Review

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Foreword

This *Review* is focused on air quality.

The first and third articles in this *Review* summarise studies conducted in collaboration with TNO, which analyse the influence that some sectors have on the ambient air quality in cities. The first article assesses the influence of international and inland shipping activity on atmospheric air quality in 19 European port cities, while the influence of aviation activity, with a focus on six European cities with large airports in their vicinities, is the theme of the third article.

The second article throws some light on ozone, a pollutant with complex chemistry that makes it difficult to identify how its concentrations can be mitigated. The article focuses on the 'Atlas of ozone chemical regimes in Europe', developed in cooperation with Ineris, for which changes in ozone metrics as a result of reductions in road transport and industrial emissions have been simulated for 22 European cities.

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Contents

This article presents results from a modelling study carried out to assess the influence that shipping emissions have on the air quality in European port cities. Using a 3-D chemical transport model (LOTOS-EUROS) and its source apportionment capabilities, the contribution of international and inland shipping emissions to atmospheric air pollutant concentrations in 19 European port cities is examined.

In the emission set used in the study, the shipping emissions on seas are derived from Automatic Identification System (AIS) data of all ships sailing in the geographic domain of the calculations. These are considered as more robust compared to the emissions reported by Member States to the European Environment Agency (EEA) as a result of the restrictive definition of maritime emissions in the national inventories which do not include any shipping emissions outside the territorial waters of the Member States.

The results show that international shipping contributes significantly to NO_2 concentrations on average over Europe (~18%), while locally in the seaports, the contribution is even higher (e.g. up to 60% in Rotterdam). The contribution remains significant in all cities located near seaports and, in many cases, international shipping is the dominant contributing source. In contrast, inland shipping generally has a low influence on NO_2 concentrations, with only a few exceptions (i.e. Rotterdam, Amsterdam). The contribution of shipping emissions for other pollutants is lower but still noteworthy (11% for SO_2, 2.5–5% for $PM_{2.5}$/PM_{10} on average over Europe).

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While concentrations of most air pollutants have been decreasing in Europe over the past 20 years, ozone (O_3) is showing variable trends, with increasing average concentrations and decreasing peak ones. The complexity of O_3 chemistry adds to the difficulty of understanding both the trends observed and how concentrations can be mitigated. This article tries to answer the following questions: which emission sectors should be targeted, and what levels of reduction could be achieved? To address these reflections, an 'Atlas of ozone chemical regimes in Europe' has been compiled. For this Atlas, 22 European cities were selected and the surrogate model Air Control Toolbox (ACT) was used to evaluate the simulated changes in several ozone metrics as a result of reductions in road transport and industrial emissions. Ozone chemical regimes have been classified and put in perspective with meteorological and emissions data in and around each city location. The O_3 sensitivity to road transport and industrial emissions differ from one city to another, but also for the same city when considering different ozone metrics and seasons (e.g. annual means versus SOMO35 or summer peaks). Counterproductive impacts yielding an increase in O_3 when emissions are reduced are mainly encountered in regions or during periods where O_3 concentrations are relatively low. In terms of meteorological factors, O_3 chemical regimes are mostly impacted by the amount of solar radiation received, but wind speed also has a considerable impact. Most cases show a higher sensitivity to reductions in road transport emissions, or equal sensitivity to emission reductions from road transport and industry. Very few cases are most sensitive to emissions from the industrial sector. However, the response of annual or seasonal average O_3 metrics to industrial and road transport emissions can be considered relatively low with a maximum reduction of 33% for a 100% reduction of both industrial and road transport emissions. This is because anthropogenic emissions can only mitigate ozone above a substantial natural tropospheric background. But it is precisely this incremental anthropogenic ozone that should be targeted by efficient policies in order to achieve significant improvements in attaining the European target values in all of the cities studied.

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The impact of shipping emissions on urban air quality in Europe — a port/city analysis

This article summarises the results of a modelling study undertaken to assess the influence that shipping emissions have on air quality in European port cities. The study compared the influence of both international and inland shipping on NO_2 concentrations both in seaports and in cities located close to the ports. It found that international shipping contributes significantly to NO_2 concentrations in Europe, while inland shipping generally has a low influence on NO_2 with only a few exceptions.

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Introduction

It is well known that elevated concentrations of atmospheric pollutants can lead to adverse effects on both human health and ecosystems. Epidemiological studies have shown that exposure to pollutants such as fine particulate matter ($PM_{2.5}$) and nitrogen dioxide (NO_2) is associated with cardiovascular and respiratory diseases, leading to increased sickness, hospital admissions and premature death.[1] Moreover, nitrogen deposition in soils and water bodies leads to eutrophication and biodiversity loss, and sulphur dioxide can contribute to acidification which can harm sensitive ecosystems.

Over the past decades, legislation has been introduced to reduce emissions of these harmful pollutants. These efforts to reduce emissions in several sectors have resulted in a decrease in the atmospheric concentrations of $PM_{2.5}$, PM_{10}, NO_2 and SO_2. The most recognised example of successful emission reduction is in SO_2. Due to abatement measures in power plants and desulphurisation of fuels, the atmospheric SO_2 concentration in European Union countries has declined by around 70% between 2000 and 2017[2] based on aggregated observations.

The strong emissions reductions in some sectors have inevitably shifted the focus to other sources with lower contributions, such as shipping for which the relative emissions contributions have a growing significance in terms of further reducing air pollutant concentrations.[3,4]

In this study, insights are gained with respect to the influence of shipping emissions on air quality in Europe and in major ports and cities. Using the chemical transport model (CTM) LOTOS-EUROS and its source apportionment capabilities, the contribution of international and inland shipping emissions to atmospheric air pollutant concentrations in 19 European port cities is computed and put into context compared to the relative contribution of other sources.

The methodology used in the study is described in the following section, which provides details about the model and the data that are used as input to perform the simulations of the atmospheric concentrations. The results of the study are presented on pages 8–13. The CTM provides labelled atmospheric concentrations over the simulation domain. Using the simulation results, the contributions of various sectors to air quality in port cities of interest are computed. The main findings are presented in the final section of this article.

Methods

Model description

LOTOS-EUROS is a 3-D chemical transport model developed by TNO. The offline Eulerian grid model simulates air pollution concentrations in the lower troposphere, solving the advection-diffusion equation on a regular latitude-longitude grid with a variable resolution over Europe.[5]

The vertical transport and diffusion scheme accounts for atmospheric density variations in space and time and for all vertical flux components. The vertical grid is based on terrain following vertical coordinates and, excluding the stacked boundary layers on top, extends to 5 km above sea level. The height of the layers on top of the 25-metre surface layer is determined by heights in the meteorological input data.

Gas-phase chemistry is simulated using the TNO CBM-IV scheme, which is a condensed version of the original scheme.[6] The LOTOS-EUROS model explicitly accounts for cloud chemistry, computing sulphate formation as a function of cloud liquid water content and cloud droplet pH as described in Banzhaf *et al.* (2012).[7] For aerosol chemistry the thermodynamic equilibrium module ISORROPIA II is used.[8] Dry deposition fluxes are calculated using the resistance approach as implemented in the DEPAC (DEPosition of Acidifying Compounds) module.[9] Furthermore, a compensation point approach for ammonia is included in the dry deposition module.[10] The wet deposition module accounts for droplet saturation following.[11]

The model is part of the Copernicus Atmospheric Monitoring Service (CAMS) regional ensemble providing operational forecasts and analyses over Europe. In this context the model is regularly updated and validated using observations from ground and satellite observations. The model performance is also subject to numerous peer-reviewed publications.[12,13,14]

Source apportionment

TNO has also developed a system to track the impact of emission categories within a LOTOS-EUROS simulation based on a labelling technique.[15] This technique provides more accurate information about the source contributions than using a brute force approach with scenario runs as the chemical regime remains unchanged. Another important advantage is the reduction of computational costs with respect to the brute force approach. The source apportionment technique has been used extensively in previous studies.[14,16,17]

As well as calculating the total concentrations of each pollutant, the contributions of selected sources to these concentrations are calculated. The labelling routine is implemented for primary, inert aerosol tracers as well as for chemically active tracers containing a C, N (reduced and oxidised) or S atom, as these are conserved and traceable.

Emissions and meteorology

The LOTOS-EUROS model is run with ECMWF ERA5 reanalysis meteorological data (2018). ERA5 provides hourly estimates of a large number of atmospheric, land and oceanic climate variables, that are necessary inputs for calculations of atmospheric concentrations. Examples of typical inputs required by LOTOS-EUROS include surface and air temperature, cloud cover, wind speed and direction, precipitation and relative humidity.

Quality-assured monthly updates of ERA5 (1959 to present) are published within three months of real time and are available through the Climate Data Store.[1] Preliminary daily updates of the dataset are available to users within five days of real time.

The CAMS-REG emission inventory data for the year 2018, version 5.1 REF2,[18] was used in this study for anthropogenic trace gas emissions. At the time of performing the study, this was the latest available dataset (an update with more recent data was published in December 2023). The inventory uses the official emissions data reported by European countries. However, for international shipping, the dataset is replaced with emissions from the Finnish Meteorological Institute (FMI) STEAM model.[19] Additional information that explains the choice of using the STEAM model to create the shipping emission inventory instead of using the officially reported European emissions data is provided below.

The importance of the emission inventory

As mentioned above, the CAMS-REG emission inventory data for the year 2018, version 5.1 REF2,[18] is used in the air quality calculations as the latest available dataset. Generally, the CAMS-REG emissions datasets are based on the officially reported emissions from the EU Member States.

To ensure robustness in the air quality calculations, it is necessary that the emissions of all sources are geographically distributed in an accurate way. However, according to reporting conventions[20] the inventory totals as reported by countries do not contain all shipping emissions (and they are not geographically referenced); only the emissions from shipping between the national harbours are accounted for in the national emission totals. The emissions from seagoing shipping leaving or coming from another country are accounted for in a so-called memo item, 'International maritime navigation'.[21] These emissions (which are commonly calculated on bunker sales) cannot be attributed to a specific country as the emissions take place at sea in international waters. The emissions included in this memo item from national inventories cannot be used in air quality calculations as the location where the emissions occur is not known (not geographically referenced). In addition, the methodology used to calculate and report inland shipping in the different Member States is not harmonised, and thus not always comparable. In most countries it relies on national fuel statistics, which do not differentiate between fuel use for inland shipping and national seagoing shipping. In those cases, international inland shipping emissions might be included in national navigation or international inland waterways as it is not possible to calculate the split between inland and maritime use.

In order to overcome these inconsistencies and increase the accuracy of the emissions dataset used, in the CAMS-REG dataset, the emissions from all seagoing vessels as reported in the national inventories are replaced with emissions from the Finnish Meteorological Institute (FMI) STEAM model.[19] This model is based on actual ship movements as registered by the Automatic Identification System (AIS) data and, moreover, they are geographically referenced. This model gives the best geographical distribution of the shipping emissions on European seas (and the Atlantic). For inland shipping the data reported in the national inventories are complemented by the spatial distribution of the emissions as calculated by the STEAM model.

1 https://cds.climate.copernicus.eu/#!/home

Model set-up

Figure 1 shows the different domains which are part of the LOTOS-EUROS simulations. A coarse resolution (circa 25 x 25 km) simulation is performed over Europe (domain shown in blue). Results from this simulation are used as boundary conditions for two nested simulations over the Mediterranean and a central part of Europe (domains shown in red) at a higher resolution (circa 6 x 6 km), covering the following ports that were studied in more detail:

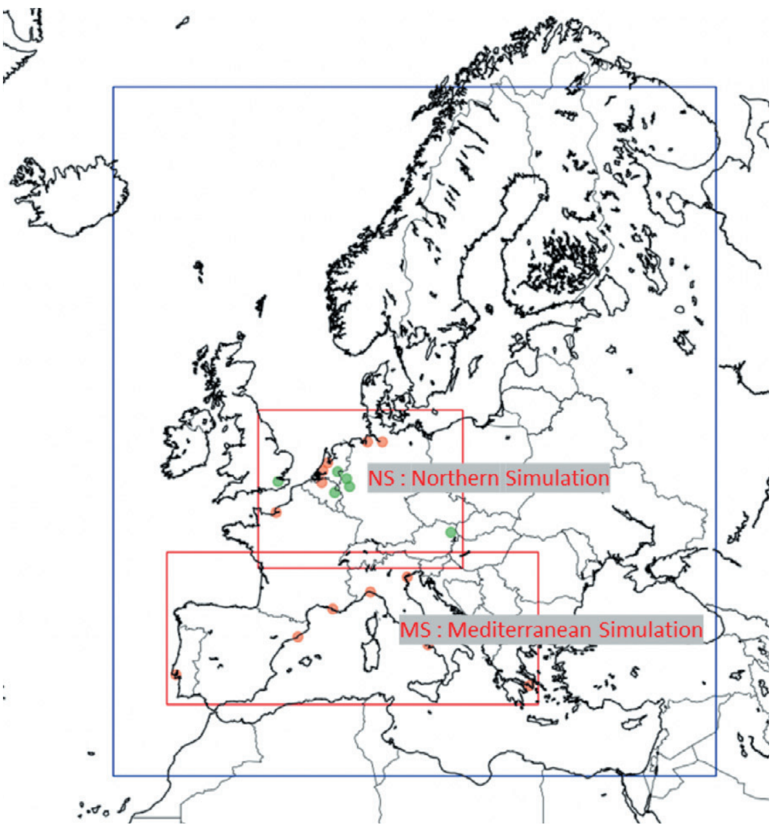
Seaports:

