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Modelling the Impact of the Introduction of the EURO 6d-TEMP/6d Regulation for Light-Duty Vehicles on EU Air Quality

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Citation: de Meij, A.; Astorga, C.; Thunis, P.; Crippa, M.; Guizzardi, D.; Pisoni, E.; Valverde, V.; Suarez-Bertoa, R.; Oreggioni, G.D.; Mahiques, O.; et al. Modelling the Impact of the Introduction of the EURO 6d-TEMP/6d Regulation for Light-Duty Vehicles on EU Air Quality. *Appl. Sci.* **2022**, *12*, 4257. https://doi.org/10.3390/app12094257

Academic Editor: Johannes Schwank

Received: 10 March 2022
Accepted: 20 April 2022
Published: 22 April 2022

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Abstract: In this manuscript, we evaluated different emission scenarios for light-duty road transport to evaluate their impact on air quality in the EU, with a focus on a number of cities by means of the EMEP (European Monitoring and Evaluation Programme) modelling system. In addition to the reference case scenario, where exhaust emission factors from COPERT (Computer Programme to calculate Emissions from Road Transport) corresponding to the existing fleet were used, we also tested future potential scenarios considering: (a) all passenger cars and light commercial vehicles meet the EURO 6 emissions standard and EDGAR (Emission Database for Global Atmospheric research) EURO 6 emission factors; (b) all passenger cars and light commercial vehicles meet the EURO 6 emissions standard and real-world emission factors derived from actual Euro 6d-TEMP/6d vehicles. Results show how the replacement of old vehicles by newer ones with better emission control technologies can help improve air quality in the EU in terms of reductions in NO_2 and $PM_{2.5}$ concentrations. However, reduced NO_x emissions in cities (as foreseen in the two scenarios analysed) will cause tropospheric O_3 to increase.

Keywords: air quality; emission scenarios; Euro 6d-TMEP/6d

1. Introduction

Air pollution is a major environmental cause of death in Europe as well as globally [1,2]. Although emissions of air pollutants in the EU have been reduced over the past two decades, poor air quality continues to cause more than 300,000 premature deaths in the EU each year and 1.6–4.8 million globally, notably due to high concentrations of particulate matter (PM) and ground-level ozone [3,4]. The European population is exposed to levels above the air quality standards set out in EU legislation, with 21 to 30% exposed to levels above the PM10 standard [5]. EU air quality standards are less strict than the specific guideline values provided recently by the World Health Organisation [6]. Almost the entire European population is exposed to particulate matter ($PM_{2.5}$), and ozone well above the levels set out by the WHO [6].

Road transport is responsible for large quantities of nitrogen oxides (NO_x), particulate matter ($PM_{2.5}$), and volatile organic compounds (VOCs) emitted in the atmosphere (EEA's Transport and Environment Reporting Mechanism [7]), and therefore for the poor air quality, especially in traffic hotspots in urban areas, through primary and secondary air pollutants such as PM, ozone (O_3), and secondary aerosols (SA). Transport emissions harm our health and need to be drastically reduced without delay [8]. Although these emissions have decreased, they have declined less than foreseen over the last 20 years [7]. This could be explained by the fact that the reduction of NO_x emissions by diesel cars has shown to be more challenging, due to the difference between real-world NO_x emissions and certification limits under Euro 4 and 5 [9]. Since 1970, road transport activities increased in Europe by 1.7 times when compared to nowadays (2018) and increased by more than 30% over the past two decades. Despite the increase of fuel combustion in this sector, most air pollutant emissions have decreased over the past two decades (with the exception of NH_3) thanks to the implementation of emission control technologies. Recent studies by [10,11] found that in 2017, emissions from road transport were lower than in the previous year. In particular, emissions of nitrogen oxides decreased at a faster rate when compared to the decreased activity in [12], specifically by 3%, and PM_{10} and $PM_{2.5}$ decreased by 1.4% and 3.6%, respectively [5] thanks, among others, to the implementation of the latest emission control technologies in new light-duty vehicles as they entered EU vehicle fleets. However, several countries did not meet the reduction targets set by the National Emission reduction Commitments Directive (NECD) for 2020 [13]. Therefore, more action is required to meet the air quality standards [14].

Aiming at reducing the emissions from the transport sector, passenger cars, and light-commercial vehicles, the EU has introduced since the 1990s a series of directives and regulations, commonly known as Euro standards (Euro 1 to Euro 6). The most recent regulatory steps within Euro 6 standard, Euro 6d-TEMP and Euro 6d, require emission tests of light-duty vehicles over a combination of on-road routes and laboratory tests. These have resulted in an important decrease of tailpipe NO_x emissions for diesel vehicles and for ultrafine particle emissions from direct injection gasoline cars equipped with gasoline particulate filters [15–19], whereas the results for ammonia (NH_3) and carbon monoxide (CO) emissions of some diesel and gasoline vehicles showed elevated emissions for these compounds [17,20,21]. It is important to note that some of these pollutants remain either non-regulated for light-duty vehicles (NH_3) or not covered by the Real Driving Emissions legislation (CO and VOCs). Vehicles meeting the Euro 6d-TEMP/6d standard constitute however, only a small part of the European fleet. While the traditional exhaust emissions (e.g., NO_x, PM) are well known and documented, several recent studies [15–18] point out the need of controlling ammonia (NH_3) emissions in vehicle exhaust, as they contribute to secondary aerosol formation.

The objective of this work was to study the contribution of NMVOCs, PM, NO_x, and NH_3 emitted by vehicles to NO_2 concentrations and to the formation of secondary aerosol (SA) and ozone, which are major contributors to urban pollution in Europe. This is achieved by changing the emission characteristics of passenger cars and light-commercial vehicles in dedicated model simulations to represent different emission reduction scenarios. The scenarios modelled are counterfactual, simplified representations of fleet renewal that help us explore the air quality benefits that could be achieved thanks to the emission reductions that could be reasonably expected from recent Euro 6 light-duty vehicles. No changes in emissions from other on-road transport sources (heavy-duty vehicles, motorcycles, etc.) or from other non-transport sources are modelled.

A novel contribution of this study is linked to the use of real-world emission factors (as measured by the European Commission Joint Research Centre (JRC) Vehicle Emissions Laboratories (VELA) laboratory) as input for air quality modelling, to project the impact of renewed traffic emissions on air quality.

The paper is structured as follows. In Section 2, the methodology and a description of the emission inventory are detailed, together with a description of the Air Chemistry

Transport Model. In Section 3 we present the results, followed by concluding remarks in Section 4.

2. Methodology

In this study, we assemble three sources of information to simulate the impact of road transport emissions on air quality:

1. Real-world emission factors.
2. EDGAR emission inventory, to project emission factors to future vehicle fleets.
3. EMEP air quality model, to convert emissions to concentrations.

2.1. Data from the Laboratory and Real-World Emission Factors

The Sustainable Transport Unit (STU) at the JRC, located in Ispra (Italy), routinely measures exhaust emissions of light-duty vehicles (including passenger cars and light-commercial vehicles) in the laboratory and on the road for a variety of purposes using state-of-the-art instrumentation.

Data from twenty-seven Euro 6d-TEMP and three Euro 6d vehicles were gathered to develop powertrain-specific emission factors for gases and solid particulates for the needs of the present study. All vehicles were tested following the World Harmonised Light-duty Vehicle Test Procedure (WLTP), which is the current type-approval laboratory test procedure for passenger cars and light-commercial vehicles in the European Union (EU) [22]. Emission factors of Euro 6d-TEMP/6d light-duty vehicles have been derived ad-hoc for the study using data gathered by the STU as the 2019 version of the emission inventory guidebook, used within the Emissions Database for Global Atmospheric Research (EDGAR), which did not contain emission factors for such vehicles. More detailed information on the methodology to derive the emission factors in the VELA laboratory is given in Figures S1–S3 of the Electronic Supplement of this paper. Additionally, information about the road traffic emission factors in EDGAR are given in Tables S1 and S2.

2.2. EDGAR Emissions

The EDGAR is a global inventory providing greenhouse gas and air pollutant emissions estimates for all countries over the time period 1970 until the most recent years, covering all IPCC reporting categories, with the exception of Land Use, Land Use Change, and Forestry (LULUCF). Note that EDGAR follows the EEA Guidelines for reporting, with a highly detailed information/data on fleet composition.

In this study, we used the emissions for aerosol and aerosol precursor gases from the EDGAR version 5.0 inventory [23–25] as baseline scenario. Furthermore, we developed different emission scenarios coupling the activity data, technologies, and abatement measures implemented in the EDGAR database with the emission factors retrieved from experiments performed at the VELA Laboratory. More detailed information about the emission inventory is given in [26] and references therein.

