Atlas of ozone chemical regimes in Europe

The complexity of ozone chemistry adds to the difficulty of understanding the observed trends in ozone concentrations and how best to mitigate potentially harmful levels of ozone in the atmosphere. This article summarises a study which aims to provide new insights into the sensitivity of changes in ozone concentrations to reductions in anthropogenic emissions, specifically transport and industrial emissions. The study results have been published as an 'Atlas of ozone chemical regimes in Europe' which presents the modelled data for ozone concentrations across 22 European cities for a range of ozone metrics.

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Introduction

Ground-level ozone (O_3) is a harmful air pollutant known to affect morbidity and acute mortality in the population[1] and to damage vegetation, affecting crops and forestry.

Ozone is a secondary pollutant, meaning that it is not emitted directly into the air. It occurs naturally in the earth's upper atmosphere, and concentrations in the lower troposphere result from the balance between mixing from above, chemical production, destruction and deposition at the earth's surface. Its chemical production results from chemical reactions between nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight. Concentrations are most likely to reach values harmful to health on hot sunny days, but can still reach high daytime values during colder months. Nitrogen dioxide (NO_2) is a precursor of O_3 but O_3 is consumed by reaction with nitrogen monoxide (NO). In the presence of high NO concentrations, O_3 concentration values can become very low. The removal of O_3 by reaction with NO to form NO_2 is referred to as titration. In the absence of NO, ozone has a long lifetime and can be transported over long distances in the atmosphere, affecting the air quality of areas far from the source of emissions. Because of the long-range transport impact and the highly non-linear O_3 chemistry, which vary depending on emissions, meteorological conditions and therefore geographic areas, it is particularly complicated to understand, simulate and predict O_3 concentrations. All these factors constitute a challenge when trying to identify relevant mitigation options, as ozone precursor reductions can lead to different responses in terms of ozone concentration changes.

The European Union (EU) has defined several standards, e.g. to characterise pollution episodes caused by ozone (information and alert threshold), to protect human health (long-term objective (LTO) and the target value for human health), and to protect vegetation (AOT40[1] and target value for vegetation).[2] In addition, a specific metric is calculated to evaluate the impact of O_3 on health (SOMO35).[2]

The response of ozone to precursor changes was formalised in atmospheric chemistry using the framework of chemical regimes. The atmospheric chemistry of ozone production is complex, and effective management of O_3 requires that the dependence on precursor emissions is understood. In several studies, north-western Europe is often found to be a VOC-sensitive regime, and southern Europe to be a rather NO_x-sensitive regime.

The present study aims to provide new insights into the sensitivity of ozone concentration changes to incremental reductions of anthropogenic emissions by focusing on road transport and industrial emissions.

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[1] AOT40 (Accumulated Ozone exposure over a Threshold of 40 ppb, expressed in $\mu g/m^3$ per hour) is the sum of differences between hourly concentrations greater than 80 $\mu g/m^3$ (= 40 ppb) and 80 $\mu g/m^3$ for a given period using the 1-hour values measured daily between 8 am and 8 pm.

[2] SOMO35 (Sum Of Means Over 35 ppb, expressed in ppb days) is the sum of maximum daily 8-hour averages over 35 ppb (= 70 $\mu g/m^3$) calculated for all days in a year.

To achieve this, a meta-modelling approach is used, where a full chemistry-transport model (CTM) is approximated with machine learning techniques. The surrogate model ACT, based on full CHIMERE CTM runs, is used to assess the comparative effect of emission reductions across two emission sectors: industry and road transport. By analogy with the classical ozone production isopleths of Sillman (1999) [3] where ozone concentrations resulting from incremental changes in NO_x or VOC emissions are presented, the results are presented here as isopleths of O_3 metric change on 2D charts of industrial (IND) versus road transport (TRA) emission reductions.

This methodology has enabled the production of an 'Atlas of ozone chemical regimes in Europe'[4] accounting for all non-linear processes and covering 22 European cities for a range of ozone metrics. The methodology is presented in detail on pages 17–20. The synthetic results are presented on pages 20–28, and a supplementary document including all the results for individual cities is also available.[5] The main findings are presented in the conclusions on pages 29–30.

Methodology

The CHIMERE model

The air quality simulations used for both the design and the everyday training of the ACT tool are performed with the CHIMERE CTM.[6,7] The CTM is widely used for air quality research and applications ranging from short-term forecasting to climate-scale projections. Concawe used a simulation set-up similar to the operational regional forecast performed under the Copernicus Atmosphere Monitoring Service,[3] albeit with a lower spatial resolution of 0.25 degree instead of 0.1 degree. The CHIMERE model version is CHIMERE2016a using MECHIOR gas phase chemistry, a two-product organic aerosol scheme, and ISORROPIA thermodynamics. Meteorological data are operational analyses of the IFS[4] (integrated forecasting system) model of the European Centre for Medium-Range Weather Forecasts[5] (ECMWF) at a temporal resolution of three hours. While the spatial resolution of the IFS evolves in time with subsequent upgrades of the operational production, it has always been higher than 0.25 since 2018, hence the spatial resolution of the meteorological driver is degraded prior to being used as a forcing to CHIMERE. The chemical boundary conditions are obtained from ECMWF, also with the IFS model.

Emissions

The anthropogenic emissions in the reference simulations are CAMS-REG-v3.1[8] data, which are regularly updated by the Copernicus Atmosphere Monitoring Service (CAMS). These emissions are based on the country reports of emissions required under the Convention for Long-Range Transboundary Air Pollution and collected by the Centre for Emission Inventories and Projections, which are available online. Emissions at the SNAP (Selected Nomenclature for Air Pollution) level 1 are used as input to CHIMERE. Where no emissions were available for a specific SNAP or a country, GAINS emissions were used. Improvements were also made to enhance consistency between countries, specifically on shipping

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[3] http://regional.atmosphere.copernicus.eu

[4] https://www.ecmwf.int/en/publications/ifs-documentation

[5] www.ecmwf.int

emissions and agricultural waste burning. The final step in the inventory was the distribution of the complete emission dataset across the European emission domain at 0.125° × 0.0625° longitude–latitude resolution using proxies and the E-PRTR database which provides information on the location (longitude, latitude) and emissions of major facilities in Europe. Temporal emissions profiles are taken from the GENEMIS project, and are available as data files from the the EMEP model website at www.emep.int. The vertical distribution profiles that are used for each SNAP sector are constant profiles depending only on the SNAP sector. Biogenic emissions are calculated online with CHIMERE using the MEGAN model.