1. Rotterdam (NL)
2. Antwerp (BE)
3. Hamburg (DE)
4. Amsterdam (NL)
5. Marseille (FR)
6. Bremerhaven (DE)
7. Barcelona (ES)
8. Le Havre (FR)
9. Genoa (IT)
10. Piraeus (GR)
11. Lisbon (PT)
12. Naples (IT)
13. Venice (IT)

Inland ports:

1. Vienna (AU)
2. Liège (BE)
3. Duisburg (DE)
4. Nijmegen (NL)
5. London (UK)
6. Cologne (DE)

Figure 1: Display of the simulation set-up domains: the port/cities of interest are displayed as dots on the map (orange for seaports and green for inland ports)



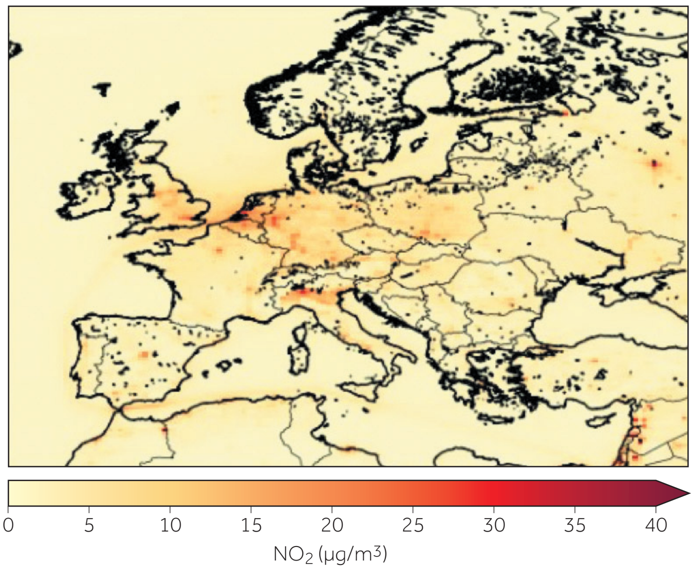
Results

Contribution of shipping emissions to air quality in Europe

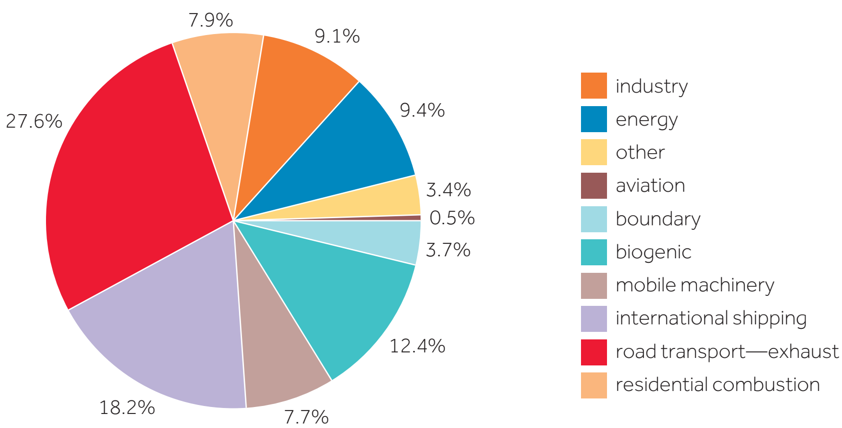
Figure 2 shows the predicted annual average surface concentration of NO_2 in 2018 for the European domain, together with the source apportionment results of the whole domain. High NO_2 concentration values are calculated in the central part of Europe (Benelux, Germany, UK) and in the Po Valley (north of Italy) with the biggest contributions, on average over Europe, being attributed to road transport (exhaust) and international shipping. The relative contribution of inland shipping is < 0.5% which is a small contribution in the European domain.

Figure 2: The annual average NO_2 surface concentration for 2018 in the simulation domain of the coarse (25 x 25 km) resolution LOTOS-EUROS simulation (a), and the relative contributions from the various labelled sectors to the surface concentration of NO_2 for the entire simulation domain (b)

a) Annual average NO_2 concentration in Europe

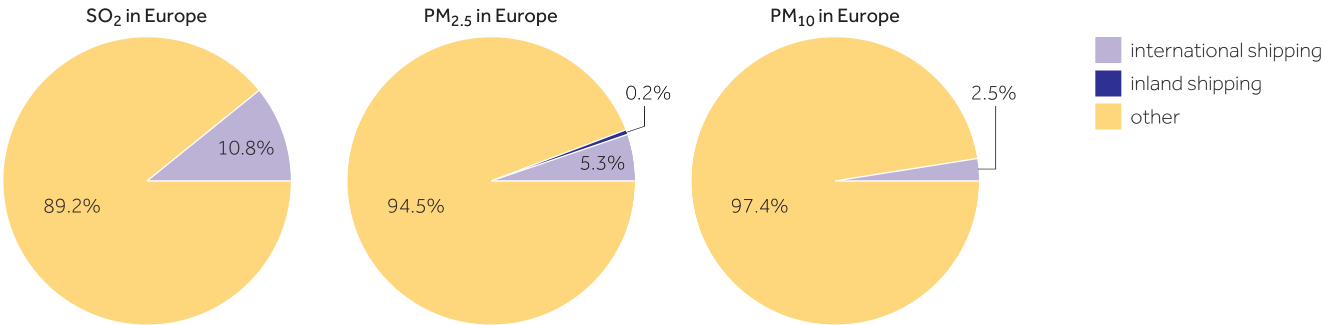


b) Contributions of the various labelled sectors to NO_2 in Europe



For the remaining pollutants examined in the study, the predicted contribution of shipping emissions compared to other sectors is shown in Figure 3. It is evident from the results that shipping has the largest relative contribution for NO_2 compared to other pollutants (Figures 2, 3). For SO_2 the contribution from international shipping is still significant (11%), while for PM the shipping contribution is somewhat smaller. In addition, inland shipping is predicted to be a negligible contributor to atmospheric pollutant concentrations on average in Europe.

Figure 3: The predicted relative contribution from international and inland shipping to SO_2, $PM_{2.5}$ and PM_{10} (annual average surface concentrations over Europe in 2018)



Contribution of shipping emissions for each port

Because the highest contribution of shipping is found for NO_2, the results of the calculated shipping contribution to NO_2 levels near the ports are presented and discussed here in more detail. Illustrative examples are used for three cases:

a) a port located near to the city centre (i.e. Piraeus);

b) a port located close to the city (i.e. Antwerp, ~10 km distance); and

c) a port located far from the city (i.e. Rotterdam, ~30 km distance).

Detailed results for all ports and pollutants can be found in the appendix of the full Concawe report of this study.[22] For the analyses, a representative central location for the port and the city centre was determined for the selected cities. The city centre locations are represented as blue dots and the port locations as green dots on Figures 4 to 6. For these locations of interest, the concentration fields were calculated as a weighted average of the four nearest grid points in the simulation domain (inversely with distance from the grid point to the coordinates of the location of interest).

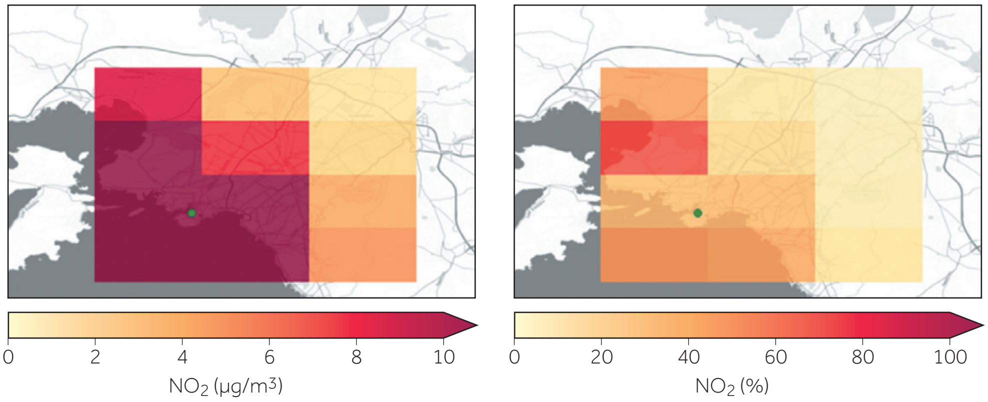
Piraeus

Figure 4 shows the absolute and relative contribution of international shipping to NO_2 annual average surface concentrations. The upper left portion of the figure shows that the absolute contribution of international shipping exceeds 5 μg/m^3 in most of the surrounding areas, and can reach up to 10 μg/m^3 or higher at the seaport. The port is located at the city centre, which causes the green and blue dots to coincide and the associated pie charts to be the same. International shipping is predicted to contribute 12 μg/m^3 (34%) to the annual average surface NO_2 concentration in Piraeus, and is the dominant source closely followed by exhaust emissions from the road transport sector.

Figure 4: The calculated absolute contributions (a) and relative contributions (b) of international shipping to the annual average surface NO_2 concentration in Piraeus in 2018. The pie charts show the relative contributions of various sectors to the NO_2 concentrations in the city centre (c) and the port (d) — these are the same because the port is essentially located at the city centre.*

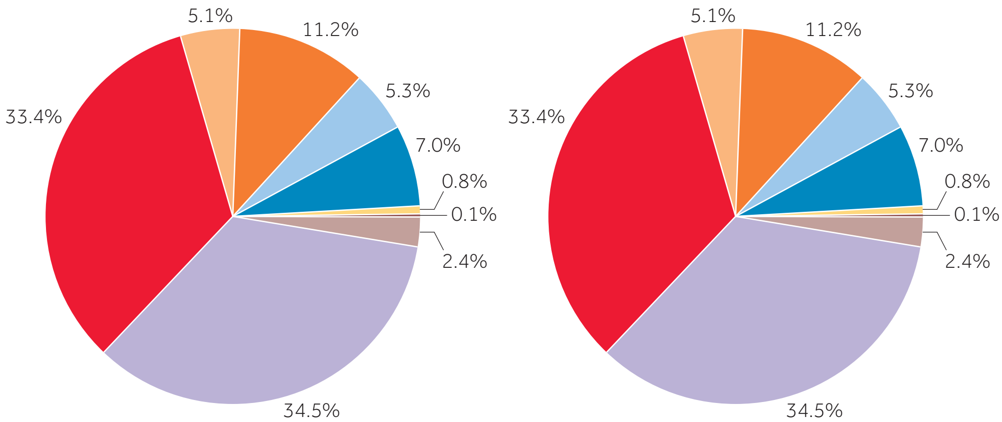
a) Absolute contributions of international shipping to NO_2 concentrations in Piraeus

b) Relative contributions of international shipping to NO_2 concentrations in Piraeus



c) Relative contributions of various sectors to NO_2 concentrations in the city centre*

d) Relative contributions of various sectors to NO_2 concentrations in the port*



Note: Inland shipping is not shown in the pie charts as there is no contribution from this sector around Piraeus.

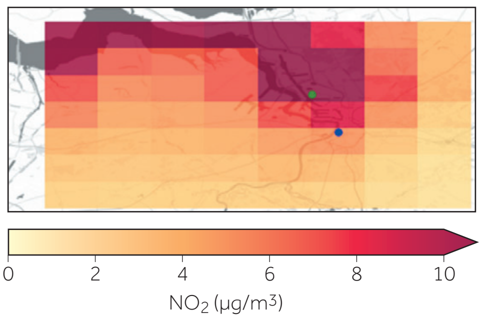
Antwerp

Antwerp is by far the biggest Belgian (sea) port, located at the river Scheldt which also features the port of Ghent closer to the sea. It plays an important role in the connection between the port of Hamburg and Le Havre in nearly all major traffic flows.

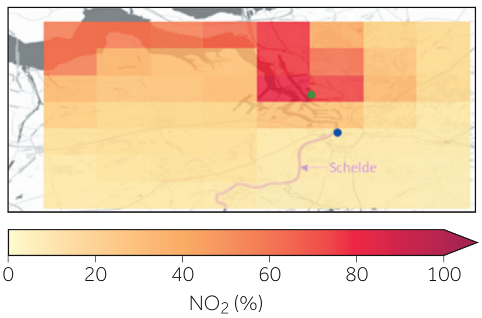
Figure 5 shows the contribution of international shipping to the NO_2 concentration in Antwerp. The absolute contribution of international shipping at the port of Antwerp (green dot) located at the delta of the Scheldt River is 16 µg/m^3 (47%). The relative contribution from international shipping to NO_2 concentration can go up to 70% following the Scheldt River further downstream. The NO_2 concentration in the ports assessed is predicted to also receive contributions from emissions from ships at berth in the ports. This also influences the air quality in the centre of Antwerp situated to the southeast of the port (blue dot). Here, international shipping contributes 5.8 µg/m^3 (24%) and inland shipping contributes 1.0 µg/m^3 (4.0%).