2.3. EMEP Air Quality Model

The EMEP model version rv_33 has been used in this work. It is an off-line regional transport chemistry model [27], used to study the impact of road transport emissions on air quality over Europe. The domain stretches from -15.05° to 36.95° longitude and 30.05° to 71.45° latitude with a horizontal resolution of $0.1^\circ \times 0.1^\circ$ longitude/latitude. The model has 20 vertical levels, with the first level around 45 m. The model uses meteorological initial conditions and lateral boundary conditions from the European Centre for Medium Range Weather Forecasting (ECMWF-IFS). The meteorological year used was 2015. The gas-phase chemistry was based on the evolution of the so-called "EMEP scheme" as described in [27] and references therein. The chemical scheme couples sulphur and nitrogen chemistry to photochemistry, using about 140 reactions between 70 species. Aqueous phase chemistry describes the SO_2 oxidation by ozone and H_2O_2, and catalysis of O_2 in clouds, to form sulphate. An important pathway of aerosol nitrate formation is through the hydrolysis of

N_2O_5 on wet aerosol surfaces that converts NO_x into HNO_3 [27]. The EMEP model has two size fractions for aerosols, fine aerosol ($PM_{2.5}$) and coarse aerosol (PM10–2.5). The aerosol components presently accounted for are $SO_4^{=}$, NO_3^{-}, NH_4^{+}, anthropogenic primary PM, and sea salt.

The MARS equilibrium module is used to calculate the partitioning between gas and fine-mode aerosol phase in the system of $SO_4^{=}$, HNO_3^{-}, NO_3^{-}, NH_3^{-}, and NH_4^{+} [27]. More information on the gas and aerosol partitioning is given in [27], Section 7.6. Regarding secondary organic aerosols (SOA), the EmChem09soa scheme is used, which is a simplified version of the so-called volatility basis set (VBS) approach. The main differences between the VBS schemes and EmChem09soa is that all primary organic aerosol (POA) emissions are treated as non-volatile in EmChem09soa. This is done to keep the emission totals of both PM and VOC components the same as in the official emission inventories. The semi-volatile biogenic and anthropogenic SOA species are assumed to oxidise (also known as ageing process) in the atmosphere by OH reactions. This will lead to a reduction in volatility for the SOA, and thereby increase partitioning to the particle phase. More information on SOA is given in [28].

Detailed information on the meteorological driver, land cover, model physics, and chemistry are described in Simpson et al. [27] and in the EMEP Status Report 2017 [29].

2.4. Counterfactual Transport Emission Scenarios

Putting together these sources of information (real-world emission factors, EDGAR emissions, EMEP air quality model), three simulations were performed:

1. A Reference Case (REF): Road transport emissions correspond to the state of the fleet in the year 2015, i.e., a mixture of Euro 1 to Euro 6. The light-duty vehicles exhaust emission factors for all pollutants are those contained in the EDGAR inventory, using Tier 2 exhaust emission factor and Tier 3 fuel economy indicators presented in the EEA guidelines released in 2019 and based on the COPERT emissions model [30].
2. Scenario 1 (SC1): This counterfactual scenario assumes that all passenger and light-commercial vehicles in the simulated domain were approved under the Euro 6b standard, that is, vehicles approved between 2015 and 2018 following the NEDC test procedure which did not include on-road testing. Exhaust PM, NO_x, NH_3, and non-methane VOCs (NMVOCS) emission factors of all pre-Euro 6b gasoline light-duty vehicles are changed to the corresponding COPERT emission factors of Euro 6b vehicles, and equivalently for diesel vehicles. The emission factors of vehicles propelled with fuels other than gasoline and diesel (e.g., natural gas, liquified-petroleum gas) are kept unchanged. Emission factors for heavy-duty vehicles, motorcycles, and mopeds also remain unchanged. In short, SC1 aimed at simulating the effect in air quality of renovating the fleet of all light-duty vehicles in Europe to Euro 6b vehicles.
3. Scenario 2 (SC2): For this counterfactual scenario, all passenger cars and light-commercial vehicles are assumed to comply with the most recent and stringent type approval regulation applicable to Euro 6d-TEMP or Euro 6d vehicles (EC Regulation 2017/1151 and amendments). Euro 6d-TEMP and 6d vehicles, available in the market since late 2017, need to comply with emission limits in the updated laboratory test procedure (WLTP) and on the road following the Real Driving Emissions (RDE) regulations. In SC2, the exhaust emission factors of PM, NO_x, NH_3, and NMVOCs for gasoline and diesel light-duty vehicles are the ones estimated by JRC STU, as described earlier. Again, in this scenario, emission factors of light-duty vehicles using fuels other than gasoline and diesel are left unchanged as compared to the reference scenario. SC2 is designed to provide evidence of the effect on air quality of the new WLTP and RDE emissions regulations in Europe.

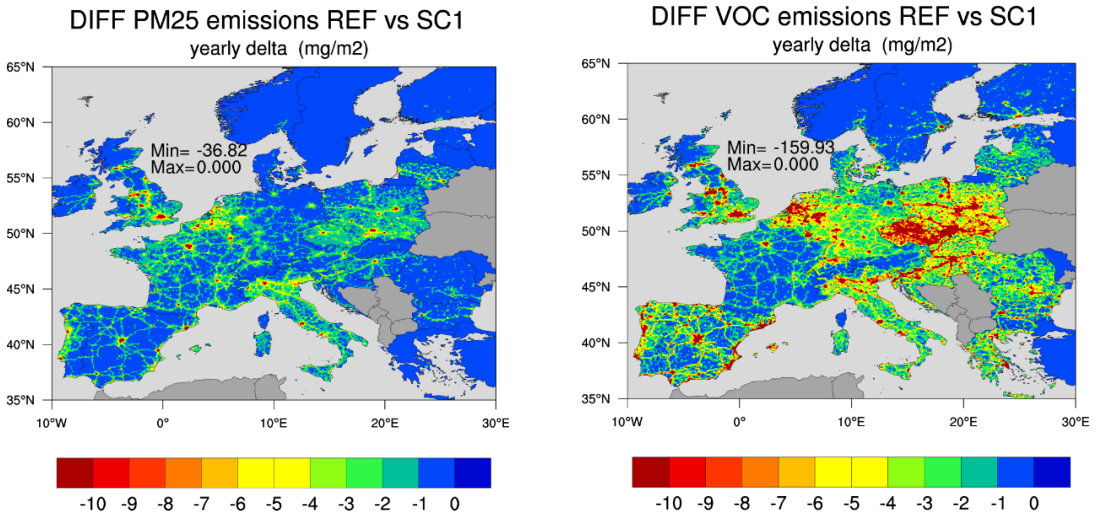
3. Results

In Section 3.1, we describe the emission used as input for the Reference Case and the two Scenarios over the EU domain. We subsequently show in Section 3.2 the impact of these scenarios on air pollutant concentrations calculated by the EMEP model.

3.1. Emissions Scenarios

In this section, we analysed the differences in terms of road transport emissions between the Reference Case and the two scenarios, i.e., SC minus REF. The impact of road transport emission reductions is analysed for the 27 EU Member States (EU27) plus Norway, Switzerland, and the United Kingdom. We consider EU27 + countries because these countries follow a common trend in the implementation of transport technologies.

As described earlier, emission changes apply only to gasoline and diesel passenger cars and light-commercial vehicles. In general, $PM_{2.5}$, NMVOC, NO_x, and NH_3 road transport emissions in the Reference Case are higher than in SC1, and those in SC1 are higher than in SC2 (see Figure 1). The unit of the emissions in the EMEP model are given in mg/m^2. The reductions in the emissions are clearly visible over highways and dense urban areas where road transport emissions mostly occur. For some countries (e.g., Germany, the Netherlands, Switzerland, Poland), higher NH_3 emissions reductions are found as compared to other countries (Spain, France). This can be explained by the difference in the share of diesel vehicles in each country's fleet and the fact that gasoline vehicles are responsible for most of the NH_3 emissions from light-duty vehicles. For example, Germany, the Netherlands, Poland, and the UK are the countries showing largest NH_3 reductions because these countries have a relatively low share of diesel vehicles (33.2, 16.8, 30.5, and 38.3% for the year 2017, respectively [31] and the pre-Euro 6 EDGAR/COPERT emission factor of NH_3 for gasoline vehicles is higher than the one attributed to Euro 6 gasolines, so that, when moving all pre-Euro 6 gasoline vehicles to Euro 6 gasoline vehicles, the mass emitted in Scenario 1 is lower than in the Reference case. For countries with a higher diesel share of the fleet (>56%, such as in the Baltic states, France, Luxembourg, Austria, or Spain), the change of scenario barely affects the total NH_3 mass emissions as the reduction on the gasoline contribution is lower than the increase of the diesel contribution. The increase of the NH_3 emission factor for diesel vehicles results from the more extensive use of the selective catalytic reduction (SCR) catalysts needed for large de- NO_x conversion.



