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Country- and manufacturer-level attribution of air quality impacts due to excess NO_x emissions from diesel passenger vehicles in Europe

Guillaume P. Chossière[a], Robert Malina[a,b], Florian Allroggen[a], Sebastian D. Eastham[a], Raymond L. Speth[a], Steven R.H. Barrett[a,*]

[a] Laboratory for Aviation and the Environment, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA, 02139, USA
[b] Centre for Environmental Sciences, Hasselt University, Martelarenlaan 42, 3500, Hasselt, Belgium

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ABSTRACT

In 2015, diesel cars accounted for 41.3% of the total passenger car fleet in Europe. While harmonized emissions limits are implemented at the EU level, on-road emissions of diesel cars have been found to be up to 16 times higher than those measured in test stands. These excess emissions have been estimated to result in increased $PM_{2.5}$ and ozone exposure causing approximately 5000 premature mortalities per year in Europe. Interventions aimed at mitigating these damages need to take into account the physical and political boundaries in Europe, where emissions from one country may have an impact on neighboring populaces (trans-boundary impacts). To date, the trans-boundary implications of excess NO_x emissions in Europe are not understood and only excess NO_x emissions have only been studied at the fleet level and for Volkswagen group cars. In this study, a distribution of emission factors is derived from existing on-road measurements for 10 manufacturers, covering 90% of all new vehicle registrations in Europe from 2000 to 2015. These distributions are combined with inventory data and driving behavior to quantify excess emissions of nitrogen oxides (NO_x) in Europe in 2015. To quantify the changes in $PM_{2.5}$ and ozone concentrations resulting from these emissions, we use a state-of-the-art chemical transport model (GEOS-Chem). Concentration-response functions from the epidemiological literature are applied to estimate the premature mortality outcomes and the number of life-years lost associated with degraded air quality. Uncertainty in the input parameters is propagated through the analysis using a Monte Carlo approach. We find that 70% of the total health impacts from excess NO_x are due to trans-boundary emissions. For example, 61% of the impacts in Germany of total excess NO_x emissions are caused by emissions released in other countries. These results highlight the need for a coordinated policy response at the European level. In addition, we find that total emissions accounting for country-specific fleet mixes and driving behaviors vary between manufacturers by a factor of 10 and mortality impacts per kilometer driven by a factor of 8. Finally, we find that if all manufacturers reduced emissions of the vehicles currently on the road to those of the best-performing manufacturer in the corresponding Euro standard, approximately 1900 premature deaths per year could be avoided.

1. Introduction

Degraded air quality in Europe has been estimated to result in approximately 400,000 premature mortalities annually (EEA, 2015). These effects are mostly caused by population exposure to fine particulate matter with an aerodynamic diameter of 2.5 μm or less ($PM_{2.5}$) and, to a lesser extent, ozone. According to the Organization for Economic Co-operation and Development (OECD, 2014), road transport accounts for 50% of these impacts. In order to reduce the road transportation-related part of this total, European regulators introduced the Euro vehicle emissions standards in 1991 (EC, 1991). Since then, they have progressively tightened the standards. Recent studies have found that the standards have successfully curbed primary $PM_{2.5}$ exhaust emissions as well as other pollutants (Shindell et al., 2011).

Recently, the European Environment Agency (EEA) revealed significant differences between NO_x emissions measured during approval tests and real-world emissions. This has since been confirmed by various independent measurements (AMS, 2016; BMVI, 2016; DfT, 2016; DUH, 2016; MEEM, 2016; Thompson et al., 2014). Investigations started in 2015 into Volkswagen shed light on potential causes of the issue, as the manufacturer has been shown to use specific devices to activate emissions control equipment only when a car is being tested and deactivate them in real-world conditions. However, several on-road measurement campaigns (AMS, 2016; BMVI, 2016; DfT, 2016; DUH,

* Corresponding author.
E-mail address: sbarrett@mit.edu (S.R.H. Barrett).

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2016; MEEM, 2016; Thompson et al., 2014) revealed that the problem was not limited to Volkswagen. Permissive testing procedures at the EU level and defective emissions control strategies (Degraeuwe and Weiss, 2016) have been found to legally allow higher NO_x emissions on the road than in a laboratory setting.

