
HHdir (25 cases)	62	94	29	33
W126 sensitive species (7 ppm h)	92	99	47	56
W126 tolerant species (15 ppm h)	62	88	11	18
NAst Canada (65 ppb)	93	98	63	75
NAst USA (80 ppb)	41	56	22	29

Table 3
Matrix of correlation (Pearson *r*) coefficients on all the years and stations. Non-parametric correlation coefficients (Spearman R) were computed for Max. See Fig. 1 for acronyms. The significance *p* of all the values was <0.001 unless stated, ° $p = 0.130$.

	M24	Max	AOT40F	AOT40dir	W126	NAst
Max	$r = 0.0794$°					
AOT40F	$r = 0.7820$	$r = 0.7552$				
AOT40dir	$r = 0.7418$	$r = 0.7001$	$r = 0.9443$			
W126	$r = 0.6809$	$r = 0.6583$	$r = 0.9686$	$r = 0.9079$		
NAst	$r = 0.2727$	$r = 0.2627$	$r = 0.6015$	$r = 0.5036$	$r = 0.6763$	
HHdir	$r = 0.6792$	$r = 0.6532$	$r = 0.9378$	$r = 0.8697$	$r = 0.9403$	$r = 0.6522$

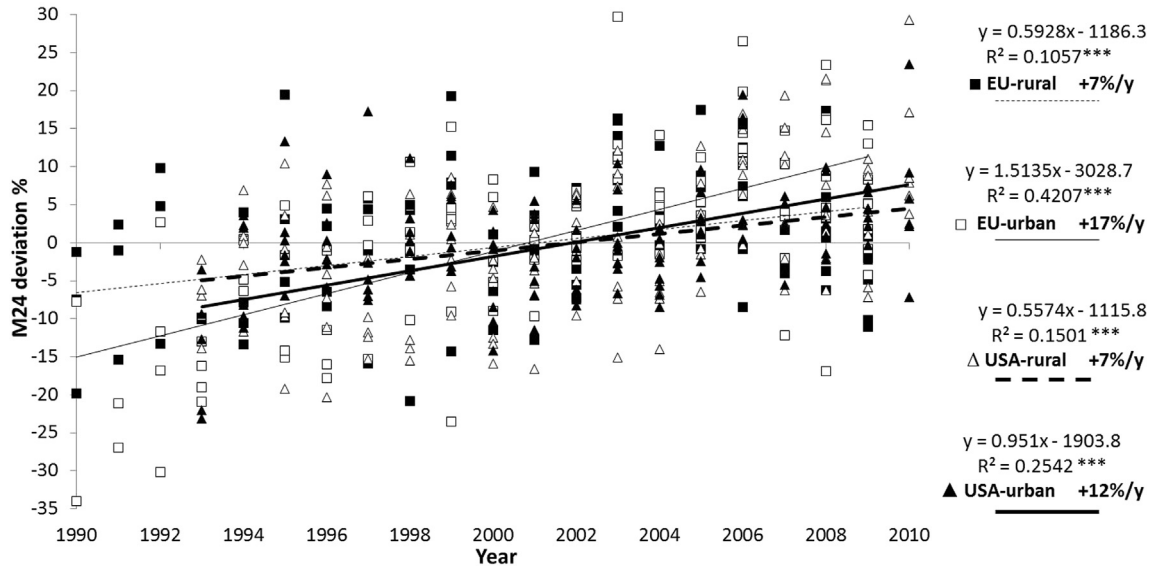

Fig. 2. Annual deviations of M24 (%) from the two-decadal (1990–2010) average at any station. Linear regressions were applied to rural and urban stations in the USA and Europe. Asterisks show the level of significance ($p < 0.001$).

Europe showed a substantial decrease of high O_3 levels (98th or 95th percentiles) over the past 15 years (Wilson et al., 2012). NAst and W126 decreased at 57% and 43% monitoring sites, respectively, since 1980 to 2005 in the USA (Lefohn et al., 2008). All these observations confirm that reductions in precursor emissions are globally reducing O_3 extremes, although background levels are still increasing. This discrepancy might be due to meteorological variability and hemispheric transport of pollution (Dentener et al.,