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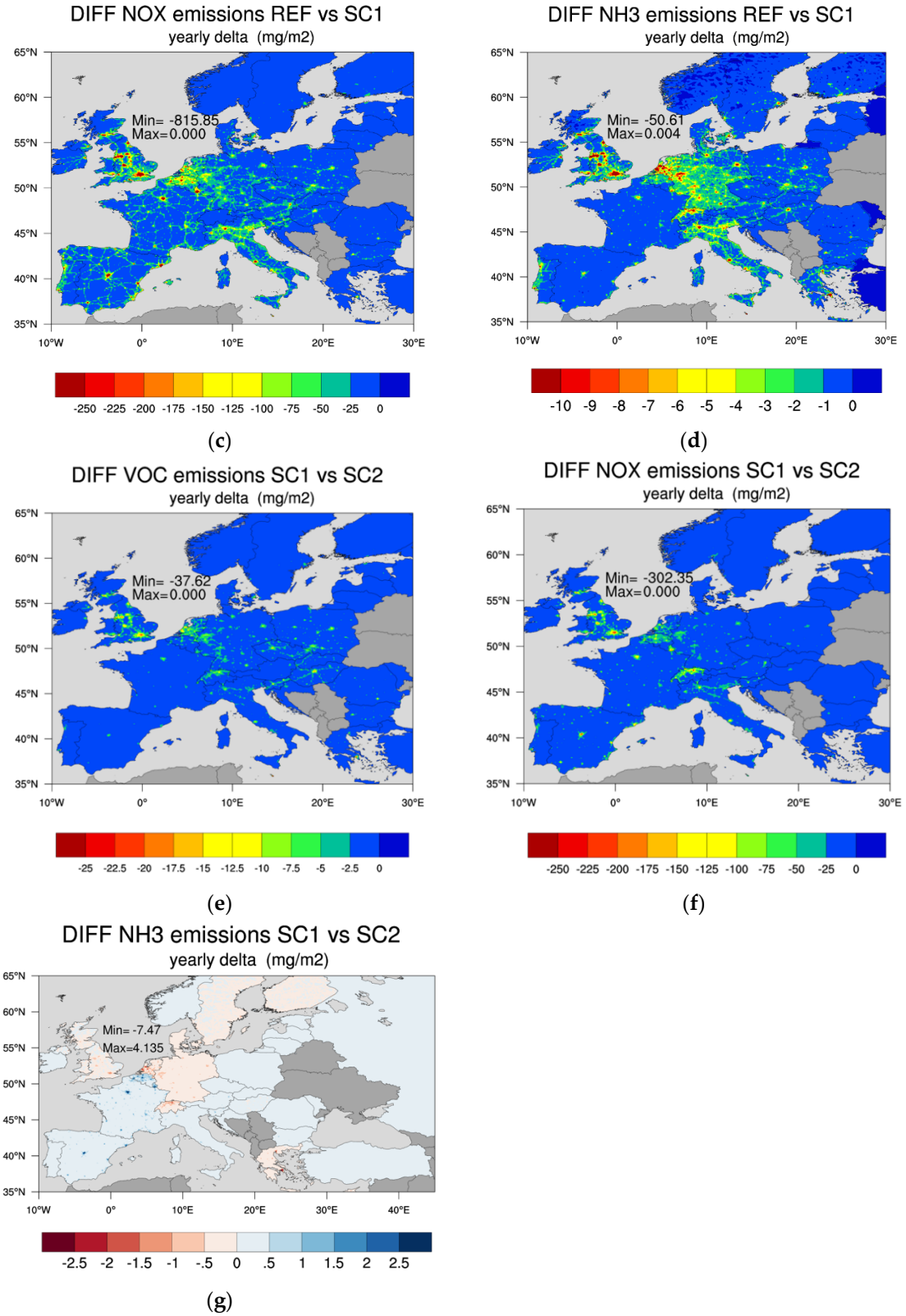


Figure 1. Yearly differences in road transport emission totals (mg/m^2) between the Reference Case and Scenario 1 for $PM_{2.5}$ (**a**), NMVOCs (**b**), NO_x (**c**), and NH_3 (**d**), together with the differences between Scenario 1 and Scenario 2 for NMVOCs (**e**), NO_x (**f**), and NH_3 (**g**).

It is interesting to note that general NH_3 emissions in SC1 are lower than SC2. In SC2, vehicles need to comply with stringent NO_x limits on the road. For that aim, diesel vehicles make a more intensive use of the after-treatment systems that need to operate throughout the whole driving duration, namely SCR catalysts. As a consequence of a wider operation of the SCR, NH_3 can be emitted, even more so if the vehicles do not mount ammonia slip catalysts. In Euro 6 pre-RDE vehicles (as in SC1), diesel vehicles resulted in very low NH_3 emissions, if any [20,32].

Between REF and SC1, the largest differences in NMVOCs are found over urban areas of the UK, the Netherlands, Poland, and Czechia, where gasoline vehicles have a relative high share of the total passenger car fleet, as diesel cars emit lower NMVOC emissions than gasoline engines [33] and Euro 6 gasoline vehicles emit less than pre-Euro 6 ones. However, Spain has a high diesel share (56.9%), and the relative difference in NMVOC emissions between the Reference Case and SC1 is up to ~30% lower by SC1 for Spain and up to ~25% lower for Czechia, considering the mass emissions on the whole country (not shown).

Differences in NO_x, NH_3, and NMVOCs between SC1 and SC2 are visible over large cities and highways (Figure 1e–g), but smaller than the Reference Case versus SC1 differences, because of the emission factors' change between Euro 6b and Euro 6d-TEMP, as described before. It is important, however, to stress than in SC2, there is an additional NO_x emissions reduction as compared to SC1, because diesel vehicles are responsible for the largest contribution of NO_x emissions as compared to cars using other fuel types; replacing all diesel cars to Euro 6b or Euro 6d-TEMP results in reducing the NO_x emissions by a factor of two for countries such as Germany, Spain, France, and Italy.

In our simulations, there were no differences in primary PM emissions between SC1 and SC2. The reason for this choice is that we focused on the impact on secondary aerosol formation only. Therefore, we changed only the aerosol precursor emissions between SC1 and SC2, but we kept primary PM emissions the same.

In real life, the primary particle emission of gasoline vehicles should be lower by Euro 6d-TEMP than Euro 6b, as Euro 6d-TEMP gasoline direct-injection cars mount gasoline particulate filters and Euro 6b cannot.

3.2. Impact on Concentrations

In this Section, the impact of the two scenarios on aerosol and gas pollutants were evaluated at ground level. We focused our analysis on yearly averages and on three main periods as done by [34], i.e., warm/summer period (May to September), mild/transition period (March, April, and October) and a cold/winter period (November to February), to highlight the seasonal dependency of the road transport emission reduction scenarios on NO_2, O_3, and $PM_{2.5}$ concentrations.

3.2.1. Nitrogen Dioxide (NO_2)

Reference Case versus Scenario 1

In general, yearly mean NO_2 concentrations by SC1 are lower over densely populated areas (e.g., Paris, London, Madrid, the Benelux, London, and the Po Valley, see Figure 2), which are characterised by intense traffic and main highways, up to 5.7 $\mu g/m^3$ (max up to −36.8%, see Figure S4 of the Electronic Supplement). The reduction in NO_2 concentration is relatively similar throughout the year, with a slightly absolute larger impact during the winter period, especially in the Po Valley and eastern parts of Europe, because these areas are characterised by frequent stagnant conditions and a low planetary boundary layer during winter period, leading to a build-up of air pollutants. The differences between the Reference Case and SC1 for the different periods are similar in absolute (around 5.8 $\mu g/m^3$) and relative terms (about 36.5%) as to the differences between the yearly averaged concentrations, indicating small differences during the various periods as mentioned before.