Previous studies have quantified the overall impacts of excess NO_x emissions in continental Europe (Anenberg et al., 2017; Jonson et al., 2017), but the relative contribution of individual manufacturers beside Volkswagen (Oldenkamp et al., 2016; Chossiere et al., 2017) to the total impacts have not been analyzed. Furthermore, given the trans-boundary nature of pollution, the efficacy of national-level regulation to mitigate air quality impacts remains unknown. This study therefore quantifies the trans-boundary impacts of excess NO_x emissions, as well as manufacturers' contribution to the total impacts. It is also the first to account for the heterogeneity of excess emissions with fleet mix between countries, thereby significantly improving the representation of on-road emissions and their impacts at the country-level, including a quantification of uncertainties in core parameters such as driving behavior and emissions factors. For each country, this study estimates the total number of premature mortalities due to excess NO_x emissions and estimates the relative importance of domestic and foreign contributions. Manufacturer-level impacts are presented on a per car, per vehicle-kilometers traveled (VKT), and per unit excess NO_x basis. These metrics distinguish the relative contributions of fleet mix, driving behavior, and geographic location of the emissions in each country.

For this purpose, this study produces a detailed inventory of the diesel car fleet on European roads in 2015, which we combine with an activity model to estimate the total number of vehicle-kilometers traveled (VKT) annually. We distinguish between three categories of vehicles based on the applicable emissions standard: pre-Euro 5, Euro 5, and Euro 6. We consider ten groups of manufacturers: the Volkswagen group (referred to as VW, which includes its brands Audi, Seat, Skoda, and Volkswagen), Renault, PSA Peugeot-Citroën (PSA), Fiat (Fiat, Jeep, Alfa Romeo), Ford, General Motors (GM, including Chevrolet, Opel and Vauxhall), BMW, Daimler (Mercedes, Smart), Toyota (Toyota and Lexus), and Hyundai. Together, these groups represent more than 90% of the total number of new vehicle registrations between 2000 and 2015 (CCFA, 2000–2015). Due to limited availability of market data and on-road measurements for other brands, we do not account for the health impacts from manufacturers not listed above. Geographically, the study covers the current (as of January 2018) 28 member states of the European Union (EU28), plus Norway and Switzerland. However, due to a lack of data on fleet composition and driving activity, excess emissions released in Croatia, Malta, and Cyprus (less than 0.5% of the total VKT in Europe in 2010 (EC, 2010)), are not included. Health impacts occurring in these countries due to excess emissions released in other countries are included in the analysis. This study focuses on the health impacts of secondary $PM_{2.5}$ and ozone resulting from excess on-road NO_x emissions. We note that primary $PM_{2.5}$ emissions from diesel cars have not raised similar concerns (Spreen et al., 2016).

We use the GEOS-Chem chemistry transport model (Bey et al., 2001) to relate the total excess NO_x emissions to the associated change in $PM_{2.5}$ and ozone concentrations in Europe. We then apply concentration-response functions and use country-specific population data to estimate the number of premature mortalities caused by excess NO_x emissions of diesel cars. We account for uncertainty in the input parameters by modeling their probability distributions and performing a Monte Carlo simulation with 10,000 independent samples. More details on the distributions are provided in the Methods section and in the Supplementary Material. Unless otherwise noted, we report the mean value of the output distribution, along with the 95% confidence interval (95% CI).

2. Methods

2.1. Vehicle fleet, activity, and geographic distribution of the excess NO_x

An inventory of diesel cars in Europe in 2015 is estimated from past fleet data and new registration numbers. Data gathered by the TRACCS projects (Papadimitriou et al., 2013) for the years 2005–2010 is used to initialize our inventory. For subsequent years, we use new registration data from the International Council on Clean Transportation (ICCT, 2016) and market share data from the Comité des constructeurs français d'automobiles (CCFA) for years 2000–2015 to track new vehicles. The market share of Toyota relative to Japanese brands (as reported by the CCFA) and of Hyundai relative to Korean brands (as reported by the CCFA) were assumed constant across the countries considered. Market shares of other brands are country-specific. The vehicle retirement rate is calibrated for each country following the methodology recommended by the TRACCS project (Papadimitriou et al., 2013). It follows the following form

$$\varphi(k) = exp\left[-\left(\frac{k + b}{T}\right)^b\right], \quad \varphi(1) = 1$$

where $\varphi(k)$ is the probability that a vehicle will still be on the road years k after its initial registration. b and T are fitted to country-specific data gathered by the TRACCS project for years 2005–2010. The lifetime function in a given country is assumed to remain unchanged until 2015.