The ACT model

Chemistry-transport models are needed to forecast air pollution episodes and, through sensitivity studies, to assess the benefits expected from mitigation strategies. However, they are complex, take time to run, and the number of scenarios they can compute is therefore limited. As part of CAMS that is dedicated to policymakers, INERIS has developed the Air Control Toolbox (ACT)[6] [9] to extend the number of scenarios that can be considered.

ACT is a surrogate model based on a polynomial function and trained on a dozen CTM sensitivity scenarios in which primary pollutant emissions are reduced. It is designed to be updated on a daily basis, i.e. the fitting of the parameters of the polynomial function is recalculated every day based on the scenario CTM runs. ACT is able to reproduce the non-linearity in the CTM response to changes in NO_x and VOC emissions that are important for O_3. In the present study, where annual metrics are considered, 365 individual ACT response model calculations are used to compute annual O_3 metrics. ACT is made available through a web interface and is able to produce daily metrics for defined areas within the underlying CTM model domain. The model is also designed to capture the daily means of both the PM_{10} and $PM_{2.5}$ fractions of particulate pollution and NO_2. The spatial coverage is the greater European continent.

The only two simplifications limiting the range of application of ACT are that emission reductions are assumed to apply (i) over the long term (meaning that it is not possible to investigate emergency mitigation measures, where emission reduction would only apply for a few days) and (ii) uniformly over the whole modelling domain (Europe).

ACT is configured to accept parametric emission changes in four activity sectors based loosely on the SNAP categorisation. These are:

- AGR: Agriculture (SNAP sector 10: including both crops and livestock)
- IND: Industry (SNAP sectors 1, 3, 4: Combustion in energy and transformation industries, combustion in manufacturing industry, and Production processes)
- RH: Residential heating (SNAP sector 2: Non-industrial combustion plants)
- TRA: Road transport (SNAP sector 7: urban and non-urban roads and motorways)

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[6] https://policy.atmosphere.copernicus.eu/documentation/act.php

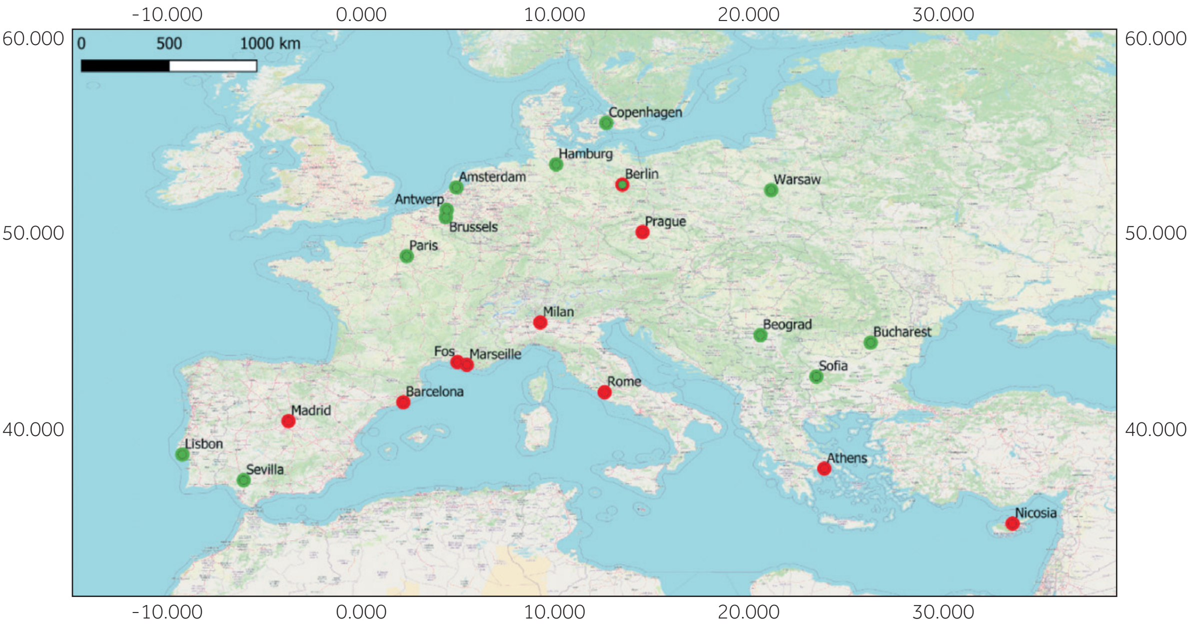
The surrogate ACT model trained on CHIMERE sensitivity simulations also allows exploring the chemical sensitivity (or regimes) within the parameter space of sectoral emission reductions. ACT is a quadrivariate second order polynomial with interactions using as predictors the four sectors considered. By plotting the surface response to two of these four sectors in a 2D parameter space, it is possible to assess chemical regimes for a given day, location and pollutant. In doing so, an analogy with the classical ozone production isopleths of Sillman (1999)[3] is performed, by substituting the NO_x and VOC emissions in the x and y axes by different activity sectors. Here, the focus is on the industrial (IND: as SNAP 1, 3 and 4) and road transport (TRA: as SNAP 7) activity sectors.