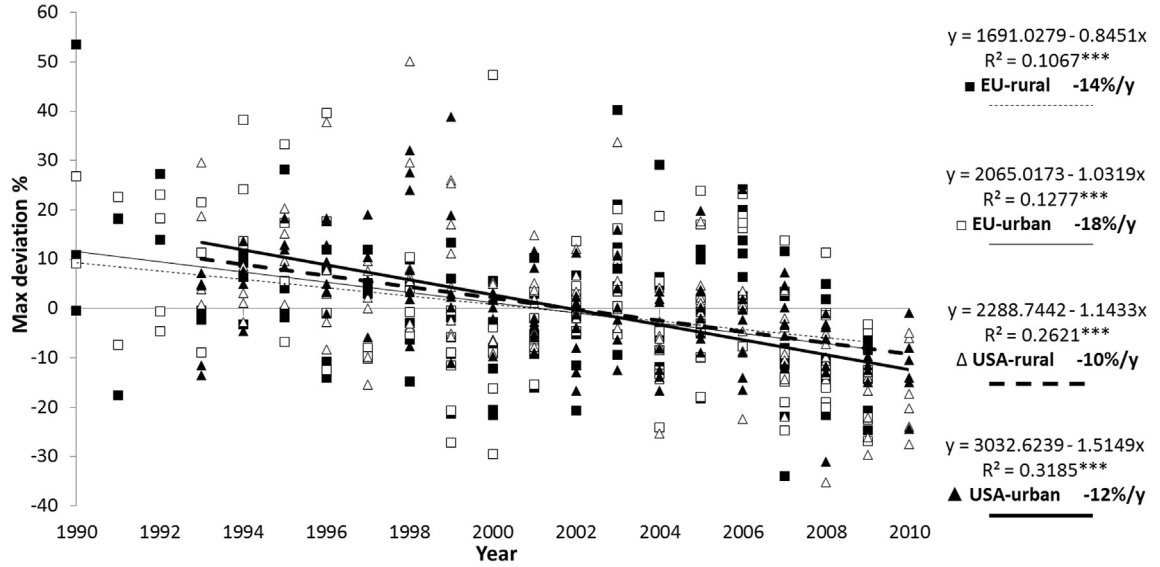

Fig. 3. Annual deviations of Max (%) from the two-decadal (1990–2010) average at any station. Linear regressions were applied to rural and urban stations in the USA and Europe. Asterisks show the level of significance ($p < 0.001$).

2010). As a result, cumulative indices, such as AOT40 for the protection of plant ecosystems, are increasing more than peak indices, such as those for the protection of people and W126.

It is a known fact, however, that O_3 exposure is not representative of actual injury to vegetation, as stomata close during unfavorable periods and avoid O_3 uptake (Paoletti and Manning, 2007). Very preliminary research, however, is available about measurements of stomatal O_3 flux to urban (Wang et al., 2012) and peri-urban (Fares et al., 2013) forests. The value of urban forests in providing ecosystem services required by urban residents is increasing as urbanization increases (Nowak et al., 2013). In parallel, O_3 is posing more serious risk to urban forests due to the increase in background levels. The main removal process for O_3 in the air is deposition to the surface, known as dry deposition (Cieslik et al., 2009). Dry deposition to terrestrial ecosystems is largely controlled by stomata, which are responsible for 30%–90% of total ecosystem O_3 uptake (Fowler et al., 2009). Therefore, more research on stomatal O_3 flux in the cities is recommended, also for quantifying the potential of urban vegetation as an ozone sink.

As expected (e.g. Landry and Cupelin, 1981), the ratio of urban to rural M24 was always lower than 1 (~0.8). Although all the indexes were higher at rural sites than in the cities, urban air did not always meet the European and USA criteria to protect human and vegetation health, confirming results for Italy by Paoletti (2009). Exceedances at the rural sites were 10–20% more frequent than at the urban sites. The exceedances were 35–40% more frequent in the US than in Europe. Further investigations are needed to test whether higher O_3 pollution in the USA than in Europe may be due to different control policies over O_3 precursors emission or to meteorological effects. The consistency of M24 and Max trends between continents in spite of higher O_3 pollution in the US is, however, remarkable.

5. Conclusions

In conclusion, O_3 levels in and around US and European cities are high enough to jeopardize human and vegetation health, according to current standards. Trends over the past 20 years suggest that O_3 risk to vegetation is rising in the cities more than at rural sites, while risks for human health are decreasing at both rural and urban sites. Such behavior is a response to control measures. Only by more stringent controls of both NOx and VOCs, Europe and the USA may meet compliance with O_3 air quality standards in both urban and rural areas.

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