Vehicle-kilometers traveled (VKT) in 2015 are calculated in each country using the Stochastic Transport Emissions Policy (STEP) light-duty vehicle fleet model developed by Bastani et al. (2012). The model is calibrated using country-specific activity data from the TRACCS project (Papadimitriou et al., 2013). Yearly variations of this quantity are taken into account, whereas seasonal variations in driving behavior are not captured.

The total emissions for each country, year, and manufacturer are allocated on a $\sim 25 \times 28\,km^2$ grid covering Europe using a spatially resolved dataset of NO_x emissions from light-duty vehicles (with an original resolution of $0.1^\circ \times 0.1^\circ$), as reported by the European Monitoring and Evaluation Programme (EMEP, 2016).

2.2. Emission factors

This study uses the on-road measurement reports published by the German Federal Motor Transport Authority (KBA) (BMVI, 2016), the Department for Transport (DfT, United Kingdom) (DfT, 2016), the Ministry of the Environment, Energy, and the Sea (MEEM, France) (MEEM, 2016), the German non-governmental organization Deutsche Umwelthilfe (DUH) (DUH, 2016), and the German online newspaper Auto, Motor, und Sport (AMS) (AMS, 2016). All the measurements use a portable emissions measurement system (PEMS). KBA, DfT, and AMS measured cars driven according to the Real Driving Emissions (RDE) testing procedures, as proposed by the EU's Technical Committee Motor Vehicles on May 19, 2015 (EC, 2015). MEEM's results were obtained using the New European Driving Cycle (NEDC) on-road instead of a laboratory setting. The NEDC cycle was in force for laboratory testing before the RDE procedure (EC, 2015) was adopted. DUH measured vehicles on a custom 32 km on-road cycle including urban driving, rural driving (up to 80 km/h), and highway driving (up to 120 km/h). All of these cycles include significant variations in speed and traffic conditions and are assumed to represent faithfully real-world NO_x emissions.

All but one of the manufacturers considered in this study have been tested by at least 2 independent entities. Table 1 below summarizes the number and origin of the samples used. We note that no comparably detailed set of measurements were available for pre-Euro 5 vehicles. We thus assume that pre-Euro 5 vehicles were performing no better than Euro 5 vehicles, and we used the Euro 5 on-road NO_x emission factors distribution for pre-Euro 5 vehicles. Excess NO_x emissions for these

Table 1
Source and number (in parenthesis) of samples used for each manufacturer.

Manufacturer	Euro 5 vehicles	Euro 6 vehicles
PSA	DfT (2), MEEM (10)	DfT (1), DUH (1), KBA (1), MEEM (8)
Renault	MEEM (7)	AMS (3), DfT (1), DUH (2), KBA (3), MEEM (8)
Fiat	KBA (6), MEEM (2)	AMS (1), DUH (4), MEEM (2)
VW	AMS (1), DfT (1), KBA (6), MEEM (8)	AMS (7), DfT (3), DUH (10), KBA (10), MEEM (3)
Ford	DfT (1), KBA (1), MEEM (3)	AMS (1), DfT (2), DUH (4), KBA (2), MEEM (3)
GM	DfT (3), KBA (1)	AMS (2), DfT (2), DUH (4), KBA (2), MEEM (3)
BMW	KBA (1), MEEM (1)	AMS (6), DfT (3), DUH (7), KBA (2), MEEM (2)
Daimler	DfT (1), KBA (2), MEEM (1)	AMS (4), DfT (1), DUH (9), KBA (3), MEEM (3)
Toyota	KBA (1), MEEM (2)	DfT (1), DUH (1), MEEM (2)
Hyundai	DfT (3), KBA (1), MEEM (1)	AMS (1), DfT (1), DUH (4), KBA (1), MEEM (1)

vehicles were calculated against the Euro 4 permissible limit value of 0.25 g/km. The limit values for Euro 5 and 6 vehicles are 0.18 g/km and 0.08 g/km respectively.