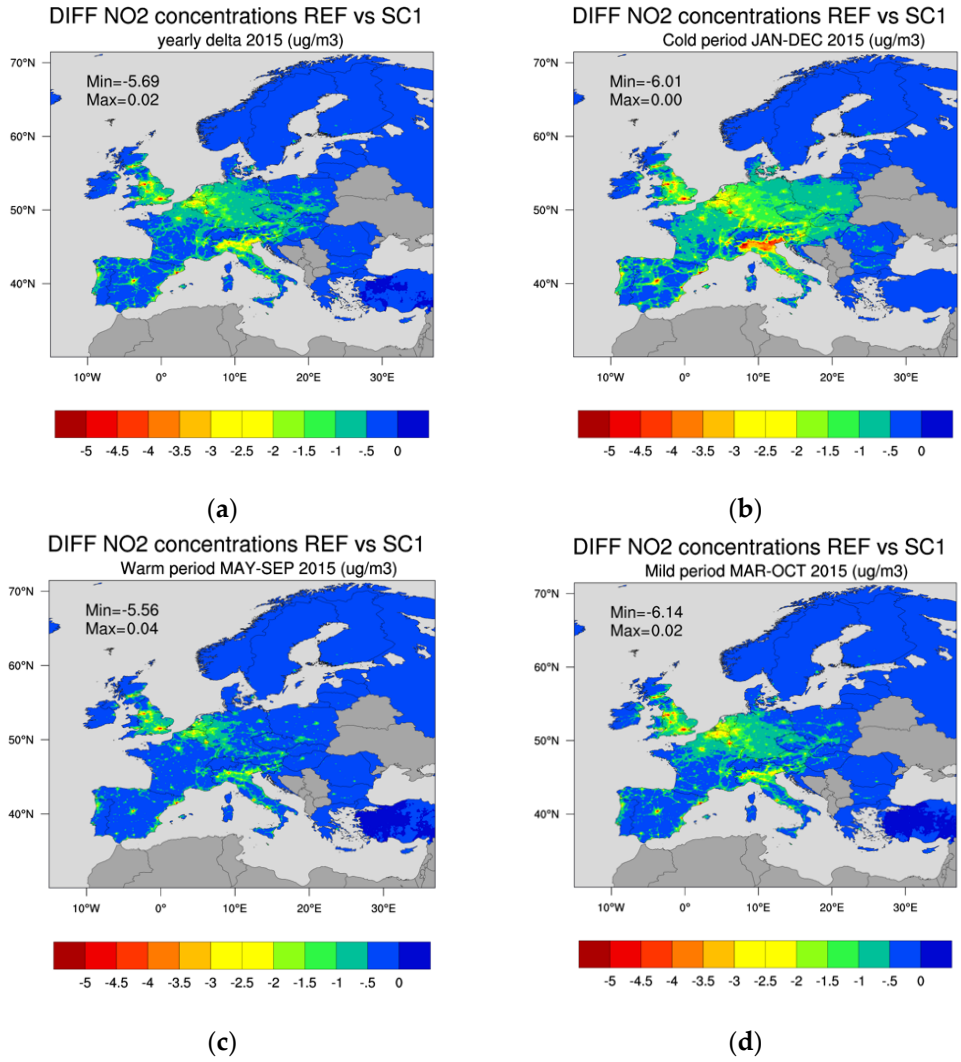


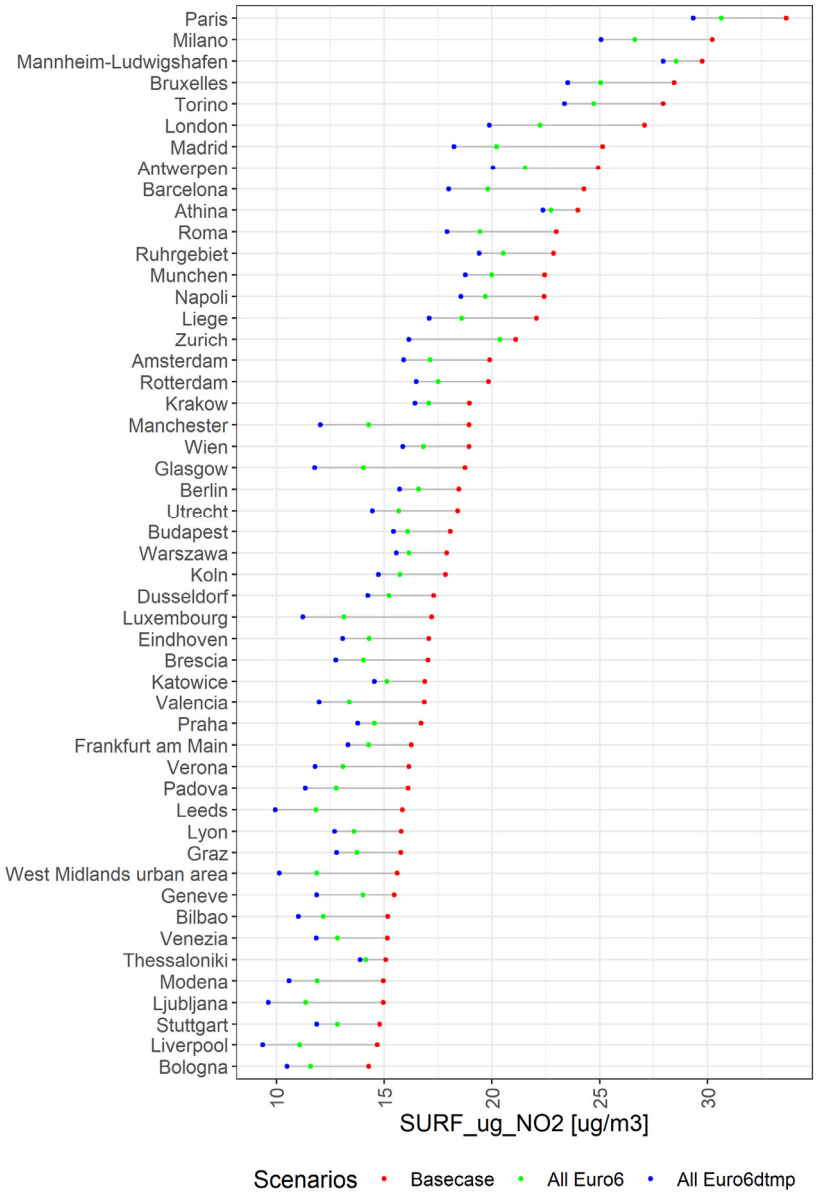
Figure 2. Calculated yearly mean differences in NO_2 concentrations ($\mu g/m^3$) between Reference Case and Scenario 1 (**a**), together with the differences for three main periods, i.e., (**b**) cold/winter period (November to February), (**c**) warm/summer period (May to September), and (**d**) mild/transition period (March, April, and October).

NO_x plays an important role in the formation and depletion of O_3 and the formation of secondary inorganic aerosols through complex chemical reactions. More will be explained in the next sections.

Scenario 1 versus Scenario 2

Differences in NO_2 concentrations between SC1 and SC2 reached a maximum reduction of 4.6 $\mu g/m^3$ (up to −41.2%. Figure S7 ES). This indicates that SC2 is more effective than SC1 in reducing NO_x emissions, mainly over urban areas where traffic is most intense. Diesel cars have the largest contribution to the total road transport NO_x emissions. The reason why SC2 shows the largest reduction over the northern part of Switzerland (not shown) is that road transport has a larger contribution of emissions to the total than other countries, and hence the change in emission factors is more evident. Additionally, most of the fleet is already Euro 6 and therefore the comparison of the Reference Case versus SC1 did not show major differences. During the summer period, the absolute differences are somewhat smaller than during the winter period. Applying Euro 6b (SC1) or Euro 6d-TEMP (SC2), shows that yearly concentrations are lower in the cities, with Euro 6d-TEMP showing the largest reduction in NO_2.

In Figure 3, the yearly NO_2 concentrations by Reference Case, SC1, and SC2 for 50 cities (the most polluted among the 150 cities considered in the $PM_{2.5}$ Atlas [35] are presented. For Bologna (~14 $\mu g/m^3$), the lowest ground level NO_2 concentration was found, while for Paris (~33 $\mu g/m^3$), the highest yearly average concentration was found for the Reference Case (these concentrations are the largest 7 km averaged value within the city functional urban areas [36] further denoted as "FUA"). Clearly, from this Figure we can see that implementing Euro 6b and Euro 6d-TEMP has a positive impact on the reduction of NO_2 concentrations at the city level.



(**a**)

Figure 3. *Cont*.

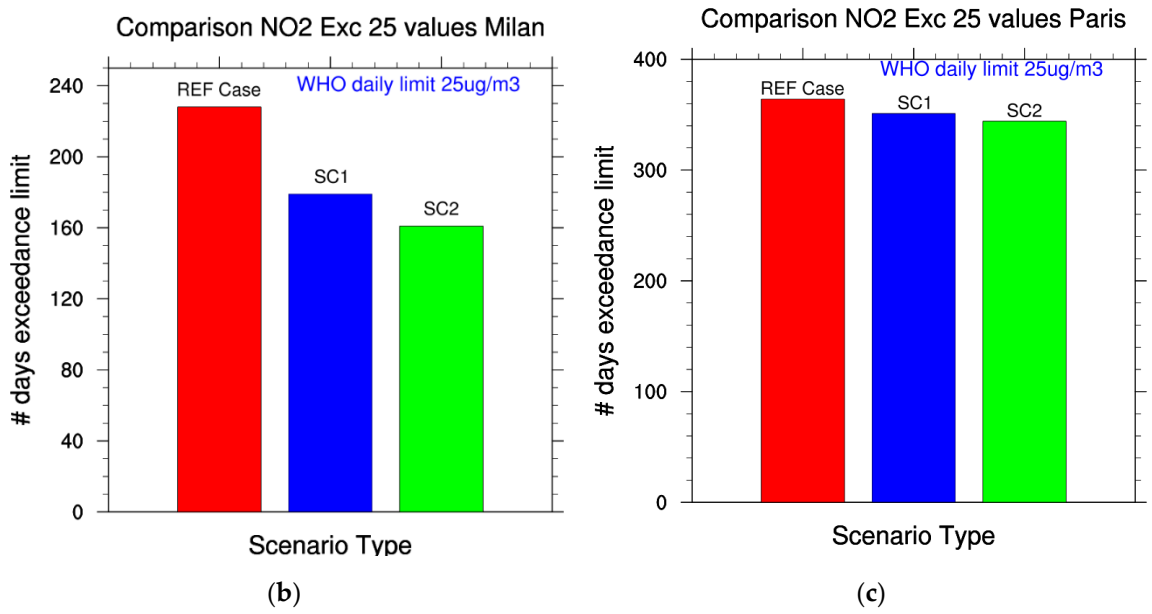


Figure 3. (**a**) NO_2 concentrations ($\mu g/m^3$) at city level for the Reference Case, and the two counterfactual emission reduction scenarios, together with the number (#) of exceedance days (Exc) of the WHO recommended concentration value for the city center of Milan (**b**) and Paris (**c**).