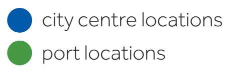
Figure 5: The calculated absolute contributions (a) and relative contributions (b) of international shipping to the annual average surface NO_2 concentration in Antwerp in 2018. The pie charts show the relative contributions of various sectors to the NO_2 concentrations in the city centre (c) and the port (d).

a) Absolute contributions of international shipping to NO_2 concentrations in Antwerp

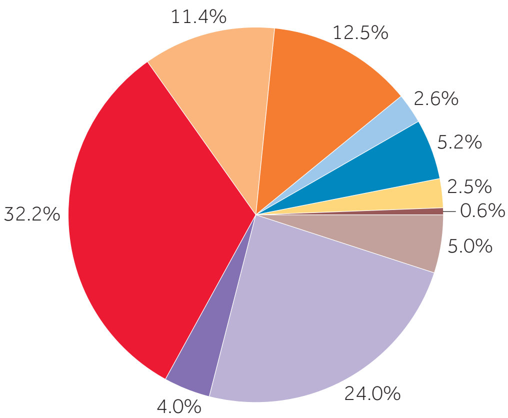


b) Relative contributions of international shipping to NO_2 concentrations in Antwerp

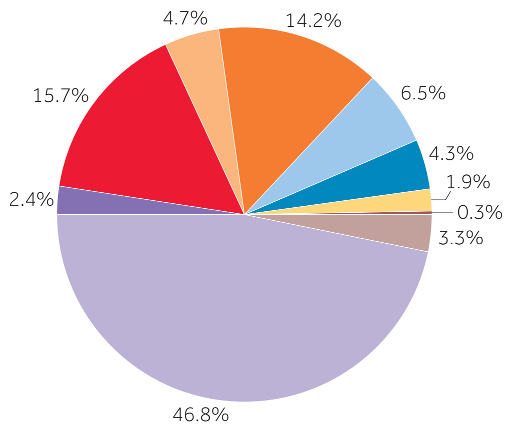




c) Relative contributions of various sectors to NO_2 concentrations in the city centre



d) Relative contributions of various sectors to NO_2 concentrations in the port

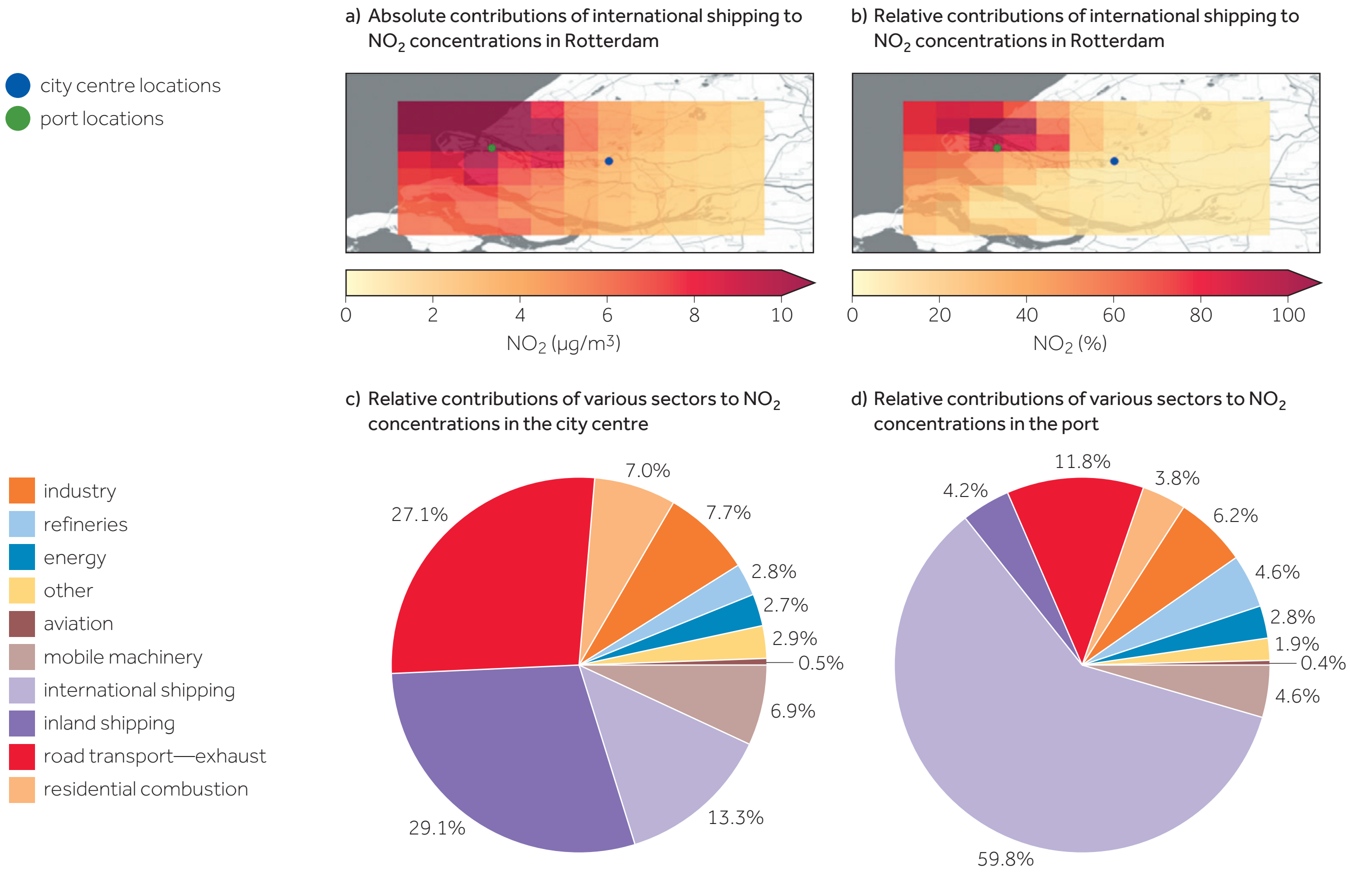


Rotterdam

The distribution of the contribution of international shipping to the annual average NO_2 surface concentration in the Rotterdam area for 2018 is shown in Figure 6. The pie charts show the relative contributions from all labelled source sectors at the main container terminal of the port (bottom right) and in the city centre of Rotterdam (bottom left).

The average annual absolute contribution of international shipping is 16 $\mu g/m^3$ (60%) at the port entrance at sea (green dot), while even at 35 km to the north, in the centre of Rotterdam (blue dot), contributions of 3.7 $\mu g/m^3$ (13%) are found. The absolute contribution, however, decreases between the port and the city centre due to dilution upon transport and the lifetime of NO_2 (the distance between the port and the city is approximately 30 km). In contrast, the absolute contribution from inland shipping is larger in the city centre than at the port location (respectively 8.1 (29%) vs 1.1 (4%) $\mu g/m^3$) and inland shipping is calculated to be the most dominant source in the city centre before exhaust emissions from the road transport sector.

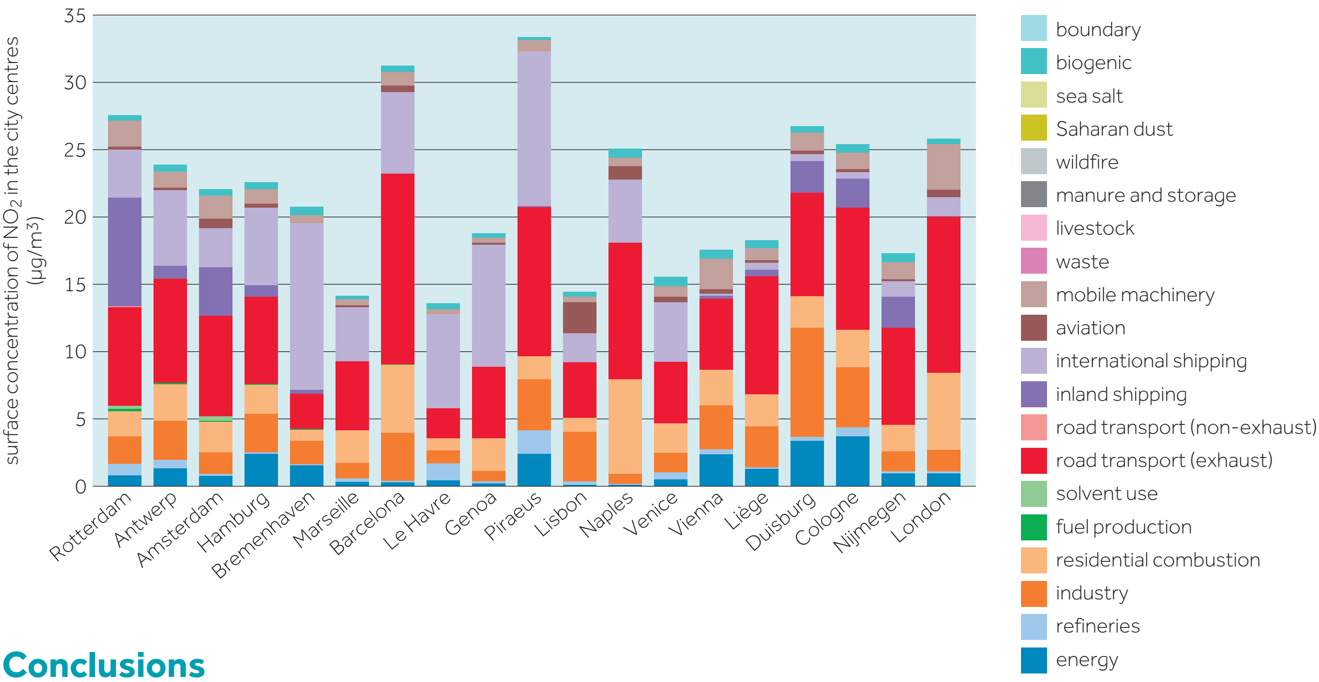
Figure 6: The calculated absolute contributions (a) and relative contributions (b) of international shipping to the annual average surface NO_2 concentration in Rotterdam in 2018. The pie charts show the relative contributions of various sectors to the NO_2 concentrations in the city centre (c) and the port (d).



The river Rhine (which is the major inland waterway linking the North Sea with industrial areas in Germany and its eastern neighbours via the Rhine-Main-Danube canal) ends in Rotterdam; this leads to the significant contribution from inland shipping to the air quality in Rotterdam city.

An overview of the absolute contributions from the labelled sectors to the centre locations of all cities examined in this study is given in Figure 7.

Figure 7: The predicted absolute contributions from the various labelled sectors to the annual average surface NO_2 concentration in 2018 for the city centres of the port cities of interest



Conclusions

The contribution of international and inland shipping to atmospheric pollutant concentrations in Europe were assessed using the chemical transport model LOTOS-EUROS and its source appointment feature that allows tracing of labelled emitted pollutants. The main findings from the study are summarised below:

- International shipping contributes significantly to NO_2 concentrations in Europe (18% on average).
- The contribution of shipping to other species is lower but still noteworthy (11% for SO_2, 2.5–5% for $PM_{2.5}/PM_{10}$).
- Locally in the seaports, the contribution of international shipping to NO_2 concentrations is higher and reaches up to 60% (Rotterdam).
- The contribution to NO_2 remains significant in all cities located near to the seaports, while in several cases, international shipping can be the dominant source (e.g. Piraeus, Hamburg).
- On average over the cities examined, the relative contribution from international shipping is 22%, whereas if only the seaport locations are considered, the contribution is higher with an average of 28%.
- Inland shipping generally has a low influence (0–4%) on NO_2, with only a few exceptions (Rotterdam, Amsterdam).

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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.

1 AOT40 (Accumulated Ozone exposure over a Threshold of 40 ppb, expressed in µg/m^3 per hour) is the sum of differences between hourly concentrations greater than 80 µg/m^3 (= 40 ppb) and 80 µg/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 µg/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

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)

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

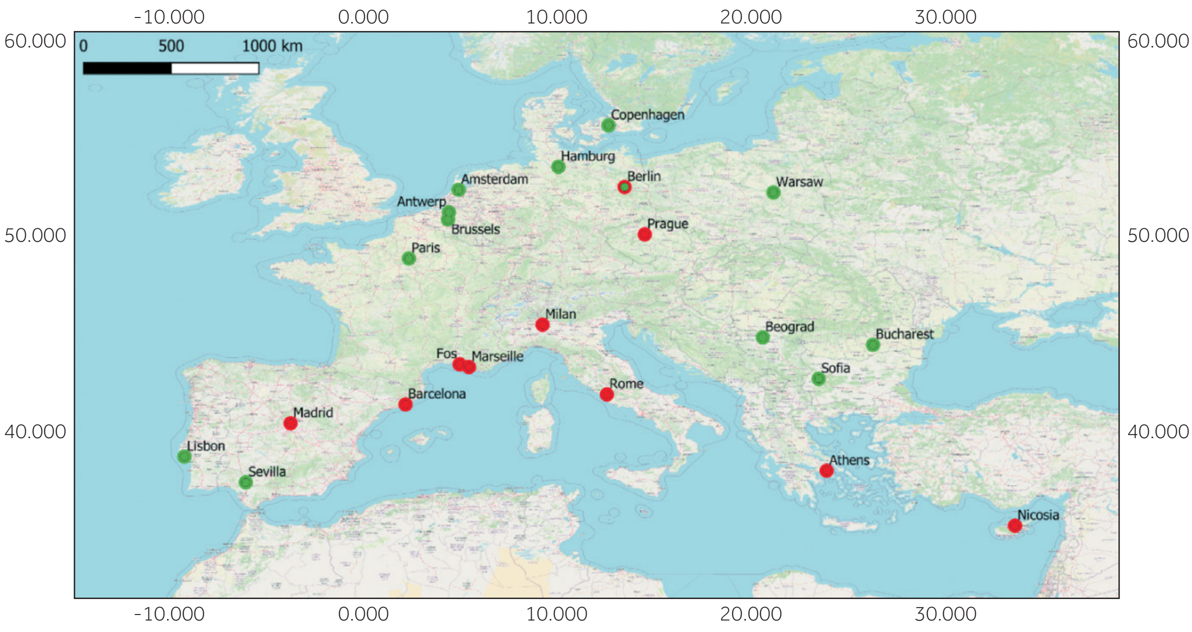
The surrogate ACT model trained on CHIMERE sensitivity simulations also allows exploring the chemical sensitivity (or regimes) within the parameter space of sectoral emission reductions. ACT is a quadrivariate second order polynomial with interactions using as predictors the four sectors considered. 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