In order to better represent each manufacturer's fleet, each sample was weighted by the relative market share of the measured model compared to the market share of the other measured models by the same manufacturer (market shares by model are taken from http://carsalesbase.com/). A gamma distribution was then fitted to the weighted samples for each manufacturer, and the resulting distributions were used to draw samples for each manufacturer's emissions indices in the Monte Carlo simulation. The distributions by manufacturers are presented in the Supplementary Material.

2.3. Air quality modeling

$PM_{2.5}$ and ozone concentrations are calculated using the GEOS-Chem chemical transport model (version 10–01) (Bey et al., 2001; Park et al., 2004; Li et al., 2014; Parrella et al., 2012), driven by GEOS-FP 2013 meteorological data provided by the Global Modeling and Assimilation Office (GMAO) at NASA's Goddard Space Flight Center. The model domain covers the area comprised between 15° W and 40° E and 33° N – 61° N, and the model is run at a resolution of 0.25° in latitude and 0.3125° in longitude (approximately 25 km × 28 km) with 47 vertical layers. This corresponds to 20,355 ground-level grid cells over Europe, covering all European Union member states in addition to Norway and Switzerland. At the northern edge, the domain excludes the northernmost parts of Norway, Sweden, and Finland. However, for each of these three countries, over 90% of the national population is captured. Boundary conditions for the domain were obtained from a global GEOS-Chem run at 4° × 5° resolution, using the same meteorological source. Simulations are run for a 15-month time period, with the first 3 months being used as a model spin-up period. 70 volatile organic species are taken into account. This approach is consistent with numerous studies that have used the GEOS-Chem chemical transport model at similar resolutions to estimate ground-level $PM_{2.5}$ and ozone concentrations (Brauer et al., 2012; Protonotariou et al., 2013).

Anthropogenic emissions are from the European Monitoring and Evaluation Programme (EMEP, 2016) 2012 emissions inventory. A baseline simulation unmodified EMEP NO_x emissions from cars is performed. Four additional runs with 270, 541, 1081, and 2162 gigagrams (Gg) of excess NO_x distributed following the spatial pattern of the EMEP (2016) inventory of road transportation NO_x emissions (EMEP, 2016) were performed (our median estimate for total excess NO_x emissions is 689 Gg). Since our median estimate for excess NO_x emissions in 2015 represents less than 10% of the total anthropogenic NO_x emissions in the inventory (EMEP, 2016), the atmospheric response to excess NO_x emitted from diesel cars is considered to be linear (see Appendix C). The five previously mentioned GEOS-Chem runs are used to verify the validity of this hypothesis. We note that the EMEP inventory already accounts for higher on-road NO_x emissions from diesel cars than standard-testing values (ERMES, 2015) but given the linearity of the atmospheric response for this range of perturbations, the impacts of excess NO_x emissions on $PM_{2.5}$ and ozone concentrations can be calculated by either adding or subtracting emissions to the baseline. One run including two times the median national amount of excess NO_x is performed for each country, in order to estimate each country's individual contribution to the Europe-wide $PM_{2.5}$ and ozone concentrations.

Non-anthropogenic emissions sources are taken from the references summarized in Table 2. Emissions are configured at run-time using the HEMCO module (Keller et al., 2014).

Simulated $PM_{2.5}$ and ozone concentrations over Europe obtained without excess NO_x are validated against air quality monitoring data from the European Environment Agency Air Quality e-Reporting dataset for each country (EEA, 2015). Given the spatial resolution of the GEOS-Chem model, we limit our analysis to background monitoring sites. Available measurements are compared point-to-point with the model prediction (see Appendix B), and we find that the relative departure of model predictions from available measurements is 0.032 (95% CI: −0.626 to 0.91) for $PM_{2.5}$ concentrations and 0.027 (95% CI: −0.013 to 0.45) for ozone concentrations. As such, the model typically overpredicts $PM_{2.5}$ concentrations by 3.2% and ozone concentrations by 2.7%. Following Caiazzo et al. (2013), the reciprocals of the biases are used as multiplicative factors to correct the GEOS-Chem model predictions in the uncertainty calculations.