Milan and Paris are among the problematic cities in Europe where yearly average NO_2 concentrations are high. In order to understand how often people are exposed to these high NO_2 levels, we took a look at the number of days for which the WHO NO_2 daily recommended concentration value of 25 $\mu g/m^3$ (WHO new guidelines recommended high-ambition level by 2030) are exceeded in Figure 3b,c. We saw that for Milan, the NO_2 concentrations by the Reference Case exceeded 228 times per year and for Paris by 364 days per year. SC1 and SC2 show a decrease in the number of exceedance days, i.e., for Milan 179 and 161 times, respectively, and a reduction for Paris to 351 (SC1) and 344 (SC2) days per year. Euro 6d and Euro 6d-TEMP vehicles have therefore a clear impact on reducing NO_2 exposure levels in cities, although it is insufficient to reach the WHO guidelines.

3.2.2. Ozone (O_3)

Reference Case versus Scenario 1

In this section, we analysed O_3 concentrations in terms of the daily 8 h maximum indicator to exclude the low night-time O_3 values in urban areas.

The production of O_3 depends on the availability of NO_x and VOCs, both emitted by road transport, among other sectors. Whether reductions in NO_x or VOC emissions will lead to lower O_3 concentrations depends on the location and type of chemical regime, i.e., whether it is a NO_x-limited or a VOC-limited regime. For NO_x-limited regimes (e.g., locations downwind of urban environments), lowering NO_x emissions results in lower O_3 peak concentrations, whereas in VOC-limited areas (e.g., highly polluted urban areas), the opposite occurs, leading to higher O_3 concentrations. On the other hand, lowering VOCs and keeping NO_x constant always leads to reduced O_3 values.

Figure 4a shows the difference in annual mean O_3 concentrations between the Reference Case and SC1. What it interesting to see is the general increase in O_3 concentration (up to 2.5 ppb, ~8.6%, Figure S5 ES), particularly in densely populated areas (e.g., the Benelux, London, Liverpool, Manchester, Madrid, Barcelona, Paris, Milan, Rome, Naples, Lisbon, Porto, the Ruhr area, Berlin, and Dublin), which was associated with lower NO_2 and NMVOCs emissions. This side-effect of cleaner vehicles could lead to higher O_3 concentrations and possible exceedances in cities that are currently below the O_3 limits.

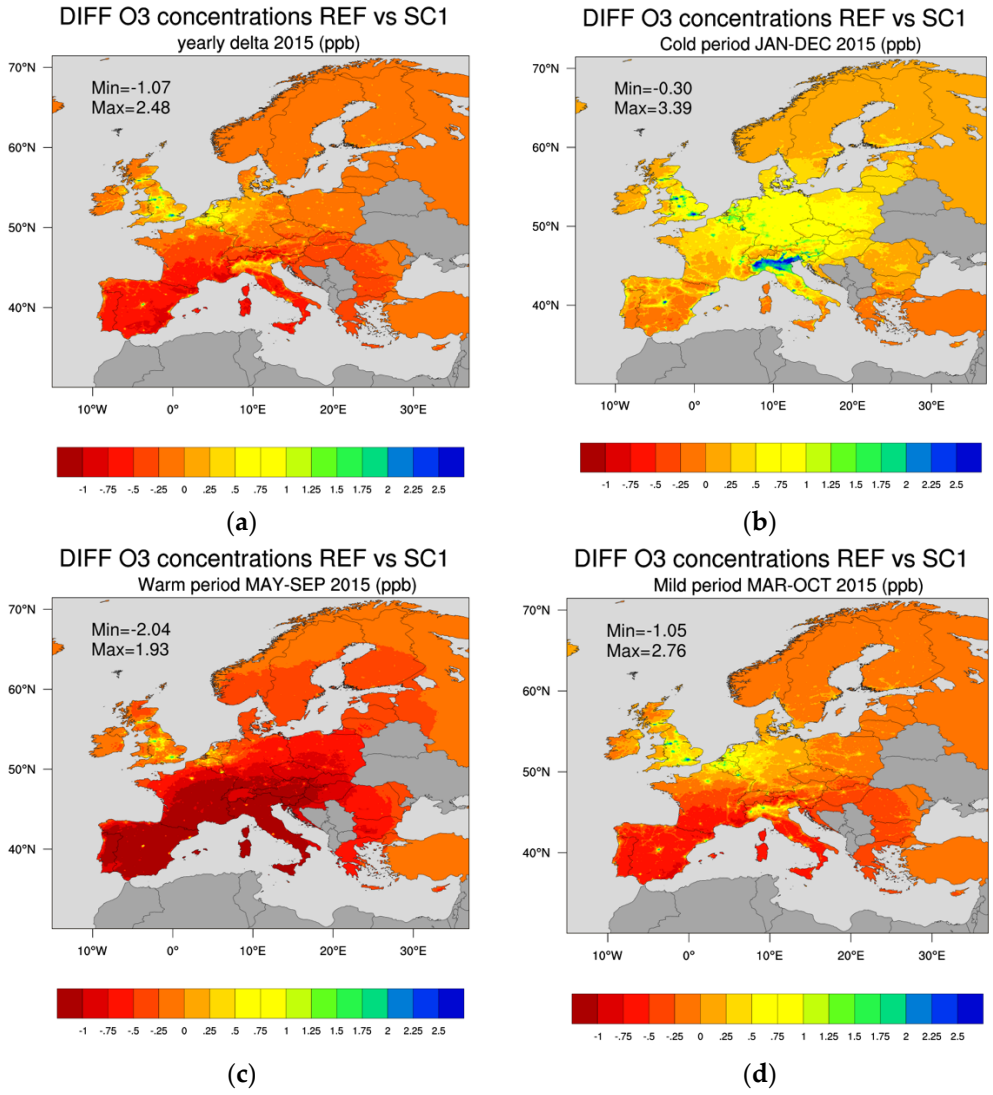


Figure 4. Calculated yearly mean differences in O_3 concentrations (ppb) between Reference Case and Scenario 1 (**a**), together with the differences for three main periods, i.e., (**b**) cold/winter period (November to February), (**c**) warm/summer period (May to September) and (**d**) mild/transition period (March, April, and October).

The underlying reason for this increase is that less O_3 is removed by NO (NO_x titration), therefore augmenting O_3 values in VOC-limited zones, as mentioned earlier. NO_x titration is an important removal mechanism of O_3 during night and wintertime [37]. Therefore, we see in general an increase in O_3 values by SC1, especially during the cold period in Figure 4b over Eastern Europe and the Po Valley (up to 19.2%).

During the warm period (May to September), there is a slight reduction of O_3 concentration over non-urban areas for the countries in the Mediterranean basin (Spain, France, Italy, up to ~2 ppb) where the concentrations are usually the highest. In principle, these reductions in O_3 could lead some regions to fulfil the current legal limit of 120 $\mu g/m^3$ (8 h maximum). Yet, higher O_3 values were found, due to the reduction of NO_x over the densely populated areas, as described above. What is interesting to see is that the seasonal variations of O_3 concentrations are more distinct than for NO_2, as described earlier. An explanation for this is that the emissions of NO_x by traffic are rather constant throughout the year, whilst O_3 is chemically formed by the oxidation process of volatile organic compounds (VOCs) in the presence of NO_x (NO + NO_2) and its formation is driven by the sunlight's intensity. The latter has a strong seasonal profile.

Scenario 1 versus Scenario 2

Yearly mean O_3 concentrations by SC2 are somewhat lower than SC1, with a minimum of 0.6 ppb. Over urban areas, O_3 values by SC2 are a little bit higher than SC1, explained by the NO_X titration, leading to higher O_3 concentrations in VOC-limited areas, as described before. Maps (Figure S8 ES) from Scenario 1 to Scenario 2 show similar results, with changes mainly localised in limited areas.

Figure 5 shows that SC1 and SC2 have, for some cities, a negative impact on the reduction of O_3 concentrations at the city level (e.g., Munich, Milan, Turin). At the same time, the Figure also shows that for most cities (27 out of 50), it has a moderate positive effect on O_3 concentration levels.