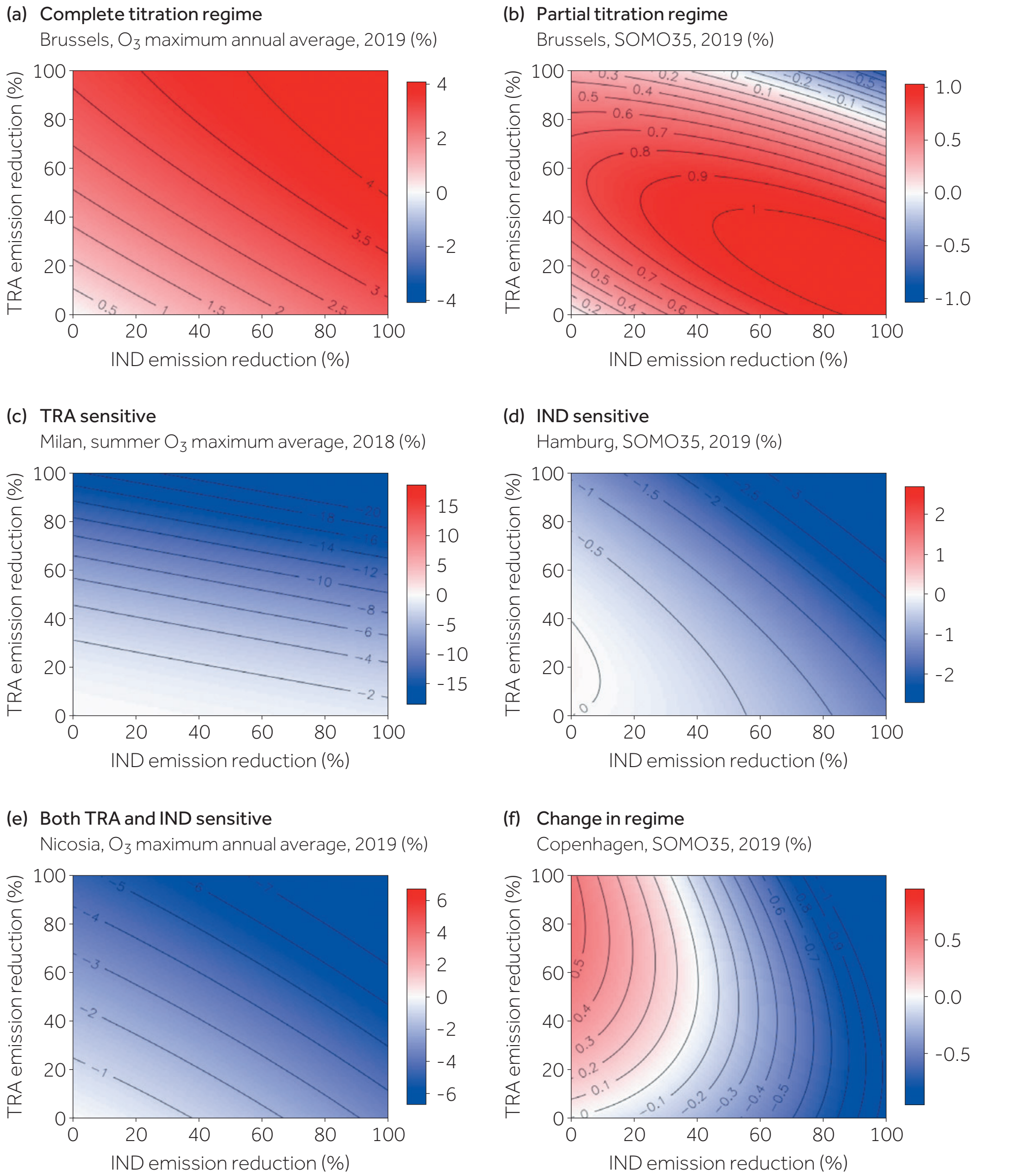
Examples of O_3 regimes and isopleths

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 µg/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 µg/m^3 threshold.

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



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.

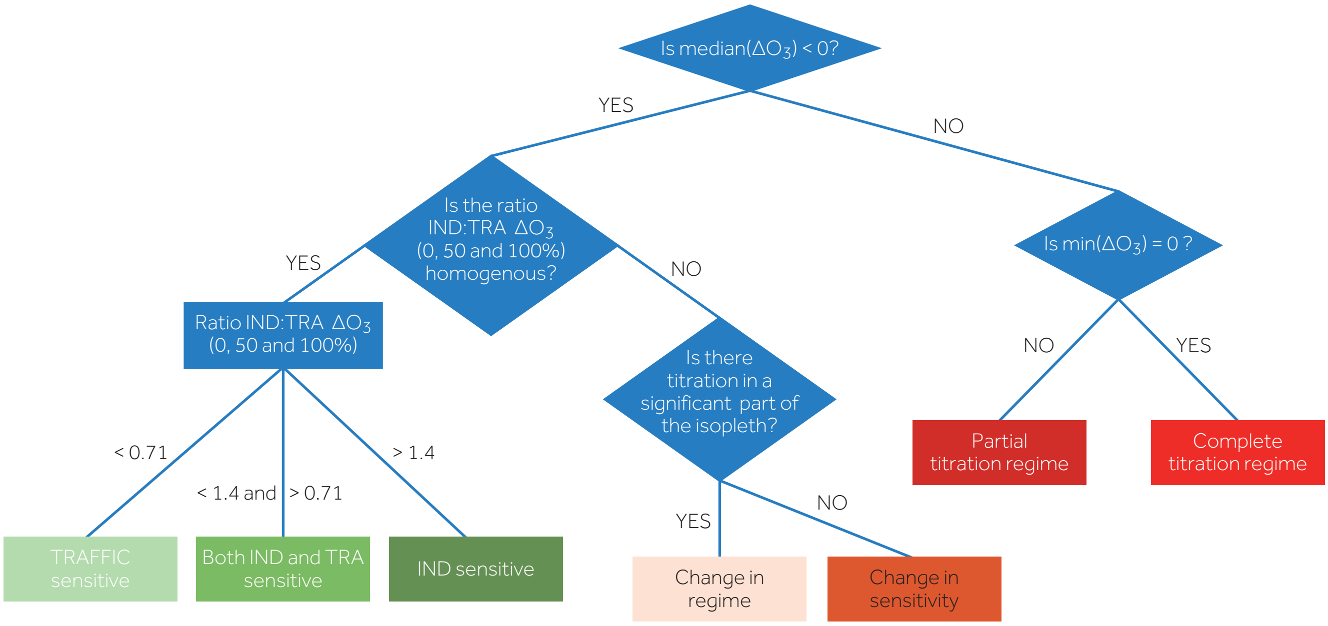
Ozone regimes

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.

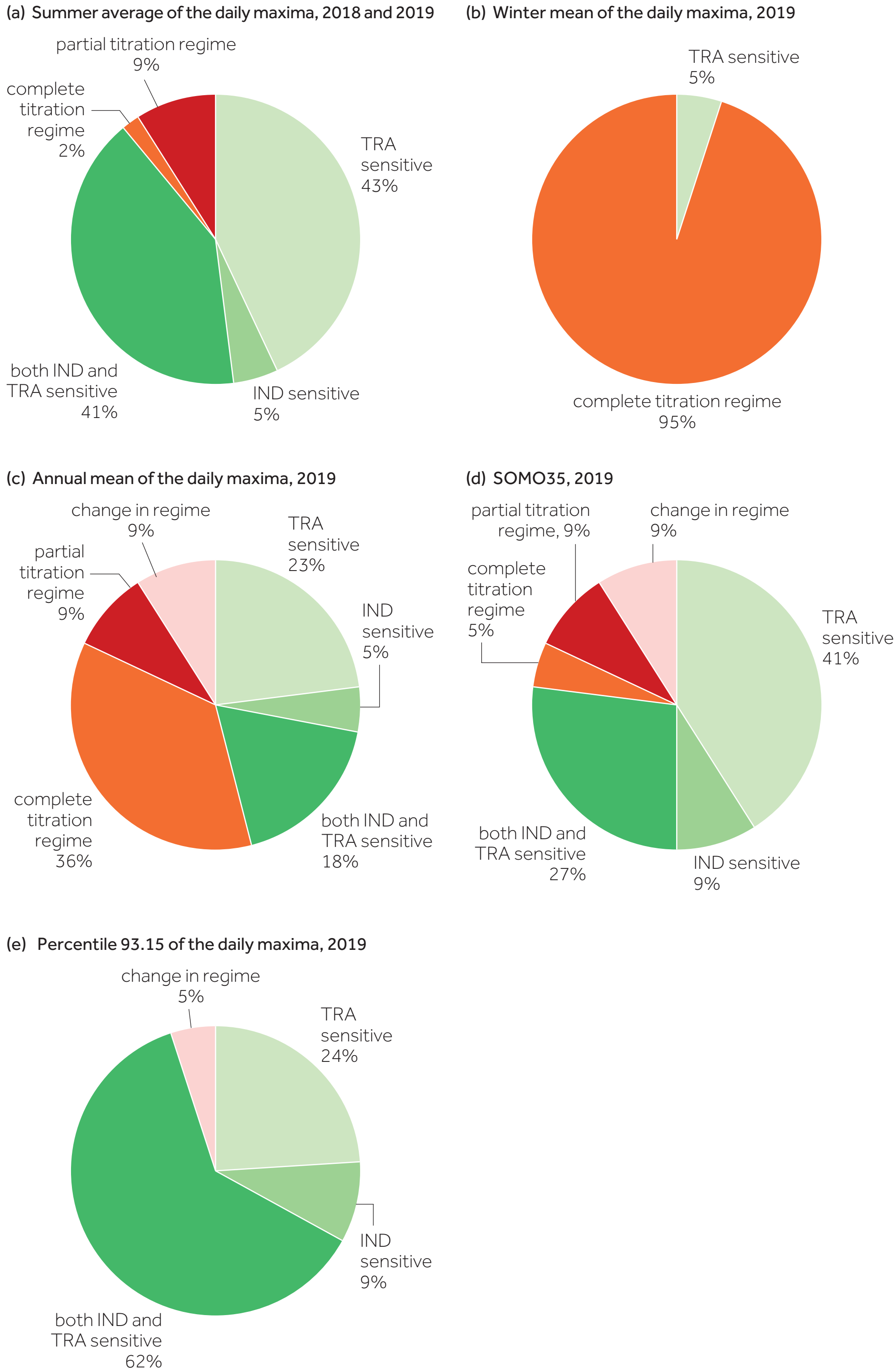
A value of the median $\Delta O_3 < 0$ indicates that reducing IND or TRA emissions yields some benefit in reducing ozone concentrations. The response can, however, be quite different depending on targeted cities, ozone metrics, or selected year/period. This response was therefore subsequently classified 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



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 µg/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.

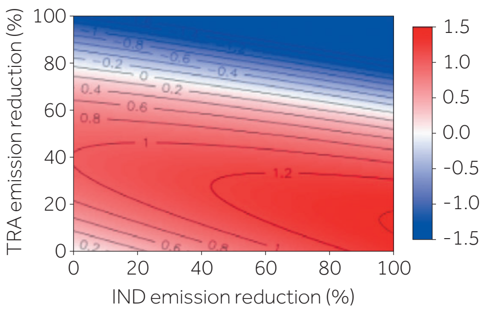
Factors influencing the differences in O_3 regimes between cities (e.g. meteorological factors, emissions speciation factors) are analysed in depth in the 'Atlas of ozone chemical regimes in Europe'.[4]

Examples from the *Supplementary Material*[5]

Milan

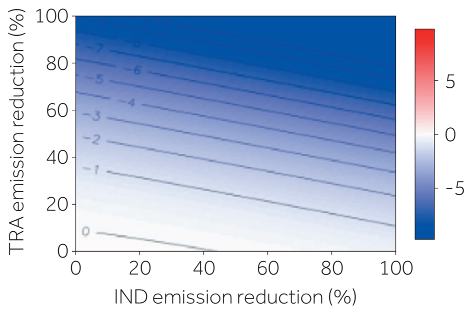
(a) **Annual maximum average, 2019**

Milan, O_3 maximum annual average, 2019 (%)



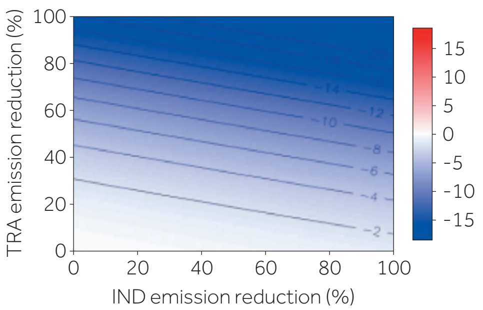
(b) **SOMO35, 2019**

Milan, SOMO35, 2019 (%)



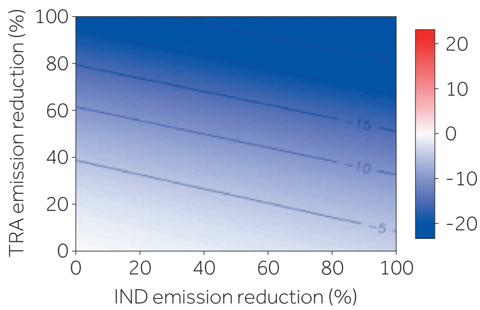
(c) **Summer maximum average, 2018**

Milan, summer O_3 maximum average, 2018 (%)