2.4. Health impacts

Epidemiologically-derived health impact functions are used to estimate premature mortalities attributable to excess NO_x emissions. The integrated exposure-response function (IER) method, which was applied in both the 2010 and 2015 Global Burden of Disease studies (Cohen et al., 2017; Burnett et al., 2014; GBD, 2013; Lim et al., 2012) is used to estimate $PM_{2.5}$-related health impacts. Four health endpoints are taken into account: adult (over 30 years old) ischemic heart disease (IHD), stroke, chronic obstructive pulmonary disease (COPD), and lung cancer. We consider age-specific IERs for 5-year age bands, taken from the 2010 Global Burden of Disease study (GBD, 2013).

Premature mortality due to exacerbation of respiratory and

Table 2
Source of non-anthropogenic emissions in GEOS-Chem v10-01.

Emissions	Source and reference
Biomass burning emissions	GFED4 (http://www.globalfiredata.org/), Giglio et al. (2013)
Dust emissions	Zender et al. (2003)
Terrestrial biogenic emissions	MEGAN v2.1 Guenther et al. (2012)
Air sea exchange fluxes	Lana et al. (2011), Jaeglé et al. (2011)
Emissions of NO_x from soils and fertiliser use	Hudman et al. (2012)
Emissions of NO_x from lightning	Murray et al. (2012)
Volcanic SO_2 emissions	Fisher et al. (2011)
Short-lived bromocarbon emissions	Liang et al. (2010)

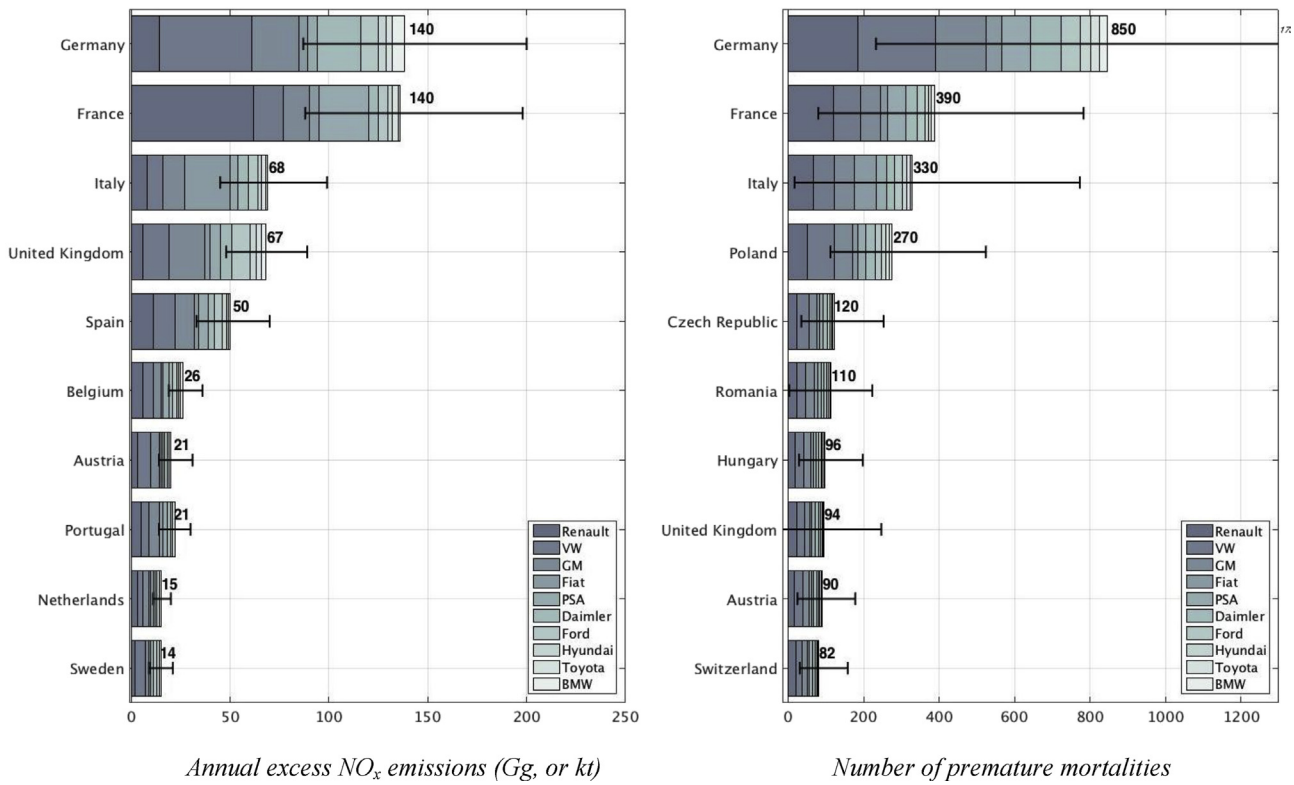

Fig. 1. Excess emissions and their total impacts by country. Mean values are shown in bold, while the solid bars represent the 95% CI. *1a (left)*. Annual excess NO_x emissions, expressed in Gg (or kilotonnes, kt) in the 10 highest-emitting countries. 1b *(right)*. Total number of premature mortalities attributable to excess emissions in the ten most affected countries (highest number of early deaths).