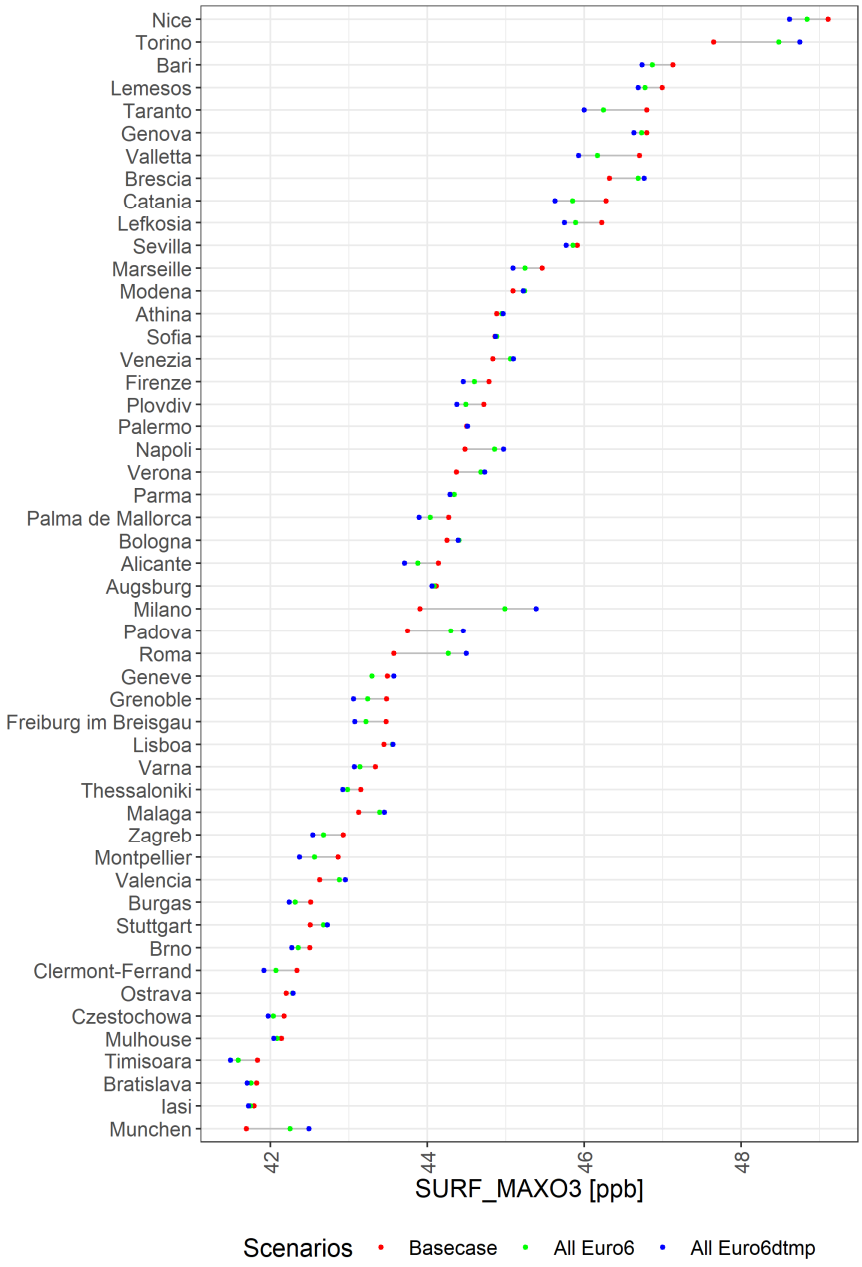


Figure 5. O_3 concentrations (ppb) at the city level for the Reference Case, and the two counterfactual emission reduction scenarios.

3.2.3. Particle Matter ($PM_{2.5}$)

Reference Case versus Scenario 1

PM2.5 yearly mean concentrations for SC1 show, in general, a reduction over the western and central part of Europe, see Figure 6. The contribution of road transport to the total emissions of $PM_{2.5}$ is relatively low (~9.9% EU average [38]) and do not show large reductions in cities or highways. The largest reductions occurred in the Po Valley (up to 2.2 $\mu g/m^3$, or ~12%, Figure S6 ES), where traffic contributes significantly to the total PM emissions [39]. During the warm period, the relative reductions in PM concentrations were higher in the Po Valley (up to 11%) than during the cold period (up to 4%), although absolute reductions were of the same magnitude (~2 $\mu g/m^3$), which can be explained by the absence of other emission sources such as domestic heating (prevalent during winter time), and hence the relative larger weight of vehicle emissions during the warm period.

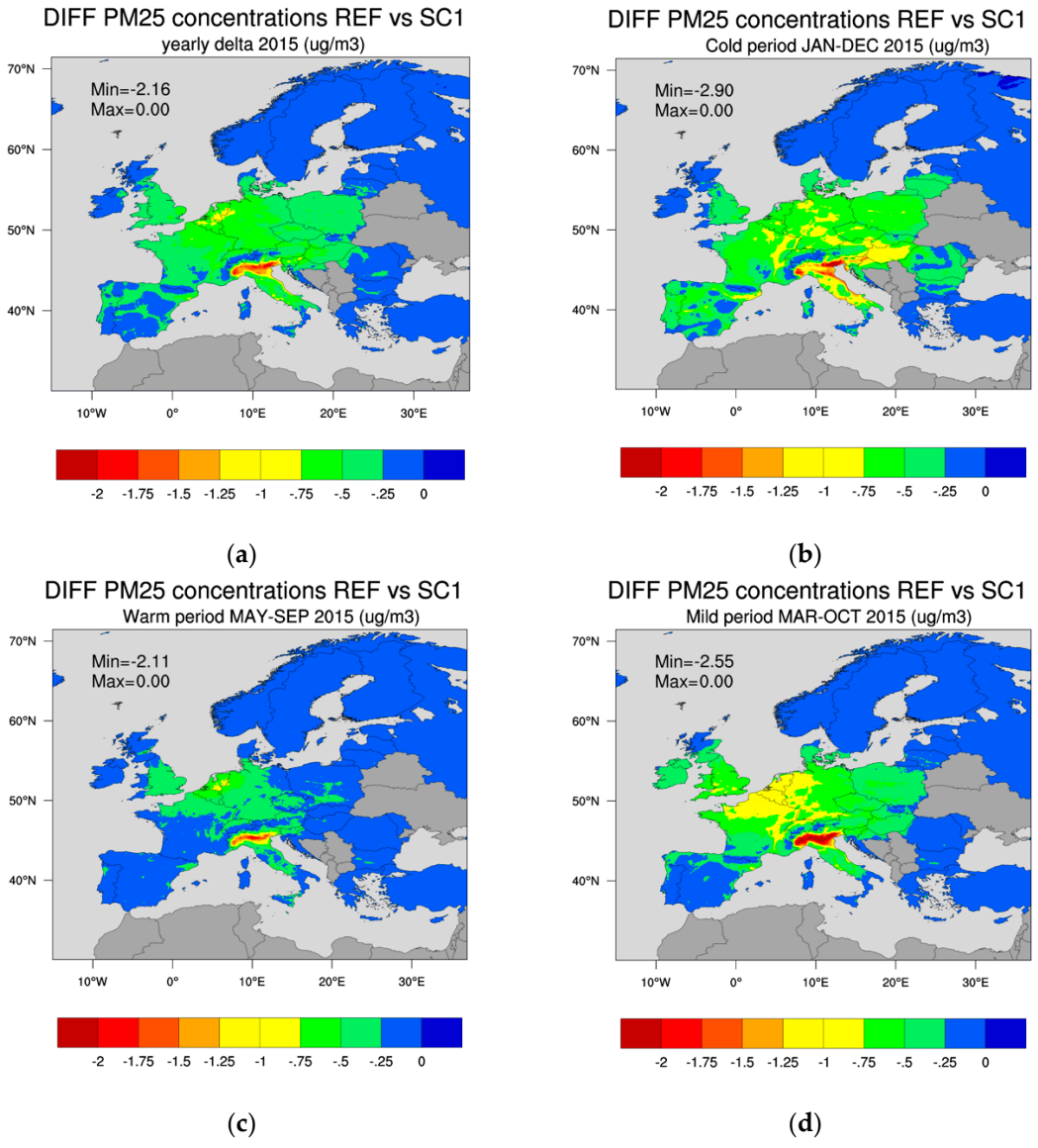


Figure 6. Calculated yearly mean differences in $PM_{2.5}$ concentrations ($\mu g/m^3$) between Reference Case and Scenario 1 (**a**), together with the differences for three main periods, i.e., (**b**) cold/winter period (November to February), (**c**) warm/summer period (May to September), and (**d**) mild/transition period (March, April, and October).

Poland, Czechia, and Hungary have around 30% of diesel vehicles, pre-Euro 5 light-duty vehicles, in their fleets (i.e., without a particle filter). Replacing old diesel vehicles by Euro 6 ones with a filter (that reduce tailpipe emissions by up to two orders of magnitude [40,41]) result in a reduction in PM emissions as observed in Figure 1, and lower $PM_{2.5}$ concentrations as shown in Figure 6. However, the large reduction in $PM_{2.5}$ emissions is not fully reflected in the reduction of $PM_{2.5}$ concentrations. A possible explanation could be that other emission sources are weakening the effect of the filters in diesel cars in these countries [42].

During the cold period, the largest reductions in $PM_{2.5}$ concentrations were found over the Po Valley, and were up to 2.9 $\mu g/m^3$. Looking at individual days, we found a reduction of up to 9 $\mu g/m^3$ for the city of Milan on 27 October (daily average $PM_{2.5}$ on 27 October is 100.8 $\mu g/m^3$ by the Reference Case).

Even though the NH_3 emitted by vehicles is lower than NH_3 emitted from agricultural sources, the NH_3 emitted by cars in urban areas, close to NO_x and SO_2 sources, is likely to increase the $PM_{2.5}$ concentration considerably, because of non-linearities in the chemical regimes that lead to the formation of secondary inorganic aerosols [34,43]. Therefore, it is important to reduce also aerosol precursors which are emitted by the road transport sector.

Our simulations showed that reducing $PM_{2.5}$, NO_x, NMVOC, and NH_3 emissions in urban areas (Figure 7) contribute to a reduction in $PM_{2.5}$ concentrations of up to ~2.9 $\mu g/m^3$ in the Po Valley during the cold period, as mentioned earlier.