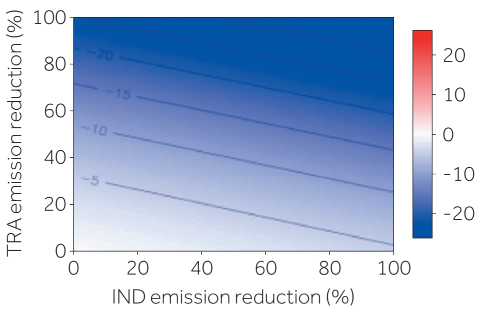
(d) **Summer maximum average, 2019**

Milan, summer O_3 maximum average, 2019 (%)



(e) **Percentile 93.15, 2019**

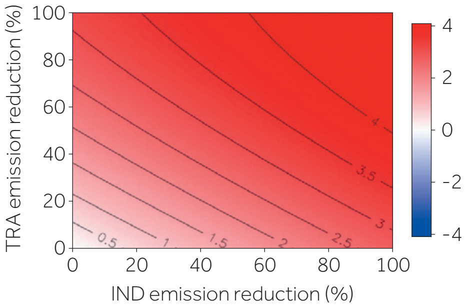
Milan, O_3 93.15 percentile, 2019 (%)



Brussels

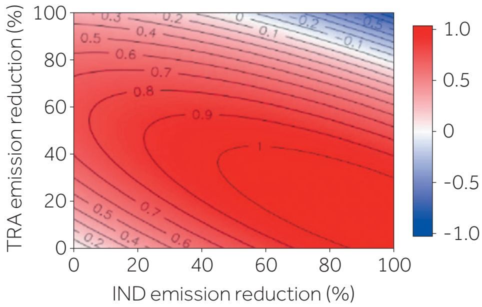
(a) Annual maximum average, 2019

Brussels, O_3 maximum annual average, 2019 (%)



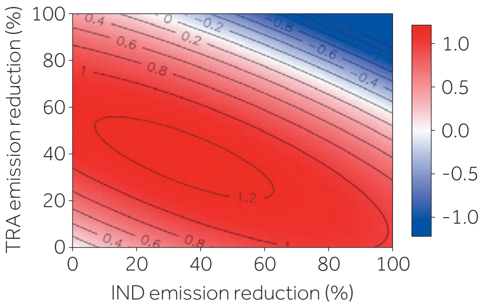
(b) SOMO35, 2019

Brussels, SOMO35, 2019 (%)



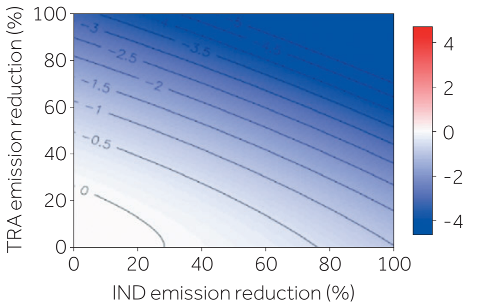
(c) Summer maximum average, 2018

Brussels, summer O_3 maximum average, 2018 (%)



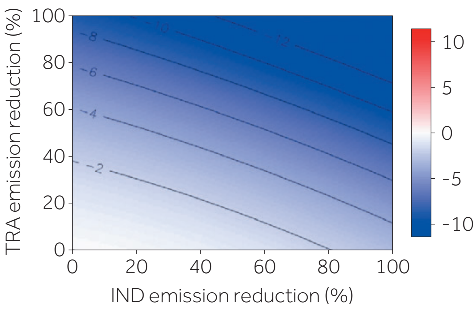
(d) Summer maximum average, 2019

Brussels, summer O_3 maximum average, 2019 (%)



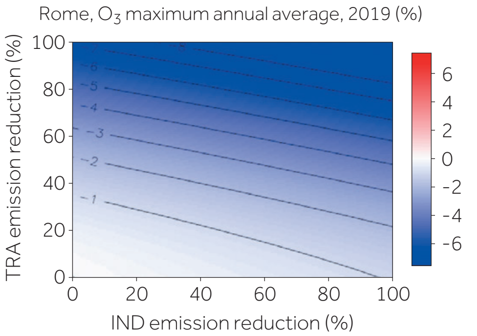
(e) Percentile 93.15, 2019

Brussels, O_3 93.15 percentile, 2019 (%)

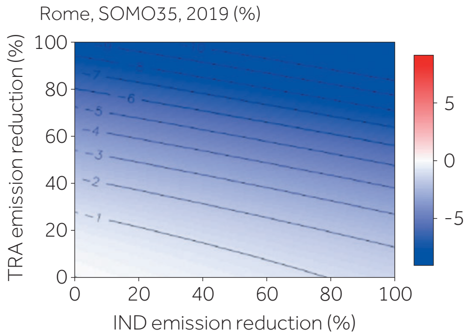


Rome

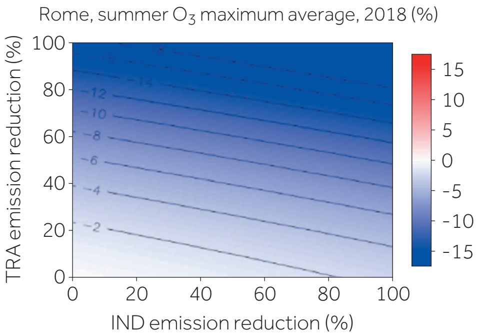
(a) **Annual maximum average, 2019**



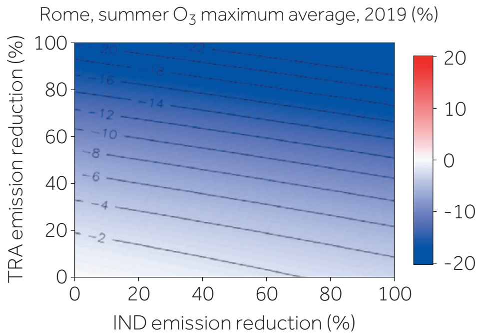
(b) **SOMO35, 2019**



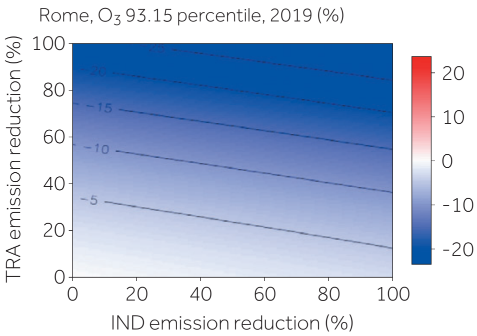
(c) **Summer maximum average, 2018**



(d) **Summer maximum average, 2019**



(e) **Percentile 93.15, 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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The impact of aviation emissions on urban air quality in Europe — an airport/city analysis

The study summarised in this article aims to enhance understanding of the influence of aviation emissions on air quality in cities with large airports nearby, compared with the influence of emissions from other sectors. This article describes the methodology, the types of data and the simulation set-up used, and presents a summary of the results for three of the six European cities studied. The complete analysis can be found in the full Concawe report on this study.[1]

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Introduction

It is well known that fuels burnt by aircraft engines result in emissions of several pollutants such as NO_x, SO_2, soot and particulate matter (PM), etc. that have a negative impact on air quality and are thus harmful to human health and ecosystems.

Despite that, on average in Europe, the aviation sector is not considered a large contributor to the emissions of air pollutants of concern, and has seen an increase in emissions compared to most other sectors. For example, between 2000 and 2018, aviation NO_x emissions showed an increase from 61 kt to 86 kt (41%), while for most of the other sectors emissions generally decreased.[2] That said, the relative contribution of the aviation sector to total emissions remains low for the whole of Europe (< 1.5% for NO_x in 2018). However, locally, in cities with major airports nearby, the effect of emissions from the aviation sector on ambient air quality is expected to be higher due to the proximity of the cities to the airports.

This work aims to provide insights and enhance understanding of the influence of aircraft emissions on ambient air quality in cities with, or near to, a major airport, by addressing the following research question: 'How is the air quality influenced by aviation emissions over Europe and specifically in cities with large airports in comparison to other sectors?'

In this context, the chemical transport model (CTM) LOTOS-EUROS and its source apportionment capabilities were used to assess the contribution of aviation emissions to atmospheric air pollutant concentrations for six cities in Europe (London, Paris, Amsterdam, Frankfurt am Main, Munich and Brussels) with large airports (Heathrow, Charles de Gaulle, Schiphol, Frankfurt am Main, Munich and Zaventem).

The methodology used in the study is described in the following section, which provides details on the model that is used and the data that are utilised as input to the model to perform the simulations of the atmospheric concentrations. The results of the study are presented in the third section of this article on pages 37–43. The CTM provides labelled atmospheric concentrations over the simulation domain. Using the simulation results, the contributions of various sectors to air emissions in airport-cities of interest are computed. The main findings are presented in the *Conclusions* on pages 43–44.

Methods

Model description

LOTOS-EUROS is a 3-D chemical transport model developed by TNO. The offline Eulerian grid model simulates air pollution concentrations in the lower troposphere, solving the advection-diffusion equation on a regular latitude-longitude grid with a variable resolution over Europe.[3]

The vertical transport and diffusion scheme accounts for atmospheric density variations in space and time and for all vertical flux components. The vertical grid is based on terrain following vertical coordinates and, excluding the stacked boundary layers on top, extends to 5 km above sea level. The height of the layers on top of the 25-metre surface layer is determined by heights in the meteorological input data.

Gas-phase chemistry is simulated using the TNO CBM-IV scheme, which is a condensed version of the original scheme.[4] The LOTOS-EUROS model explicitly accounts for cloud chemistry, computing sulphate formation as a function of cloud liquid water content and cloud droplet pH as described in Banzhaf *et al.* (2012).[5] For aerosol chemistry the thermodynamic equilibrium module ISORROPIA II is used.[6] Dry Deposition fluxes are calculated using the resistance approach as implemented in the DEPAC (DEPosition of Acidifying Compounds) module.[7] Furthermore, a compensation point approach for ammonia is included in the dry deposition module.[8] The wet deposition module accounts for droplet saturation following.[9]

The model is part of the Copernicus Atmospheric Monitoring Service (CAMS) regional ensemble providing operational forecasts and analyses over Europe. In this context the model is regularly updated and validated using observations from ground and satellite observations. The model performance is also subject to numerous peer-reviewed publications.[10,11,12]

Source apportionment

TNO has also developed a system to track the impact of emission categories within a LOTOS-EUROS simulation based on a labelling technique.[13] This technique provides more accurate information about the source contributions than using a brute force approach with scenario runs as the chemical regime remains unchanged. Another important advantage is the reduction of computational costs with respect to the brute force approach. The source apportionment module for LOTOS-EUROS provides a source attribution that is valid for current atmospheric conditions as all chemical conversions occur under the same oxidant levels. For details and validation of this source apportionment module, the reader is referred to Kranenburg *et al.*[13] The source apportionment technique has been used extensively in previous studies.[12,14,15]

As well as calculating the total concentrations of each pollutant, the contributions of selected sources to these concentrations are calculated. The labelling routine is implemented for primary, inert aerosol tracers as well as for chemically active tracers containing a C, N (reduced and oxidised) or S atom, as these are conserved and traceable.

Meteorology

The LOTOS-EUROS model is run with ECMWF ERA5 reanalysis meteorological data (2018). ERA5 provides hourly estimates of a large number of atmospheric, land and oceanic climate variables, that are necessary inputs for calculations of atmospheric concentrations. Examples of typical inputs required by LOTOS-EUROS are surface and air temperature, cloud cover, wind speed and direction, precipitation and relative humidity.

Quality-assured monthly updates of ERA5 (1959 to present) are published within three months of real time and are available through the Climate Data Store.[1] Preliminary daily updates of the dataset are available to users within five days of real time.

[1] https://cds.climate.copernicus.eu/#!/home

Emissions

The CAMS-REG inventory emission data for the year 2018 version 5.1 REF2[16] is used in this study for anthropogenic trace gas emissions. At the time of performing this study, this was the latest available dataset (an update with more recent data was published in December 2023). The inventory uses the official emissions data reported by European countries. The aviation emissions in the national inventories only include the total national emissions during the landing and take-off cycle (LTO). The LTO covers four modes of engine operation, namely idle, approach, climb-out and take-off, each of which is associated with a specific engine thrust setting and a time in mode. The emissions during cruise flights (above 3,000 feet) are not reported in the national inventories. The national aviation emissions (LTOs) in a country are distributed over the contributing airports in that country based on flight statistics per airport on an annual basis. All emissions from an airport are represented as a point source at the location of the airport in the CAMS-REG inventory.

Since this study aims to better assess the local effects and spatial extent of aviation emissions at an airport and the impact they have on air quality in its vicinity, modelling simulations are performed at a 1 x 1 km resolution. Because the CAMS-REG inventory has a 6 x 6 km resolution, it is not suitable for use in the 1 x 1 km resolution simulations, so for this purpose a 1 x 1 km regridded dataset is used that contains CAMS-REG emissions for NO_x at a 1 x 1 km resolution. The regridding is done based on high-resolution proxy data, such as road and rail networks and land use maps, while keeping the annual total emissions per sector unchanged. For SO_2 and PM, this level of detail is not available from the CAMS-REG inventory. However, for the Netherlands and Germany, emissions for all pollutants of interest are available at this resolution from other national datasets, namely the Emission Register (ER) in the Netherlands and GrETA (Gridding Emission Tool for ArcGIS) in Germany,[17] which provide a more detailed representation of airports with runways rather than point sources.