The choice of cities

Twenty-two European cities were chosen to be representative of different meteorological conditions (ranging from southern to northern Europe), different O_3 regimes and different emission profiles. The set of selected cities is shown in Figure 1. The situation of the cities relative to the target value for human health (the maximum daily 8-hour mean may not exceed 120 µg/m^3 on more than 25 days) and vegetation (AOT may not exceed 18,000 µg/m^3 per hour) for the year 2019 is represented by coloured circles, with red for annual exceedances and green to indicate compliance with the target values. The cities exposed to exceedances of the EU target values are mainly Mediterranean cities that receive large amounts of solar radiation.

Figure 1: Cities selected for the 'Atlas of ozone chemical regimes'

Data from the EEA's 'AQ eReporting' statistics for 2019 (https://www.eea.europa.eu/data-and-maps/dashboards/air-quality-statistics)



For each city, compliance with the human health target value for the year 2019 is represented by a large green circle, and compliance with the target value for vegetation by a small green circle. In contrast, a large red circle is used when the target value for health is not met, and a smaller red circle for the target value for vegetation. See also the city characteristics in the *Supplementary Material*.[5]

Metrics, period and classification

The ACT tool explores the response, in terms of the ozone metric, to emission reductions ranging from 0 to 100%. The model can consider emission reductions for four sectors, but the focus of this study is on the reduction of emissions from the industrial and road transport sectors (referred to as IND and TRA, respectively, in the remainder of this article). Emissions from agriculture (AGR) and residential heating (RH) are held constant.

For each city, isopleths are established for the change in ozone metric drawn on charts on which the axes represent emission reductions applied to the TRA and IND sectors.

Results

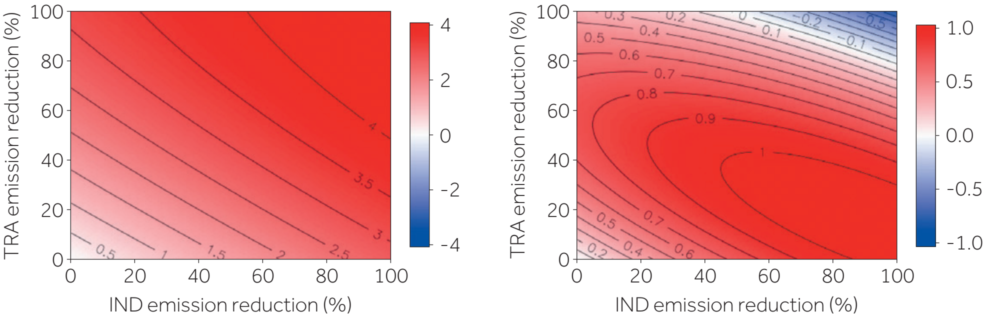
Examples of O_3 regimes and isopleths

For producing isopleths, emissions from traffic and industry are each reduced from 0 to 100% (with a 1% reduction step, this amounts to studying the distribution of indicators according to 10,000 reduction scenarios) and the resulting change in O_3 is calculated. All the isopleths produced for the different O_3 metrics, seasons and cities are provided in the *Supplementary Material*.[5] They all represent the difference between the value of a metric after the application of an emission reduction and the value without any reduction. This difference, referred to as ΔO_3 hereafter, is negative (in blue) if the metric has decreased as a result of emission reductions, and positive (in red) if the metric has increased. Figure 2 on page 21 presents some typical examples of O_3 regimes and the associated isopleths for illustration purposes (the full set of results are provided in the *Supplementary Material*.[5]

When the isopleths are completely red, it means that, whatever the reduction of emissions from road transport and industry is, the ozone metric values are increasing rather than decreasing: this is a case of O_3 titration. Conversely, a blue isopleth means that the emission reductions are indeed reducing ozone. The importance of this reduction can be read directly on the isopleths; this is shown in Figure 2 as a % reduction of the ozone metric. These isopleths also enable an assessment of whether industrial emission reductions allow a greater reduction of ozone than road transport emission reductions, and vice versa, depending on the slope of the isopleths. In the examples from the *Supplementary Material* presented on pages 26–28 it can be seen that the set of isopleths can be classified into six different classes in terms of chemical regimes.

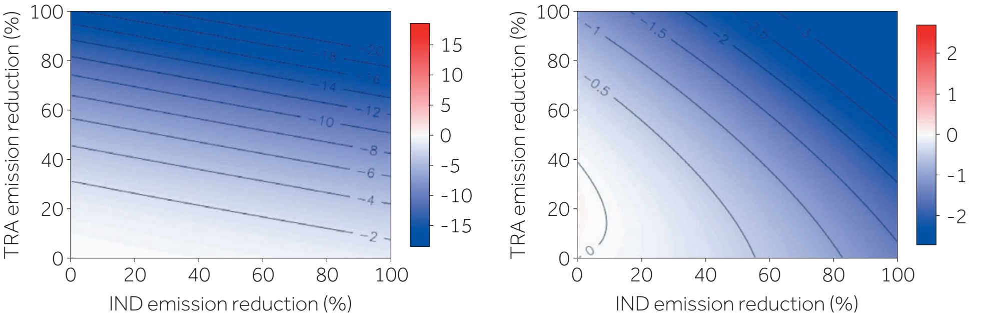
In winter, a complete titration regime is found for all cities except Nicosia. Indeed, in winter, solar radiation is much lower at the zenith than in summer, and the nights are longer. O_3 production is therefore low and O_3 is mainly consumed by its reaction with NO. A decrease in NO emissions (from IND or TRA) will therefore lead to less O_3 destruction, and in most cities will effectively result in an increase in O_3. The largest wintertime O_3 increase is simulated for Milan, with a median daily max O_3 increase of 26% (i.e. 9 $\mu g/m^3$), and a maximum of 66% (for 100% reduction of IND and TRA emissions). However, this increase in O_3 is tempered by the fact that O_3 values in Europe are low in winter with very few exceedances of the 120 $\mu g/m^3$ threshold.