However, the complexity of secondary inorganic $PM_{2.5}$ formation is high, as described in [34,43]. It is, for that reason, key to identify the main precursors on which to act and to which extent when air quality plans are designed. For example, a study by [43] showed that in the Po Valley, the formation of secondary $PM_{2.5}$ is characterised by contrasting chemical regimes within distances of a few hundreds of kilometres, as well as non-linear responses to emission reductions during wintertime. They also showed that for some areas in the Po Valley, a small increase in $PM_{2.5}$ values were found when NO_x emission reductions were applied in NO_x-rich areas. During COVID-19 lockdown, NO_x emissions were in general lower in many cities due to the absence of road transport, but a significant reduction in $PM_{2.5}$ concentrations in many European cities was surprisingly not found [44,45]. The reason for that absence of the reduction or even a small increment in $PM_{2.5}$ concentrations is that over urbanised areas, lower NO_2 concentrations at constant or similar NMVOC concentrations lead to an increase of O_3 values, which is a reactive oxidant. Therefore, increasing levels of O_3 concentrations lead to an enhancement of the atmospheric oxidising capacity, which might lead to an increase in the Secondary Organic Aerosol (SOA) formation [46–50]. For example, SOA is formed through a series of chemical reactions of gaseous precursors, mainly volatile, intermediate-volatile, or semi-volatile organic compounds (IVOCs) with O_3, OH, and nitrate radical oxidants (NO_3) [50]. IVOCs are mainly emitted by the road transport sector.

Scenario 1 versus Scenario 2

Yearly $PM_{2.5}$ concentrations are lower by SC2 than SC1, up to 1 $\mu g/m^3$ (~8%). We could see (ES Figure S9) differences between SC2 and SC1 in the geographical distribution of the concentrations between the different seasons, although the differences were relatively small, up to ~1.51 $\mu g/m^3$ during the mild/transition period.

In Figure 8, we can see that Warsaw and Milan are some of the problematic major cities in Europe for which yearly average $PM_{2.5}$ levels are very high. In order to understand how often people are exposed to high PM levels, we looked at the number of days in the year for which the WHO $PM_{2.5}$ daily limit value had exceeded. As there is no EU daily limit value for $PM_{2.5}$, we evaluated how often the WHO-recommended, high-ambition level (by 2030) of 15 $\mu g/m^3$ was exceeded in Figure 8a,b. We can see that for Warsaw, $PM_{2.5}$ concentrations by the Reference Case were exceeded 323 times per year, and for Milan 249 times. SC1 and SC2 show a decrease in the number of exceedance days, i.e., for Warsaw 318 and 317 times for SC1 and SC2, respectively, and a larger reduction for Milan, being 228 (SC1) and 223 (SC2).

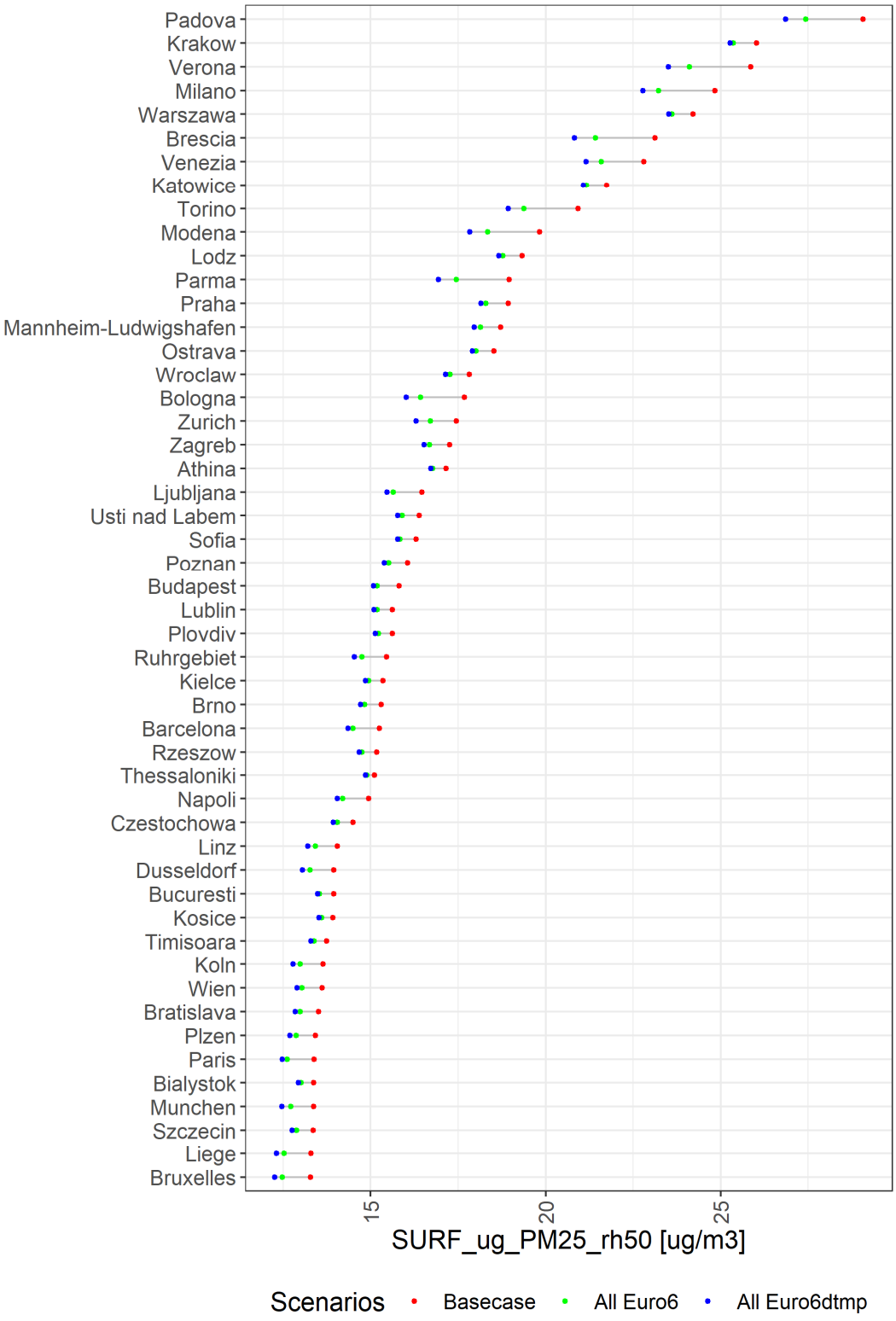


Figure 7. $PM_{2.5}$ concentrations ($\mu g/m^3$) at city level for the Reference Case, and the two counterfactual emission reduction scenarios.