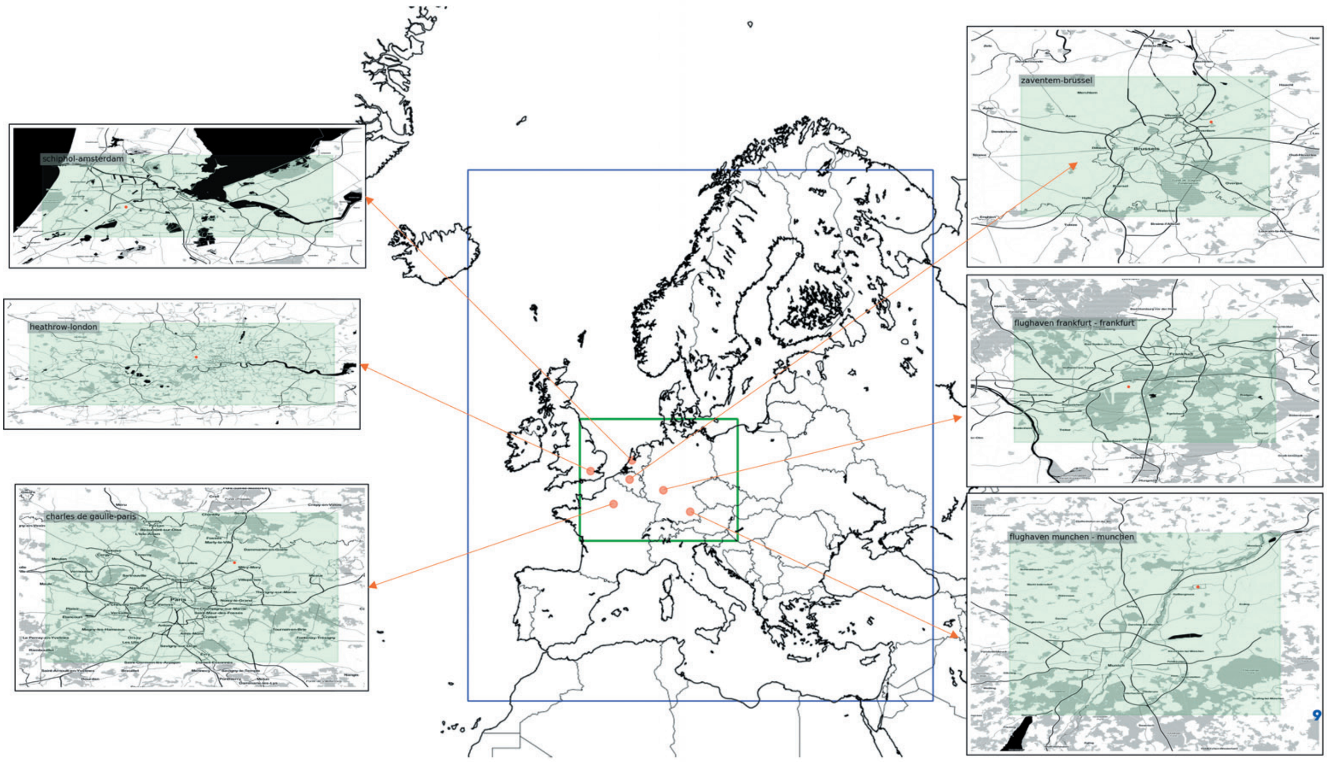
As multiple emissions datasets are available, an exploratory study was undertaken to find the most appropriate option to use in the simulations. For this purpose, the city of Amsterdam was chosen as a test case since all three emissions datasets (CAMS-REG version 5.1 6 x 6, CAMS-REG version 5.1 1 x 1 and ER 1 x 1) are available, and were used as input for the LOTOS-EUROS model for a simulation of the pollutant concentrations in January and July 2018. Details on the analyses can be found in the respective Concawe report.[1] Based on the results, the decision was made to use emissions at the highest available resolution for as many of the relevant pollutants as possible to avoid unrealistic patches in the simulated concentrations. Hence, for the Netherlands and Germany, ER and GrETA emissions data are used because high-resolution data are available for all pollutants of interest. Unfortunately, this dataset does not cover Paris, London or Brussels. For these domains, the CAMS-REG v5.1 1 x 1 dataset was used.

Model set-up

Figure 1 shows the different domains which are part of the LOTOS-EUROS simulations. In the middle of the figure, a coarse resolution (circa 25 x 25 km) simulation performed over Europe (domain edge in purple) is shown. Results from this simulation are used as a boundary condition for the nested simulation over north-western Europe (domain in green) at a higher resolution (circa 6 x 6 km). As a next step, simulations are performed for the cities that are part of this study at higher resolution (circa 1 x 1 km). The chosen cities and domains are shown with orange dots in the centre figure. On the outsides of the figure, the domains of the 1 x 1 km high-resolution zoom runs are shown in more detail. The following major cities are studied in the high-resolution zoom runs:

1. London (UK)
2. Paris (FR)
3. Amsterdam (NL)
4. Frankfurt am Main (DE)
5. Munich (DE)
6. Brussels (BE)

Figure 1: Display of the simulation set-up domains
(the chosen cities and domains are shown by orange dots on the map at the centre)



In order to distinguish emissions sources from different sectors, a set of labels was applied during the source apportionment simulations. The complete set of labels is as follows:

1. Aviation
2. International shipping (all sea-going shipping)[2]
3. Inland shipping (all river-going shipping)
4. Public power/Energy
5. Residential combustion
6. Industry
   a. Solvent use[a]
   b. Fuel production
   c. Refineries[b]
   d. Other industry
7. Mobile machinery[c]
8. Road transport—exhaust
9. Road transport—non-exhaust (only contributes to PM)
10. Waste management
11. Agriculture
    a. Livestock
    b. Manure management
12. Biogenic[3]
13. Wildfires (GFAS,[4] daily[18])
14. Sea salt (only contributes to PM)
15. Saharan dust (only contributes to PM)
16. Boundary[5]

[a] Even though 'Solvent use' is considered a subcategory of industry, domestic solvent use is included here. This is, however, a relatively small contribution.
[b] Oil, gas and petroleum refining is incorporated in this label. The label 'Fuel production' contains emissions that occur during production, distribution, exploration, gas flaring and venting, and oil and coal handling.
[c] 'Mobile machinery' contains emissions from railways, small agricultural, forestry and fishing equipment, compressors, gardening, off-road vehicle usage, etc.

There is a strong variation in the influence of these emission sources on surface concentrations of $PM_{2.5}$, PM_{10}, NO_2 and SO_2. In the analysis of the results, only those sectors that contribute significantly (> 2%) are presented graphically, with the exception of sectors of special interest (aviation) that are always reported if they contribute. The less-contributing sectors are aggregated and labelled as 'Other'.

The results of the study are presented in the following section of this article.

[2] A detailed analysis of the contribution from shipping can be found in Concawe Report no. 2/23, *The impact of shipping emissions to urban air quality in Europe – Detailed port-city analysis.*

[3] Biogenic emissions include isoprene and monoterpene from vegetation and soil NO_x emissions.

[4] Global Fire Assimilation Service, https://atmosphere.copernicus.eu/global-fire-emissions

[5] The label 'Boundary' is used to describe contributions from the CAMS global simulation results that are used as a boundary condition to the simulation over the European domain.

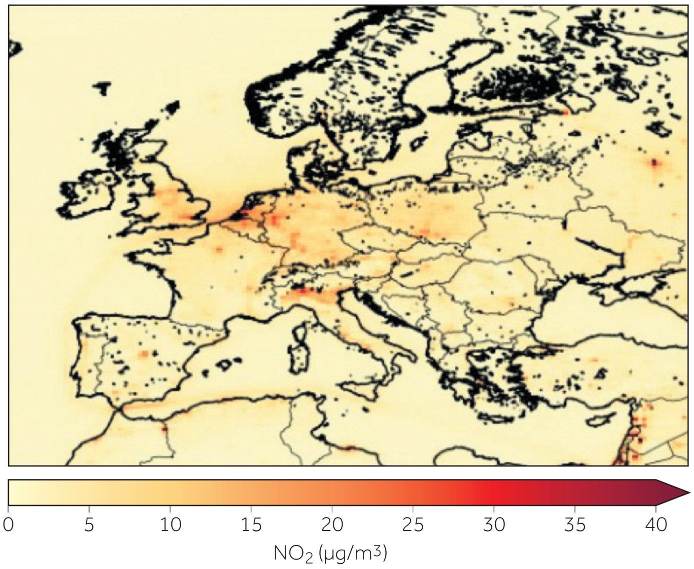
Results

Contribution of aviation emissions to air quality in Europe

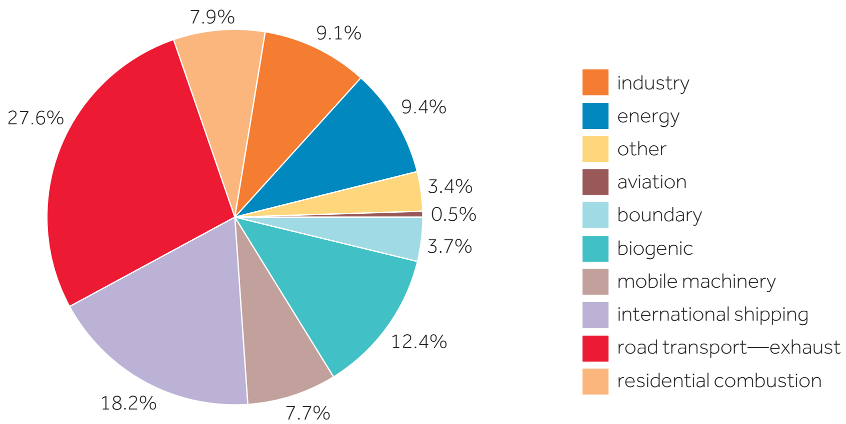
Figure 2 shows the predicted annual average surface concentration of NO_2 in 2018 for the European domain, together with the source apportionment results of the whole domain. High NO_2 concentrations are mainly predicted in the central part of Europe (Benelux, Germany, UK) and in the Po Valley (north of Italy) with 'Road transport—exhaust' and 'International shipping' being the two largest contributors.

Figure 2: The annual average NO_2 surface concentration for 2018 in the simulation domain of the coarse (25 x 25 km) resolution LOTOS-EUROS simulation (a), and the relative contributions from the various labelled sectors to the surface concentration of NO_2 for the entire simulation domain (b)

a) Annual average NO_2 concentration in Europe



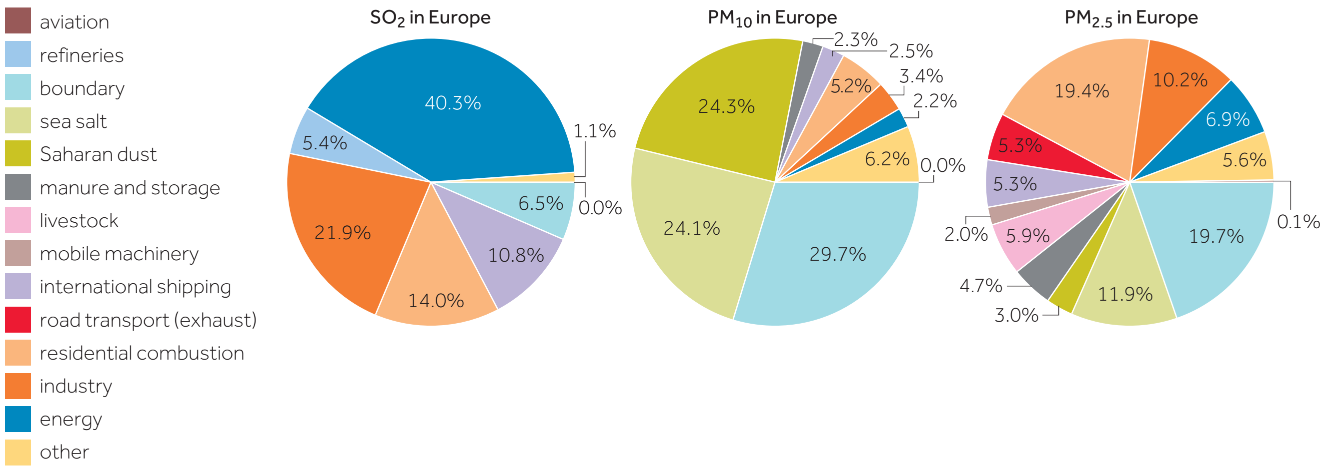
b) Contributions of the various labelled sectors to NO_2 in Europe



In contrast, the aviation sector contribution, averaged over the whole European domain, is relatively small (0.5%), which is to be expected due to its local nature and the short lifetime of NO_2 (this is discussed in more detail in the city/airport analyses results).

Aviation is also predicted to be a negligible contributor to the average surface concentrations of the remaining pollutants examined in the study, as shown in Figure 3. On average over the European domain, the results show that for $PM_{2.5}$, PM_{10} and SO_2 the contributions are 0.14% (a 1.7 ng/m^3 contribution to a domain average of 1.2 µg/m^3), 0.04% (a 2.2 ng/m^3 contribution to a domain average of 5.5 µg/m^3) and 0.03% (a 6.6 ng/m^3 contribution to a domain average of 21 µg/m^3), respectively.