Figure 2: Examples of O_3 isopleths and O_3 regimes for different cities, O_3 metrics and periods (reference scenario)



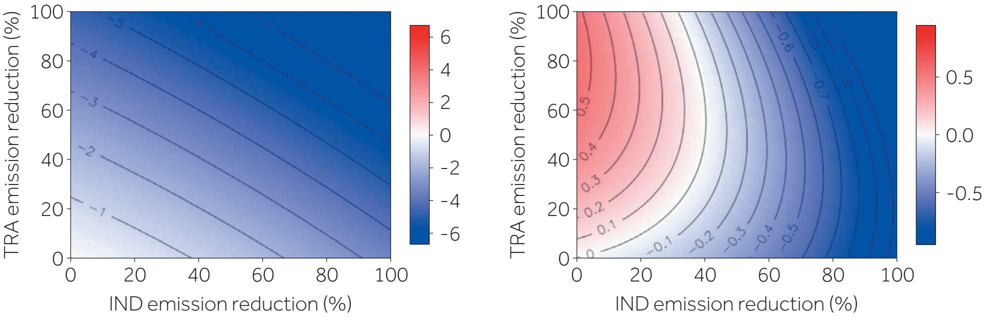
(a) **Complete titration regime**
Brussels, O_3 maximum annual average, 2019 (%)

(b) **Partial titration regime**
Brussels, SOMO35, 2019 (%)



(c) **TRA sensitive**
Milan, summer O_3 maximum average, 2018 (%)

(d) **IND sensitive**
Hamburg, SOMO35, 2019 (%)



(e) **Both TRA and IND sensitive**
Nicosia, O_3 maximum annual average, 2019 (%)

(f) **Change in regime**
Copenhagen, SOMO35, 2019 (%)

Some cities are not only in a titration regime in winter, but also show titration or very low reduction of O_3 for the summer average of the daily maximum and SOMO35 indicator; these are Paris, Antwerp, Brussels, Amsterdam and Copenhagen. However, in such cases the failure or ineffectiveness of emission reductions in lowering ozone levels must be put into perspective, as target values for health and vegetation are not exceeded in these cities.

For the annual average O_3 daily maximum metric, only Beograd, Nicosia, Bucharest, Sofia, Seville and Rome show O_3 reductions whatever the emission reductions are. But even for those cities, the reduction in the O_3 metric is limited to 4% for the median reduction and 13% for the maximum. Emissions reductions are slightly more efficient when considering the annual metric SOMO35 that does not take into account O_3 concentrations lower than 70 µg/m^3. In particular, for the cities of Barcelona, Milan, Copenhagen, Berlin and Hamburg, emission reductions do lower SOMO35 in most cases, while their annual average ozone levels tend to rise as a result of these emission reductions.

Summer is the period for which O_3 reductions associated with emission reductions are greatest, due to the large amount of O_3 production at this time. The largest reductions are found in Rome, Milan, Madrid, Prague, Bucharest, Fos-sur-Mer, Sofia and Seville, with for example a median reduction of 11% (-16 µg/m^3) in Milan in summer 2019. For the large majority of summer isopleths, this median level is obtained for TRA and IND emissions reductions larger than 50% (see the *Supplementary Material*[5]). When TRA and IND emissions are reduced by 100%, the highest summer reductions occur in Milan, and can reach -32% (50 µg/m^3) in Milan during summer 2019. However, in the majority of the cities examined, the highest reductions do not exceed 20%. O_3 reductions associated with the annual metric SOMO35 are half way between those simulated for the summer average of the daily maximum O_3 and for its annual average, with O_3 reductions in the majority of cities but limited to 5% for the median reduction and 15% for the maximum.

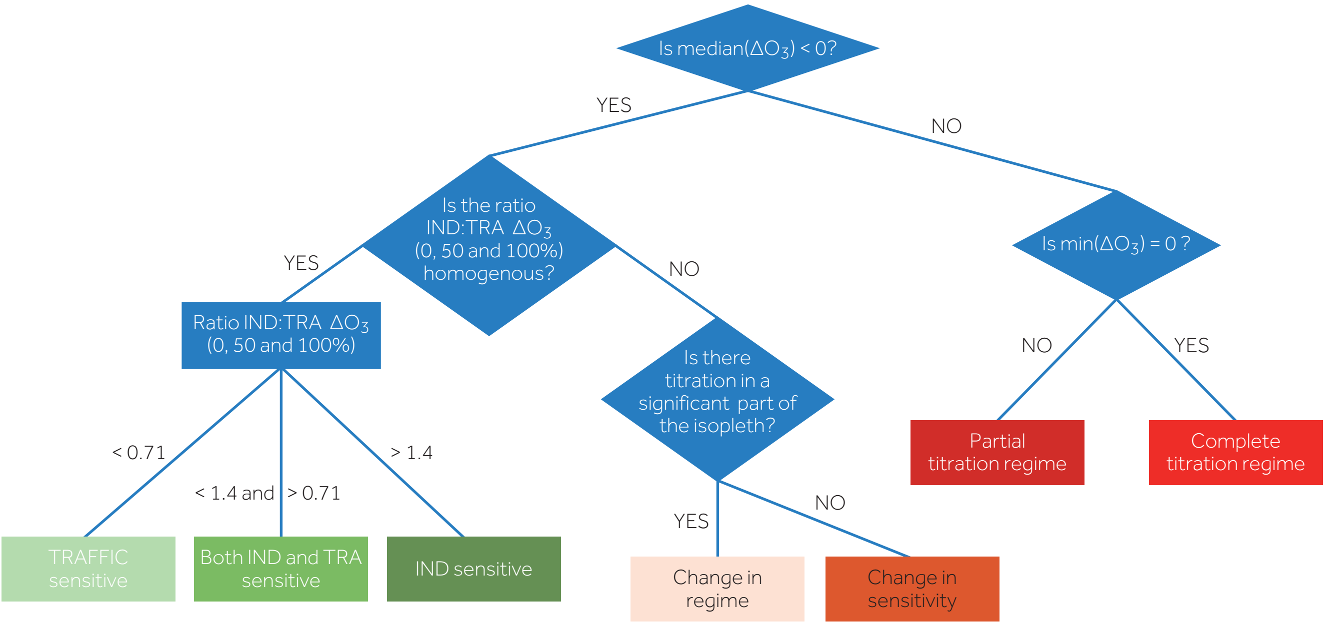
Ozone regimes

The set of isopleths for all ozone metrics, cities and periods studied have been classified into six different O_3 classes in terms of chemical regimes:

1. Titration regime (complete or partial): reductions in emissions (IND or TRA or both) lead to an increase in the O_3 metrics (positive ΔO_3). This can be the case for any reduction (complete titration regime) or only for some part of the IND:TRA reduction space (partial titration regime).
2. TRA sensitive: reductions in road transport emissions produce a greater reduction in the considered O_3 metric than that produced by reductions in industrial emissions.
3. IND sensitive: reductions in industrial emissions produce a greater reduction in the considered O_3 metric than that produced by reductions in road transport emissions.
4. TRA and IND sensitive: road transport and industrial emission reductions have a similar impact on the considered O_3 metric.
5. Change in regime: an increase in the O_3 metric occurs in a part of the IND:TRA reduction space, and a decrease in the O_3 metric occurs elsewhere.
6. Change in sensitivity: there is a clear shift from a regime that is sensitive to road transport emissions reductions to a regime that is sensitive to industrial emissions reductions (or the reverse). This case was not encountered in the cities and over the period selected.

An example of each ozone regime is given in Figure 2 on page 21. The procedure used to classify the O_3 regime results for each city is described below and presented in Figure 3.

Figure 3: Representative flow chart of the regime classification based on the median and minimum ΔO_3, and the ratio between O_3 responses to road transport and industrial emissions reductions



A value of the median $\Delta O_3 > 0$ indicates a titration regime. This is classified as a:

- complete titration regime if the minimum ΔO_3 value is = 0; and
- partial titration regime if this minimum value is < 0.

A value of the median $\Delta O_3 < 0$ indicates that reducing IND or TRA emissions yields some benefit in reducing ozone concentrations. The response can, however, be quite different depending on targeted cities, ozone metrics, or selected year/period. This response was therefore subsequently classified as one that explicitly occurs if the sensitivity was mainly attributed to IND, TRA or both IND and TRA, if it changes with sensitivity regime, or if some part of that response still exhibited a titration regime.

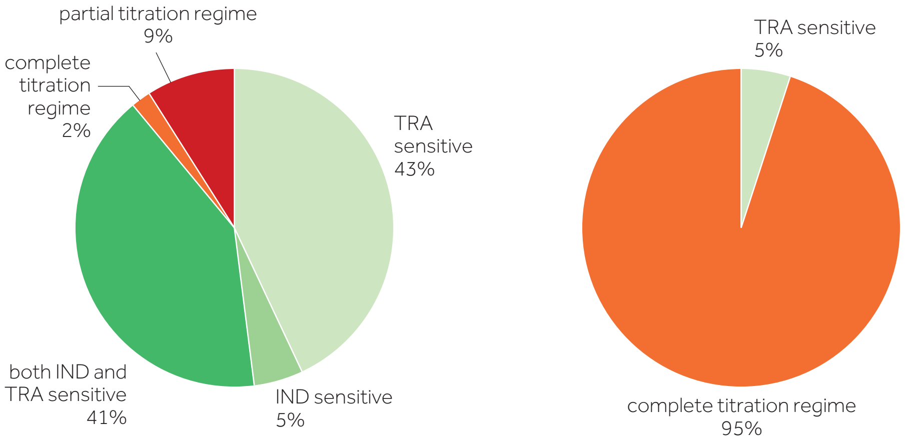
Figure 4 on page 24 clearly shows the differences between the periods (summer, winter, yearly average) and the O_3 metrics in terms of classification of ozone regimes.

Figure 4: Summary classification of ozone regimes for different ozone metrics over the 22 target cities

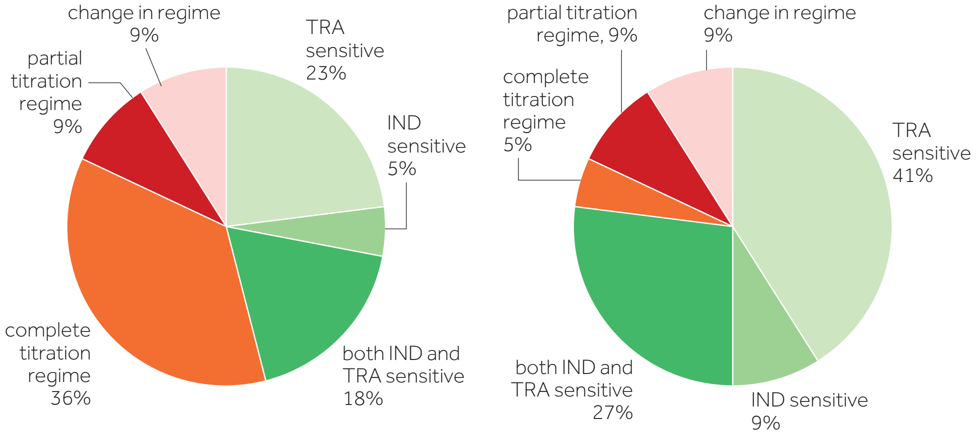
Each pie chart indicates the distribution of regimes across the 22 selected cities:

- 🟩 TRA sensitive
- 🟩 IND sensitive
- 🟩 IND and TRA sensitive
- 🟧 complete titration
- 🟥 partial titration
- ▨ change in regime

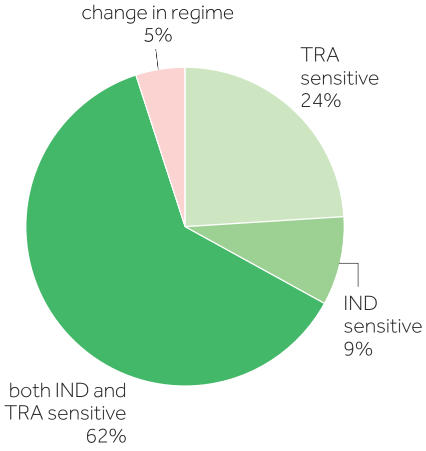
(a) Summary average of the daily maxima, 2018 and 2019 (b) Winter mean of the daily maxima, 2019



(c) Annual mean of the daily maxima, 2019 (d) SOMO35, 2019



(e) Percentile 93.15 of the daily maxima, 2019



In summer (Figure 4a), the titration regime is marginal as it occurs only in 11% of the target cities. For 43% of the target cities, the average summertime daily maximum O_3 is reduced more by road transport emissions reduction than by industrial emissions reduction, compared to 5% having a higher sensitivity to industrial emissions reduction. A large fraction (41%) is sensitive to emissions reductions in both the industrial and road transport sectors.