circulatory diseases (ICD-10 codes I00-I99 and J00-J98) as a result of exposure to the annual average of 8-hour maximum ozone concentration is calculated using a two-pollutant models adjusted for $PM_{2.5}$ from Turner et al. (2015). The relationship between exposure and mortality is assumed to be log-linear. Turner et al. find a central relative risk for circulatory diseases of 1.03 (95% CI: 1.01 to 1.05) and a central relative risk of 1.12 (95% CI: 1.08 to 1.16) for respiratory diseases.

Premature mortalities due to the exposure to $PM_{2.5}$ and ozone resulting from excess NO_x emissions from passenger cars are estimated using the well established method of the population-attributable fraction in each grid cell.

$$M_h = P \times B_h \times \frac{RR_h - 1}{RR_h}$$

where M_h is the number of premature mortalities from disease h in that grid cell, P the population count by age group, B_h the vector of baseline incidence rate by age group, and RR_h the relative risk.

The spatial distribution of population in Europe is taken from the LandScan database for 2013 (Bright et al., 2014), and country-specific population count and age breakdown are from the UN World Population Prospects Division (UNDESA, 2015). Following US EPA (2004) recommendations, we apply a 20-year cessation lag to the estimated number of premature mortalities. 30% of the mortalities due to exposure are assumed to occur in the same year, 50% in the following 4 years, and the remaining 20% are assumed to be spread equally over the following 15 years.

The parameters of each of the concentration response functions (CRFs) are treated as independent, uncertain variables. We assume triangular distributions with mode and 95% confidence interval taken from the corresponding epidemiological study.

For each estimated number of premature mortalities, we also report the corresponding number of life-years lost. This quantity is the product of mortalities in each age group with the age group's corresponding standard life expectancy, taking into account the cessation lag described above. Life expectancies are obtained from UN population forecasts (UNDESA, 2015) for the appropriate year. These results are presented in the Supplementary Material.

2.5. Monetization of health impacts

Following common practice in the literature and in government agencies (see OECD, 2012 for a detailed overview), mortality effects due to changes in exposure to $PM_{2.5}$ and ozone because of excess on-road NO_x emissions are valued using two different techniques: the Value of Statistical Life (VSL) and the Value Of a Life Year (VOLY). These methods represent two distinct approaches for attributing a monetary value to air pollution-attributable health impacts and should be considered independently. For VSL, we use a triangular distribution from the OECD (OECD, 2012) (after the appropriate conversion between 2010 USD and 2015 EUR, see Supplementary Material), with a base value of 3.65 million year–2015 EUR, lower bound of 1.82 million EUR, and upper bound of 5.48 million EUR. Health costs in a given year resulting from excess NO_x emissions are calculated by multiplying the estimated number of premature mortalities occurring that year (following the EPA-recommended lag structure (US EPA, 2011)) by the VSL estimate for that year. For premature mortalities occurring in future years, the VSL distribution is adjusted for changes in GDP per capita. VOLY methodology is detailed in the Supplementary Material.

Total health costs are expressed in 2015-EUR using a social rate of time preference of 3% (discount rate), as recommended by the EU ExternE methodology (Bickel and Friedrich, 2005) and by the US EPA (2014).

3. Results and discussion

On the basis of our fleet inventory data, activity model and of the

results of real-world drive test cycles (AMS, 2016; BMVI, 2016; DfT, 2016; DUH, 2016; MEEM, 2016), we estimate the total amount of excess NO_x released by each manufacturer in 2015. The total amount of excess NO_x emitted annually in each country reflects both the country's fleet composition and driving habits, and the differences in the real-world emissions between manufacturers. Results for the ten countries with the highest excess emissions are presented in Fig. 1a. The relative importance of each manufacturer in this total varies by country and reflects manufacturers' market share. Overall, total excess emissions are dominated by the countries with the largest fleets, France and Germany, at the country-level, and by the two major manufacturers in terms of sales, Renault and Volkswagen, at the manufacturer-level.