Figure 3: The predicted relative contributions from the various labelled sectors to SO_2, PM_{10} and $PM_{2.5}$ (annual average surface concentrations over Europe in 2018)



Contribution of aviation emissions in cities/airports

Looking at the surface contributions, averaged over the European domain, it would appear that aviation seems to be a sector of limited significance. Aviation activities could nevertheless be relevant, since these are commonly concentrated in densely populated areas. It is therefore worthwhile to take a closer look at the fractional contributions of the various source sectors in the vicinity of large airports.

Because the highest contribution of aviation is found for NO_2, the results of the calculated aviation contribution to NO_2 levels near the cities where airports are located are presented and discussed here in more detail. The cities of London, Paris and Amsterdam are used as illustrative examples while detailed analyses for all cities and pollutants can be found in the full Concawe report on this study.[1] For the analyses, a representative central location for the airport and the city centre was determined for the selected cities. The city centre locations are represented as blue dots and the airport locations as turquoise dots on Figures 4 to 6. For these locations of interest, the concentration fields were calculated as a weighted average of the four nearest grid points in the 1 x 1 km simulation domain (inversely with distance from the grid point to the coordinates of the location of interest).

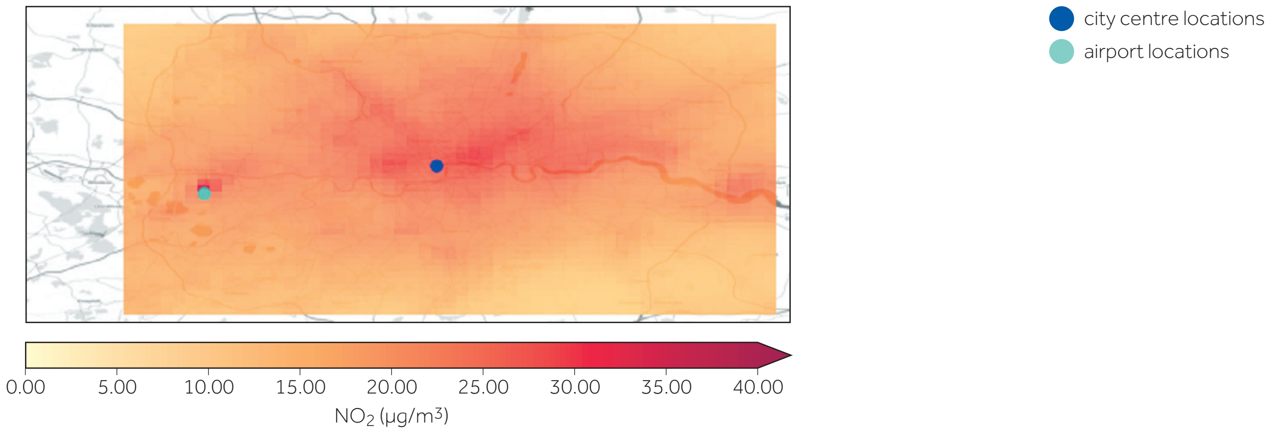
London

London city centre is located 25 km east of Heathrow airport (IATA[6] code: LHR) the busiest airport in Europe with respect to passenger throughput (i.e. more than 80 million passengers passed through Heathrow in 2018). At Heathrow, the predicted elevated NO_2 concentrations can be largely attributed to aviation activities, with a contribution of 54.9% (17.2 μg/m^3) (Figure 4).

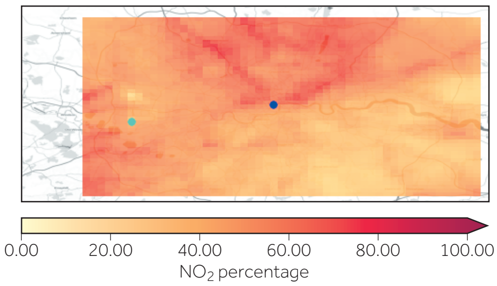
[6] International Air Transport Association

Figure 4: Predicted annual average NO_2 concentration in and around London (a), and the relative contributions of road transport (the largest contributor in the region) (b) and aviation (c) to this concentration. The pie charts show the contributions from various sectors to NO_2 concentrations in the city centre and near Heathrow airport.

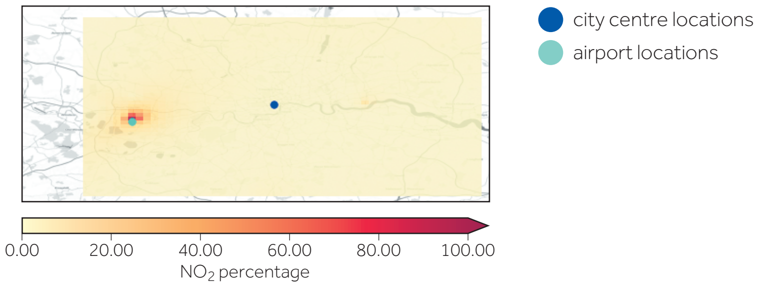
a) Predicted annual average NO_2 concentration in London



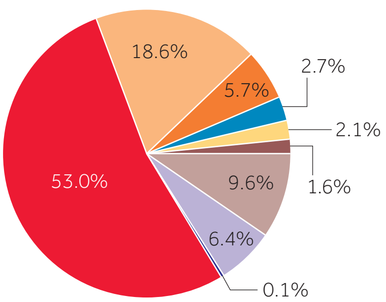
b) Relative contributions of road transport to NO_2 concentrations in the vicinity of London



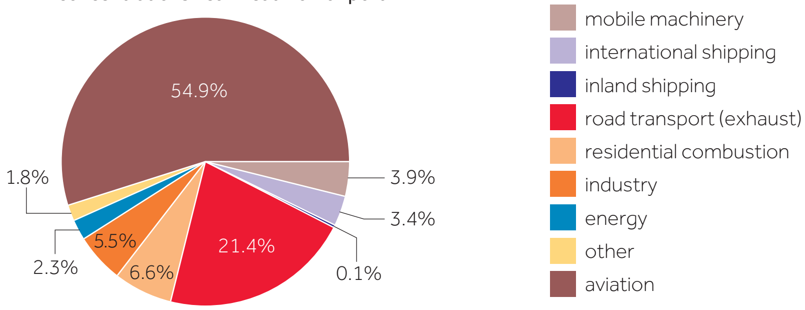
c) Relative contributions of aviation to NO_2 concentrations in the vicinity of London



d) Relative contributions of various sectors to NO_2 concentrations in London city centre



e) Relative contributions of various sectors to NO_2 concentrations near Heathrow airport



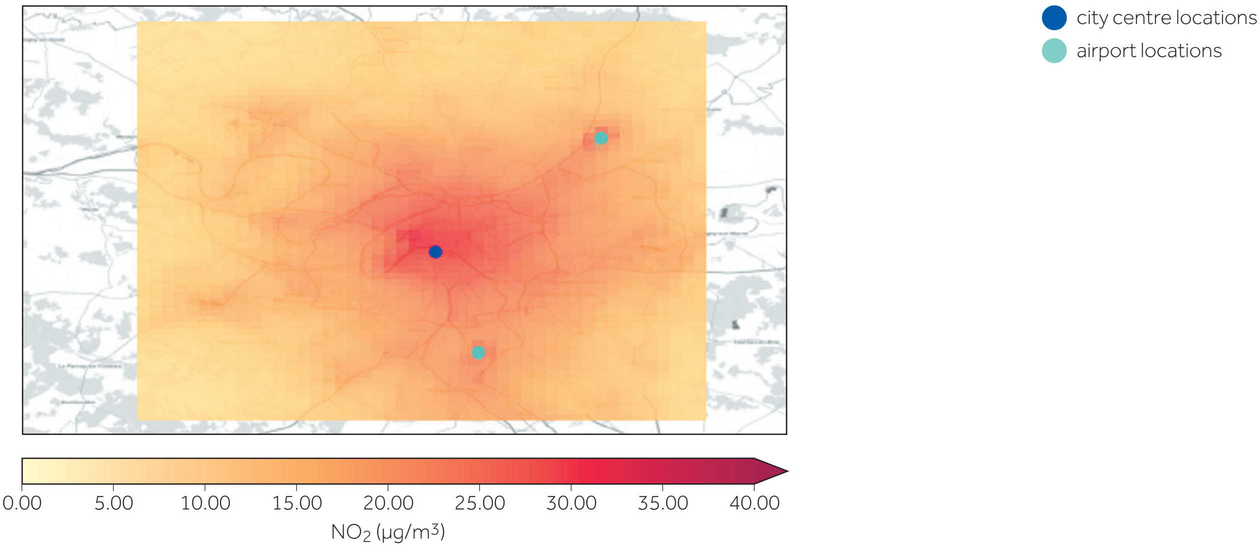
In the city centre, the contribution from aviation diminishes to 1.6% (0.44 μg/m^3) due to dilution upon transport and the lifetime of NO_2 in the atmosphere. If an exponent is fitted to the declining contribution of aviation as a function of distance from the airport, a 63% reduction in the relative contribution can be seen for every 2.8 km separation from the airport toward the city centre. This means that, at 2.8 km from Heathrow airport, aviation contributes 20% of the NO_2 concentration present. More details on this analysis can be found in the Appendix of the full Concawe report on this study.[1] The declining trend as a function of distance is also a result of a larger absolute contribution from other sources in the city of London (e.g. road transport and residential combustion), hence the relative contribution from aviation is reduced.

Paris

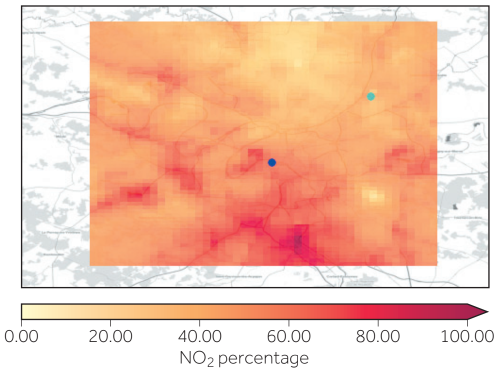
Paris has two airports in relatively close proximity to the city centre. Charles de Gaulle airport (IATA code: CDG) is located about 30 km north-east of the city centre and is Europe's second largest airport with a throughput of 72 million passengers, while to the south of Paris is Orly (IATA code: ORY), the second largest French airport with a throughput of 33 million passengers. At Charles de Gaulle airport, aviation is predicted to contribute around 58% (15.5 μg/m^3) of the NO_2 surface concentration. In the city centre, aviation activities at both Orly and Charles de Gaulle airports contribute 2.3% (0.68 μg/m^3) of the NO_2 surface concentration; the dominant sectors contributing to NO_2 emissions in the city centre are road transport and residential combustion activities, which together account for ~80% of the total NO_2 concentration. At 4.3 km from CDG airport, the relative contribution from aviation to the NO_2 concentration is predicted to reduce by 63% with respect to the relevant contribution at the airport (i.e. to 21%). This drop-off is less steep than the one found for London, and can be due to the presence of contributions from the Orly airport, or a smaller relative contribution from other sources. See Figure 5 on page 41.

Figure 5: Predicted annual average NO_2 concentration in and around Paris (a), and the relative contributions of road transport (the largest contributor in the region) (b) and aviation (c) to this concentration. The pie charts show the contributions from various sectors to the NO_2 concentration in the city centre and near CDG airport.

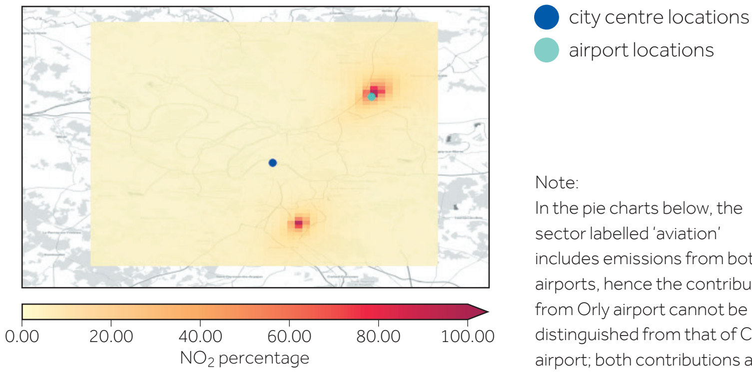
a) Predicted annual average NO_2 concentration in Paris



b) Relative contributions of road transport to NO_2 concentrations in the vicinity of Paris

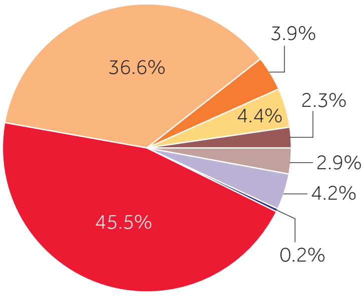


c) Relative contributions of aviation to NO_2 concentrations in the vicinity of Paris

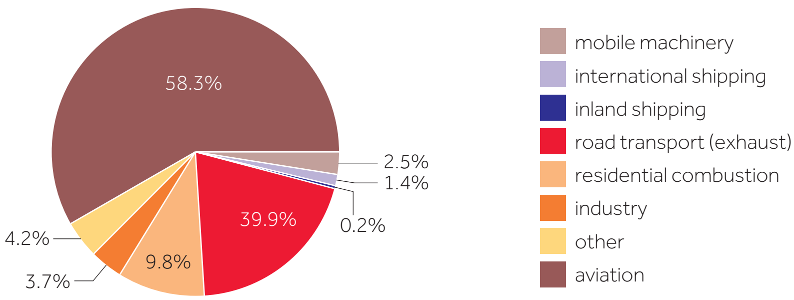


Note:
In the pie charts below, the sector labelled 'aviation' includes emissions from both airports, hence the contribution from Orly airport cannot be distinguished from that of CDG airport; both contributions are incorporated together in the pie charts and represented by the brown slice.