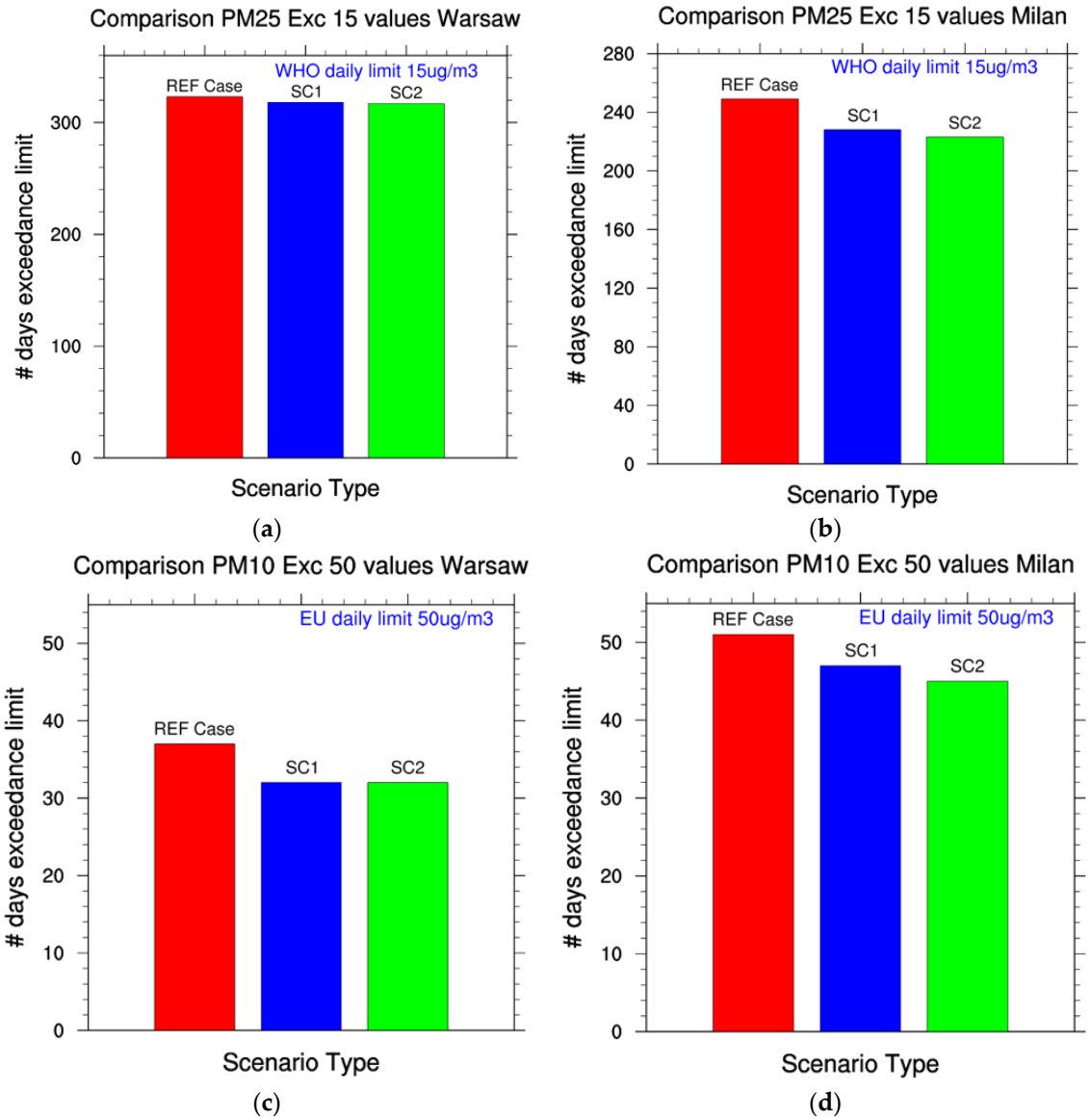


Figure 8. The number (#) of days for which the WHO $PM_{2.5}$ limit values of 15 $\mu g/m^3$ (**a**,**b**) and PM_{10} European limit value of 50 $\mu g/m^3$ are exceeded for Warsaw and Milan (**c**,**d**).

Evidently, also the EU PM_{10} limit value of 50 $\mu g/m^3$ is exceeded by the Reference Case. Under current EU air quality legislation, this daily limit value may not be exceeded more than 35 times per year. We can see that for Warsaw and Milan, the PM_{10} limit values are exceeded 37 and 51 times per year respectively, see Figure 8c,d.

In SC1 and SC2, the number of exceedance days for Warsaw reduced to 32 days for both scenarios. While for Milan, the number of exceedance days dropped to 47 and 45 days for the two scenarios, respectively. This indicates that applying the counterfactual scenarios reduces, to some extent, the number of days for which the daily limit values of PM_{10} are exceeded, which is important for decision-makers.

4. Conclusions

In this work we studied the contribution of NMVOCs, PM, NO_x, and NH_3 emitted by vehicles to the formation of Secondary Aerosol and urban air pollution in Europe. We performed model simulations with the Edgar emission inventory (V5) and changed the EU 2015 (a combination of Euro 1 to Euro 6) emission characteristics of light-duty vehicles with two different counterfactual emission reduction scenarios, one where all vehicles are

modelled as Euro 6b standard (SC1) and one where all vehicles meet the most stringent Euro 6d-TEMP/6d standard (SC2).

The results for both counterfactual scenarios show that total NO_x, NMVOC, and NH_3 emissions by diesel and gasoline vehicles are reduced when compared to the reference scenario (with a mix of Euro 6 and pre-Euro 6 vehicles in the national vehicle fleets). For NO_x, we found a reduction of almost 50% in total emissions for Italy, Spain, Germany, and France. Since diesel vehicles are responsible for the largest contribution of NO_x emissions as compared to cars using other fuel types, replacing all diesel cars to Euro 6b or Euro 6d-TEMP results reduces the NO_x emissions by a factor of two for countries such as Germany, Spain, France, and Italy. This is very relevant, as these countries (together with Poland and UK) are responsible for the largest road transport emissions. A strong decrease was also found in total emissions per country and for NMVOCs and NH_3 for both counterfactual scenarios.

Road transport emissions of $PM_{2.5}$, NMVOC, NO_x, and NH_3 in the Reference Case are higher than in either SC1 or SC2. NO_x and NMVOC emissions by SC2 are lower than SC1 over urban areas, whilst for some areas NH_3 mass emissions are higher by SC2, depending on the diesel share of the fleet.

The impact of these emission changes on concentration highlights that for yearly mean NO_2 concentrations, the Reference Case shows higher NO_2 values than SC1 over urban areas, roads, and highways, being up to 5.7 $\mu g/m^3$. During the cold, warm and transition periods, the differences between SC1 and the Reference Case are similar to the yearly differences. Also, NO_2 concentrations by SC_2 are lower than SC1, up to 41% (4.6 $\mu g/m^3$), mainly over urban areas. At the city level, we showed that both SC1 and SC2 have a significant impact on reducing NO_2 exposure levels.

Yearly mean $PM_{2.5}$ concentrations are higher in the Reference Case (up to 2.2 $\mu g/m^3$). The largest differences were found over the Benelux and the Po Valley. During the cold, warm and transition periods, the differences between SC1 and the Reference Case are similar to the yearly differences. Similar, yearly $PM_{2.5}$ concentrations by SC2 are lower than SC1, up to ~7% (~1 $\mu g/m^3$). Note that when we analysed the $PM_{2.5}$ concentrations for individual days, we found a reduction of up to 8 $\mu g/m^3$ for the city of Milan on 26 October (daily average $PM_{2.5}$ is 56.2 $\mu g/m^3$ by REF). This highlights the importance of analysing the daily limit values as well. For cities for which $PM_{2.5}$ concentrations are problematic, the number of days for which the daily limit values are exceeded dropped drastically when Euro 6d and Euro 6d-TEMP emission factors are implemented.

Throughout the year, SC1 shows higher O_3 concentrations as compared to the Reference Case (up to ~2.5 ppb) over urban areas, with the largest differences found during the cold period. The reason for this is that when NO_x emissions are reduced in VOC-limited areas, higher O_3 concentrations are found, because less O_3 is removed by NO_x titration. During the warm period, higher O_3 monthly mean values were found by the Reference Case over southern Europe, with the exception of the Benelux and urban areas. The largest differences between SC2 and SC1 were found during the cold period (~3.8 ppb), mainly over urban areas due to titration.

The side-effect of cleaner vehicles could lead to higher O_3 concentrations and possible exceedances in cities that are currently below the O_3 limits. A similar behaviour was also found for $PM_{2.5}$, where a small increase in PM2.5 concentrations was found over urbanised areas when NO_x emissions were reduced. For this reason, it is important to identify the main precursors on which to act and to which extent when air quality plans are designed.

Both counterfactual scenarios modelled in this study were used to explore, in a simplified manner, the impact of the Euro 6 standard. SC1, in particular, indicates that pre-RDE Euro 6 vehicles have had a positive contribution to air quality, and SC2 points to the further potential for the latest Euro 6 standard to continue to deliver air quality improvements before vehicles compliant with the new Euro 7 standard (currently under preparation) are gradually introduced into EU vehicle fleets in the second half of the present decade.

However, even if this emission reduction potential were fully realised, this would not translate into compliance with current EU air quality standards in many EU urban areas.

Further reductions than modelled in this study are in any case possible through a combination of a technology-neutral Euro 7 standards with lower emissions, through the accelerated introduction of zero tailpipe emission light-duty vehicles and also with emission standards and other measures aimed at reducing the emissions from sectors other than road transport (none of which were modelled in our scenarios).

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/app12094257/s1, ref. [17,20,51–55]. Figure S1: Instrumentation and sampling scheme in VELA laboratory; Figure S2: Test cycles speed profile WLTC; Figure S3, Set up and vehicle in the VELA laboratory; Table S1: Emission Factors for Road Traffic in Edgar; Table S2: Average fuel-specific emission factors of diesel and gasoline Euro 6d-TEMP/Euro 6d vehicles over WLTP; Figure S4: Calculated yearly relative mean differences (%) between Reference Case and Scenario 1 for NO_2. Together with the differences in averaged concentrations between the two simulations for three main periods, i.e., summer period (May till September), transition period (March, April and October) and a winter period (November till February); Figure S5: Same as Figure S4, but for O_3; Figure S6: Same as Figure S4, but for PM_{25}; Figure S7: Calculated yearly mean relative differences (%) between Scenario 1 and Scenario 2 for NO_2. Together with the differences in averaged concentrations between the two simulations for three main periods, i.e., summer period (May till September), transition period (March, April and October) and a winter period (November till February); Figure S8: Same as Figure S7, but for O_3; Figure S9: Same as Figure S7, but for PM_{25}.

Author Contributions: Conceptualisation, A.d.M., P.T. and E.P.; methodology, A.d.M., P.T. and E.P.; software, A.d.M., M.C., D.G., O.M. and G.D.O.; validation.; A.d.M., P.T., E.P., V.V. and R.S.-B.; formal analysis, A.d.M., P.T., E.P., V.V. and R.S.-B.; investigation, A.d.M., P.T., E.P., V.V. and R.S.-B.; resources, A.d.M., M.C., D.G., O.M. and G.D.O.; writing—original draft preparation A.d.M., P.T., E.P., V.V. and R.S.-B., writing—review and editing, A.d.M., C.A., P.T., E.P., O.M., G.D.O., V.V., R.S.-B. and V.F.; project administration, C.A.; funding acquisition, C.A. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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