In winter (Figure 4b), almost all target cities show a complete titration regime with daily maximum O_3 concentrations increasing for all emission reductions.

The annual average of O_3 daily maxima (Figure 4c) shows a behaviour between the two extremes shown for summer and winter. It can be seen that 45% of the target cities are in a titration regime (partial or complete), 23% are TRA sensitive, 5% IND sensitive, and 18% are both TRA and IND sensitive. In addition, 9% of the target cities are classified as 'change in regime', meaning that titration is observed for a significant part of the IND:TRA emissions reduction space, but the regime changes to an O_3 net decrease when emission reductions reach a higher level.

For SOMO35 (Figure 4d), the number of cities displaying a titration regime is logically lower than for the annual mean because of the definition of the SOMO35 metric. Indeed, the effect of the titration is the consumption of O_3, resulting in lower O_3 concentrations. For SOMO35, being the sum of the maximums of O_3 over 8 hours that are higher than 70 $\mu g/m^3$, the days of strong titration are not counted in the calculation of SOMO35. The proportion of cities that show greater sensitivity to IND than TRA reductions for SOMO35 is slightly greater (at 9%) than for the other metrics. The Figure shows that 41% of the target cities are TRA sensitive, and 27% are both TRA and IND sensitive.

The last indicator studied is the percentile 93.15. On this high ozone peak indicator, the majority of cities are both TRA and IND sensitive (62%). Around 24% of the cities are TRA sensitive and 9% are IND sensitive.

Overall, partial or complete titration regime aside, most indicators are either equally sensitive to traffic and industrial emission reductions, or more sensitive to traffic emission reductions. Some cities are more sensitive to reductions in industrial emissions, but not necessarily on all indicators (e.g. on SOMO35 but not on percentile 93.15): these include Madrid, Hamburg, Copenhagen, Lisbon, Warsaw and Beograd.

Some cities have been identified here as being in a titration regime, or showing very low O_3 reductions when reducing road transport and industrial emissions; this is the case for all O_3 metrics. These are Paris, Antwerp, Brussels, Amsterdam and Copenhagen. When only considering the annual average O_3 maximum metric, the list also includes Berlin, Warsaw, Hamburg, Barcelona and Milan.

The cities showing the largest relative reduction in the annual average O_3 maximum metric when reducing road transport and industrial emissions are Bucharest, Belgrade, Nicosia, Rome, Sofia and Seville.

Milan shows a very different behaviour depending on which O_3 metric is considered: it is one of the cities showing the largest relative reduction for SOMO35 but, when looking at the annual average O_3 maximum, it shows a titration regime. For the summer O_3 metrics, Milan and Rome are clearly the cities with the largest relative reduction, followed by Bucharest, Seville, Fos-sur-mer, Sofia, Nicosia, Madrid and Prague.

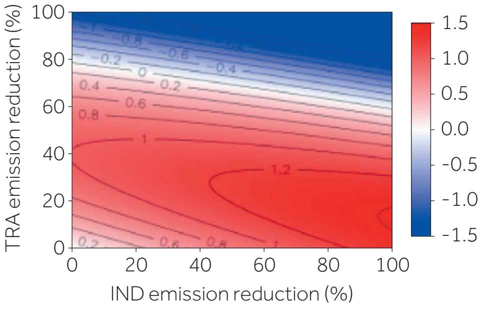
Factors influencing the differences in O_3 regimes between cities (e.g. meteorological factors, emissions speciation factors) are analysed in depth in the 'Atlas of ozone chemical regimes in Europe'.[4]

Examples from the *Supplementary Material*[5]

Milan

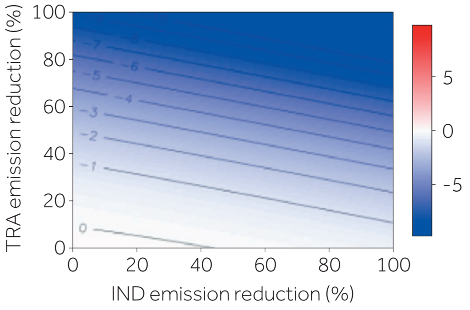
(a) Annual maximum average, 2019

Milan, O_3 maximum annual average, 2019 (%)



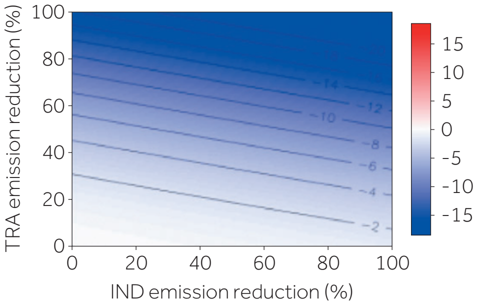
(b) SOMO35, 2019

Milan, SOMO35, 2019 (%)



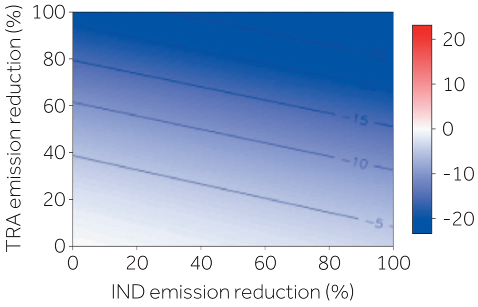
(c) Summer maximum average, 2018

Milan, summer O_3 maximum average, 2018 (%)