d) Relative contributions of various sectors to NO_2 concentrations in Paris city centre



e) Relative contributions of various sectors to NO_2 concentrations near CDG airport



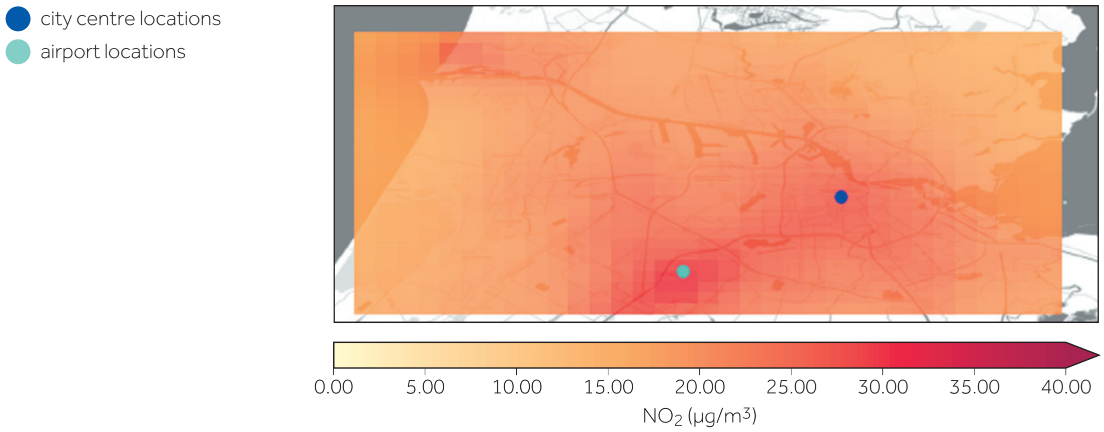
The impact of aviation emissions on urban air quality in Europe — an airport/city analysis

Amsterdam

Schiphol airport (IATA code: AMS) lies approximately 15 km south-west of Amsterdam city centre. It is the largest airport in the Netherlands and the third largest airport in Europe (after Heathrow and Charles de Gaulle) with a throughput of 71 million passengers. The predicted NO_2 concentrations in and around Amsterdam, and the relative contributions of the various sectors are shown in Figure 6.

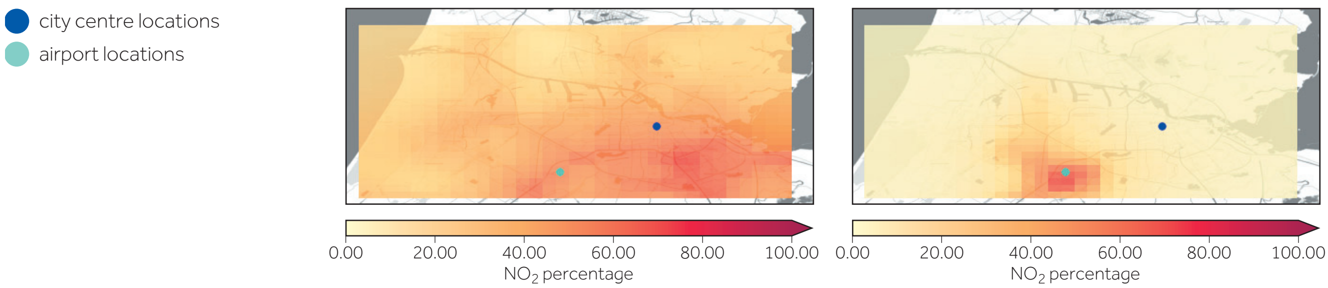
Figure 6: Predicted annual average NO_2 concentration in and around Amsterdam (a), and the relative contributions of road transport (the largest contributor in the region) (b) and aviation (c) to this concentration. The pie charts show the contributions from various sectors to the NO_2 concentration in the city centre and near Schiphol airport.

a) Predicted annual average NO_2 concentration in Amsterdam



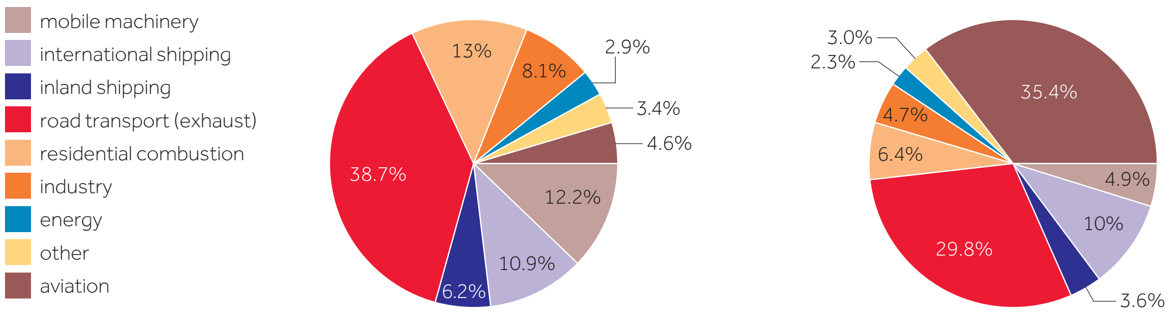
b) Relative contributions of road transport to NO_2 concentrations in the vicinity of Amsterdam

c) Relative contributions of aviation to NO_2 concentrations in the vicinity of Amsterdam



d) Relative contributions of various sectors to NO_2 concentrations in Amsterdam city centre

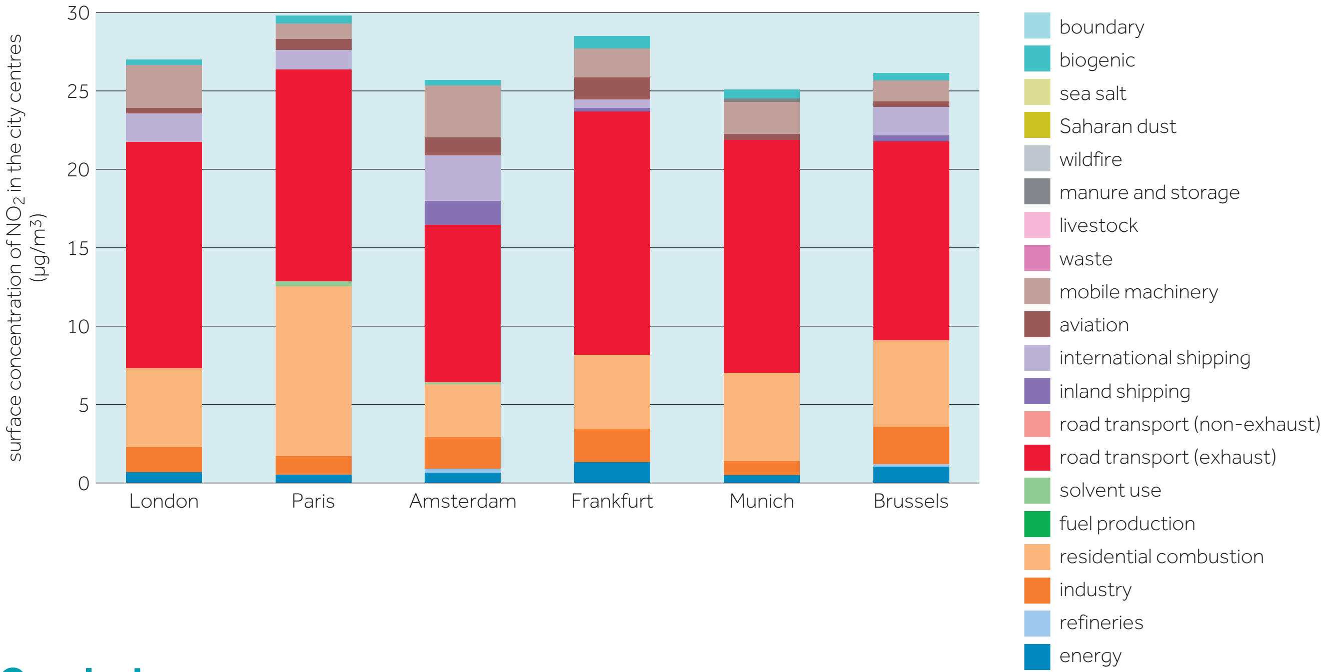
e) Relative contributions of various sectors to NO_2 concentrations near Schiphol airport



The modelling simulations predict that aviation activity at Schiphol airport contributes about 35% (10.9 µg/m^3) of the NO_2 concentration at the airport location and 4.6% (1.19 µg/m^3) of the NO_2 concentration in the city centre. A 63% reduction of the relative contribution is seen for every 4.2 km separation from the airport toward the city centre.

An overview of the total absolute contributions from the labelled sectors to the city centre locations is given in Figure 7.

Figure 7: The predicted absolute contributions from the various labelled sectors to the annual average surface NO_2 concentration in 2018 for the city centres in the vicinity of the airports of interest



Conclusions

The contribution of aviation emissions to atmospheric pollutant concentrations in Europe were assessed using the chemical transport model LOTOS-EUROS and its source apportionment feature that allows tracing of labelled emitted pollutants. In addition to the modelling simulation covering the whole European domain, six European cities (London, Paris, Amsterdam, Frankfurt am Main, Munich and Brussels) with large airports nearby were chosen for additional analyses.

Due to the spatial characteristics of the aviation contribution (i.e. mainly a local issue with respect to ambient air pollutant concentrations), an initial exploration into various emissions datasets was performed from which it was concluded that emissions data at the highest available resolution should be used for as many of the relevant pollutants as possible depending on the data availability.

The main findings from the study can be summarised as follows:

- An average contribution from aviation to the NO_2 concentration in the respective city centres of the six cities examined of 2.5% is predicted, ranging from 0.5% (Munich) to 4.6% (Amsterdam).
- For the other pollutants, the relative contribution is smaller with, respectively, 1.9%, 0.5% and 0.3% for SO_2, $PM_{2.5}$ and PM_{10}. This suggests that aviation is not a significant contributor to pollutant concentrations in the city centres compared to other sources, e.g. road transport and residential combustion.
- Closer to the airports, the average relative contributions from aviation to the NO_2 concentration in the six airports examined is significantly higher at 40%, varying from 26% (Zaventem) to 58% (Charles de Gaulle).
- This is also the case for the other pollutants, with aviation contributing, respectively, 45%, 6.2% and 4.6% to concentrations of SO_2, $PM_{2.5}$ and PM_{10}.
- The relative contribution of aviation declines as a function of distance from the airport. On average over the six airports examined, pollutant concentrations decrease with a reduction rate of 63% for every 3.8 km separation from the airport toward the city centre, ranging from 1.8 km (Brussels) to 4.9 km (Frankfurt am Main and Munich).

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Abbreviations and terms

ACT	Air Control Toolbox
AGR	Agriculture
AIS	Automatic Identification System
AMS	IATA code for Schiphol airport, the Netherlands
AOT40	Accumulated Ozone exposure over a Threshold of 40 ppb
C	Carbon
CAMS	Copernicus Atmospheric Monitoring Service
CAMS-REG	A state-of-the-art high-resolution European emission inventory for air quality modelling
CDG	IATA code for Charles de Gaulle airport, France
CBM-IV	Carbon Bond Mechanism IV — a chemical kinetics mechanism for simulating urban and regional photochemistry
CTM	Chemical Transport Model
DEPAC	DEPosition of Acidifying Compounds
E-PRTR	European Pollutant Release and Transfer Register
ECMWF	European Centre for Medium-Range Weather Forecasts
EEA	European Environment Agency
EMEP	European Monitoring and Evaluation Programme
ER	Emission Register (Netherlands)
ERA5	Fifth generation atmospheric reanalysis of the global climate covering the period from January 1940 to present, produced by the Copernicus Climate Change Service at ECMWF
FMI	Finnish Meteorological Institute
GAINS	Greenhouse gas and Air pollution INteractions and Synergies
GFAS	Global Fire Assimilation Service
GrETA	Gridding Emission Tool for ArcGIS (Germany)
IATA	International Air Transport Association
IFS	Integrated Forecasting System
IND	Industry
ISORROPIA II	A computationally efficient thermodynamic equilibrium model used for modelling aerosol gas systems
LHR	IATA code for Heathrow airport, UK
LOTOS-EUROS	A 3-D chemical transport model developed by TNO
LTO	Landing and Take-Off cycle *(aviation article)*
LTO	Long-Term Objective *(ozone article)*
N	Nitrogen
NMVOC	Non-Methane Volatile Organic Compound
NO	Nitrogen monoxide
NO_x	Nitrogen oxides
NO_2	Nitrogen dioxide
O_3	Ozone
ORY	IATA code for Orly airport, France
PM	Particulate matter
$PM_{2.5}$	Particulate Matter with an aerodynamic diameter of less than or equal to 2.5 µm
PM_{10}	Particulate Matter with an aerodynamic diameter of less than or equal to 10 µm
RH	Residential Heating
S	Sulphur
SNAP	Selected Nomenclature for Air Pollution
SOMO35	Sum Of Means Over 35 ppb
SO_2	Sulphur dioxide
STEAM	Ship Traffic Emission Assessment Model
TNO	Dutch Organisation for Applied Scientific Research
TRA	Road transport
VOC	Volatile Organic Compound

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