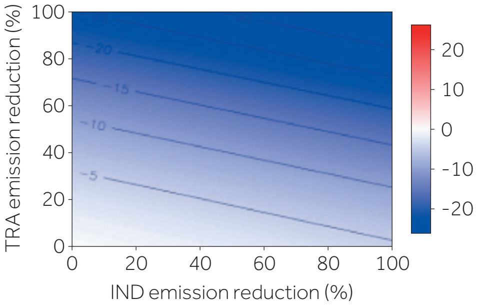
(d) Summer maximum average, 2019

Milan, summer O_3 maximum average, 2019 (%)



(e) Percentile 93.15, 2019

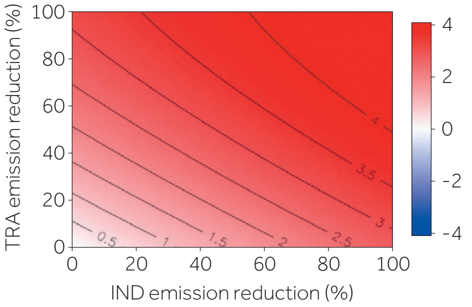
Milan, O_3 93.15 percentile, 2019 (%)



Brussels

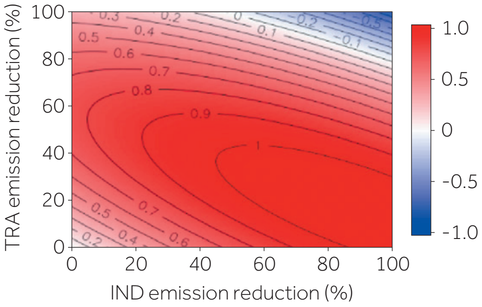
(a) Annual maximum average, 2019

Brussels, O_3 maximum annual average, 2019 (%)



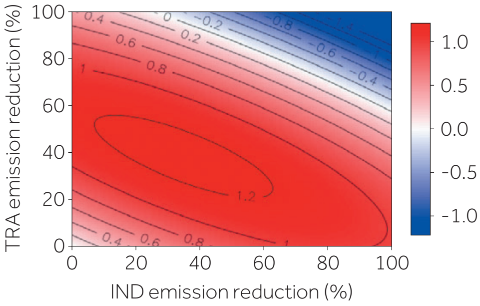
(b) SOMO35, 2019

Brussels, SOMO35, 2019 (%)



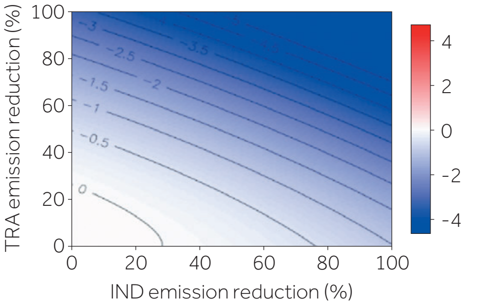
(c) Summer maximum average, 2018

Brussels, summer O_3 maximum average, 2018 (%)



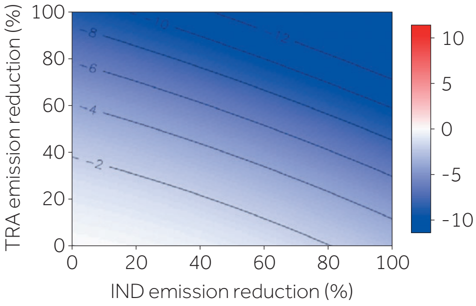
(d) Summer maximum average, 2019

Brussels, summer O_3 maximum average, 2019 (%)



(e) Percentile 93.15, 2019

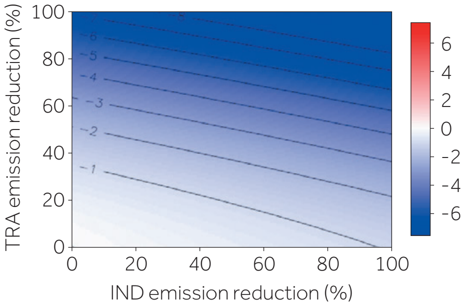
Brussels, O_3 93.15 percentile, 2019 (%)



Rome

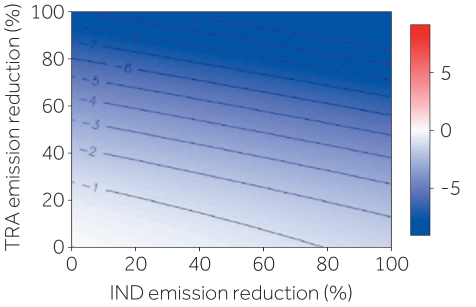
(a) Annual maximum average, 2019

Rome, O_3 maximum annual average, 2019 (%)



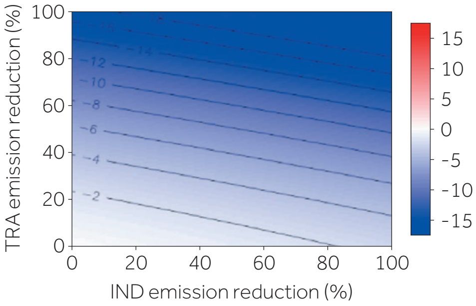
(b) SOMO35, 2019

Rome, SOMO35, 2019 (%)



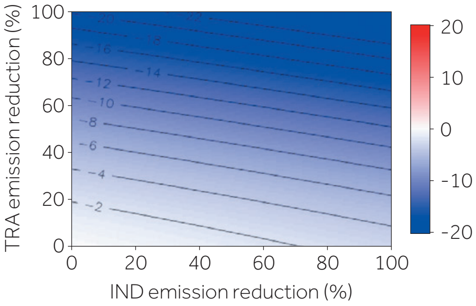
(c) Summer maximum average, 2018

Rome, summer O_3 maximum average, 2018 (%)



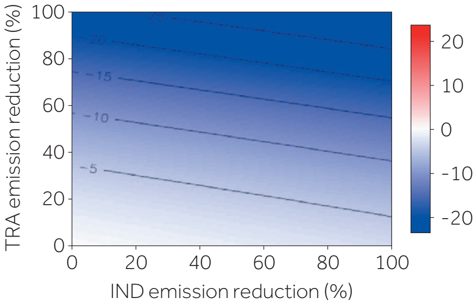
(d) Summer maximum average, 2019

Rome, summer O_3 maximum average, 2019 (%)



(e) Percentile 93.15, 2019

Rome, O_3 93.15 percentile, 2019 (%)



Conclusions

The Air Control Toolbox, ACT, is a surrogate model trained on the full chemistry-transport model CHIMERE that allows capturing the effect of a wide range of emission reductions in the road transport, industrial, residential and agricultural sectors on ozone, NO_2 and particulate matter.

In this study, ACT was used to examine the change in surface ozone that might be brought about through reductions in emissions from road transport and a combination of industry sources represented by two pseudo-categories: road transport (TRA) and industry (IND). Both TRA and IND are associated with NO_x and non-methane VOC (NMVOC) emissions, the amounts varying city by city. The results of these calculations have been presented as an atlas of two-dimensional emission reduction charts showing ozone metric changes (ΔO_3) as isopleths.

A total of 22 target cities across Europe were selected and O_3 changes analysed for the years 2018/2019. The results have been supplemented with information on O_3 regime, meteorological parameters and emissions information. Focus was on three metrics for O_3: the daily 1-hour maximum averaged over a season; SOMO35, a health metric; and the 93.15 percentile of the daily maximum O_3 concentrations, corresponding to the 26th highest O_3 concentration (not to exceed the EU target value of 120 µg/m^3). The results have been expressed as a change in O_3 metrics (ΔO_3) with change in emissions. Detailed results are available in the *Supplementary Material*.[5]

The ΔO_3 charts were classified into six O_3 classes in terms of chemical regimes. The O_3 sensitivity to road transport and industrial emissions differ from one city to another, but also for the same city when considering the different ozone metrics and from one period of the year to another (winter vs summer), or even from one year to another (2018 vs 2019).

Six classes in terms of chemical regimes are considered in the analysis: either (i) road transport (TRA) or (ii) industry (IND) if emission reductions for one of those activity sectors is found to lead to ozone reductions. Sensitivity to both IND and TRA is considered as an individual class (iii). A fourth class differentiates the cases where TRA and/or IND emission reduction yields an increase in ozone metrics (referred to as partial or complete titration regimes (iv)). A final class is where the model indicates that both increases and decreases in ozone occur over the range of emission reductions (referred to as change in regime (v)). A sixth class was also considered which would have involved switching from a TRA-sensitive regime to an IND-sensitive regime (referred to as a change in sensitivity (vi)); however, no cases were found in the cities studied.

The proportion of cases (city/period/metrics) for which the O_3 regime is a titration regime is significant, especially in winter (96%) and for the annual average of the O_3 daily maximum (45%). This is particularly the case for northern European countries with low solar radiation (and thus low O_3 production) but also for some countries further south but with high NO_x emissions at local and/or regional scale. In these cases, measures to reduce NO_x emissions are counterproductive for reducing O_3. Ozone titration (i.e. counterproductivity of NO_x reduction measures) is not observed at very high O_3 levels, since the principle of titration is consumption of O_3 by its reaction with NO. That is why reduced titration, which leads to an increase of ozone, is essentially a concern where and when ozone concentrations are low in the reference case and the EU target values are not reached.

The greater the focus on summertime months, and on yearly indicators with a high threshold, the more effective emission reductions can be, and the fewer cases of titration there are. This is because emission reductions mainly reduce the high ozone peak when daily averaged O_3 can be increased due to a lower impact of titration. The cases of complete titration decrease significantly to 2% and 4%, respectively, when considering the summer period and SOMO35 compared to the winter case (96%). For about 10% of the cities, the regime is a partial titration regime, i.e. emission reductions will primarily contribute to increasing O_3 but when high emission reductions are assumed, O_3 reductions are predicted. For the remaining cases (more than 75% of the cities) the emission reductions from road transport and industry are expected to reduce O_3 metric values, but this reduction is limited with a maximum reduction of summer average daily maximum of 32% in Milan assuming the elimination of both IND and TRA emissions. This is a fairly limited O_3 reduction in comparison to the major reduction in emissions (100%). For other cities in summer (2019) O_3 maximum reductions are more in the range of 20–25%, so even less responsive to major reductions in road transport and industrial emissions. The indicator most sensitive to emission reductions is the percentile 93.15 with median reductions ranging from 3% to 13% and a maximum reduction of 37% for Milan. Emission reductions are never counterproductive for this indicator, except in Paris for low emission reductions. Moreover, in all cities, significant improvements in attaining the European target value was shown with the associated emissions reductions. This study therefore suggests that reducing ozone precursor emissions from the traffic and industrial sectors may have counterproductive effects on certain ozone indicators, but is unlikely to lead to exceedances of the current target value; on the contrary, it may reduce the number of exceedances if the emissions reductions are significant.

NO_x and NMVOC emissions from other sectors have not been reduced in this study. Inventories show higher NMVOC from solvent use than from TRA and IND in several cities. Biogenic VOC emissions also contribute to ozone production.

The cities that show the largest relative O_3 reductions are southern European cities where either NO_x emissions are not too high, or which have high NO_x emissions but also high VOC emission levels. Climatic conditions favour O_3 production, particularly the amount of solar radiation received and the propensity for stagnation of air masses, for which annual average wind speed was used as a surrogate.

Outside the titration regime, most cases show a higher sensitivity to emission reductions from road transport or equal sensitivity to emission reductions from road transport and industry. Very few cases are most sensitive to emission reductions from the industrial sector.

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