Excess emissions combined are estimated to cause 2700 premature mortalities (95% confidence interval (CI): 660 to 5500) in 2015. These health impacts are equivalent to about 12% of the number of road fatalities registered in 2015 (EC, 2016). The Europe-wide health costs associated with these excess emissions are estimated to be 9.2 billion EUR (95% CI: 2.2 to 19) using the Value of Statistical Life (VSL) valuation method (OECD, 2012). For each premature mortality, we also compute the associated number of life-years lost, based on country-specific life expectancy data. These results, along with cost estimates using the Valuation of a Lost Year (VOLY) method, are presented in Appendix A.

The breakdown of impacts attributed to each manufacturer in the ten most affected countries is presented on Fig. 1b. In order to control for fleet size and driving habits, we show the estimated number of premature mortalities attributed to each manufacturer per ten billion (10^{10}) VKT driven in Europe and per million vehicles in the fleet in Table 3. We note that the health impacts of excess NO_x emissions vary with location, depending on background atmospheric conditions and population density. In order to account for this geographical effect, we present the number of attributable premature mortalities per gigagram (Gg, or kilotonne, kt) excess NO_x emissions for each manufacturer.

Significant differences arise between manufacturers, with an average of 58 and 52 additional premature mortalities per million vehicles on the road attributed to Renault and GM cars, respectively, which corresponds to an average 33 and 36 premature mortalities per 10 billion vehicle-kilometers traveled (VKT). BMW and PSA vehicles show the lowest mean excess NO_x emissions, estimated to result in an additional 13 and 18 premature mortalities per million cars (or 4.6 and 9.6 premature mortalities per ten billion VKT) on average, respectively. Overall, mean relative health impacts per VKT vary by a factor of 8 among manufacturers, while impacts by car vary by a factor of 4.5 between manufacturers. We note that if the vehicles from all manufacturers in a given Euro standard category emitted at the best-performing manufacturer's level in the category, all other things being equal, approximately 1900 annual premature mortalities would be avoided.

The geographical distribution of cars and corresponding location of emissions does not fully explain the differences between manufacturers, as premature mortalities per gigagram (kilotonne) excess emissions only vary by a factor of 1.2. The remaining differences in health impacts per unit (VKT or cars) are therefore an indication of the variance in excess emission levels between manufacturers. In turn, these difference point towards the technological feasibility to limit on-road excess emissions.

Some countries bear a disproportionate share of the health impacts of the continent-wide excess NO_x emissions: the ratio of the number of premature mortalities to domestic excess NO_x emissions (in gigagram, Gg) ranges from 0.15 for the Netherlands (3 for France, 6 for Germany and Italy) to 78 in for Romania (75 for Lithuania, 23 for Poland, 16 for Switzerland). Results for other countries are presented in Appendix A. Prevailing westerly winds in Europe as well as local geography and background atmospheric conditions partially explain why Eastern countries bear a disproportionate share of the overall health impacts of excess NO_x emissions.

Fig. 2a presents the breakdown of the impacts by country for the ten most affected countries, distinguishing between premature mortalities resulting from domestic excess emissions ("domestic" impacts) and premature mortalities resulting from excess emissions released abroad ("imported" impacts). Fig. 2b provides additional details on the trans-boundary effects of excess NO_x emissions: some countries are net importers of premature mortalities (Poland and Germany, for instance), while others are net exporters (e.g., France and Austria). "Exported" impacts designate impacts due to a country's excess NO_x emissions but occurring abroad. The ratio of domestic to imported impacts varies by two orders of magnitude between countries, driven by prevailing winds and geography as outlined above. Overall, we find that 70 percent of the health impacts stem from trans-boundary emissions, the remainder stemming from domestic emissions. This underlines the need for a coordinated policy response at the EU level.

In order to further understand the nature of transboundary impacts, we control for population effects by normalizing the results in Fig. 2 by population. Fig. 3a shows the domestic impacts per million inhabitants in each country, Fig. 3b the imported impacts per million inhabitants, and Fig. 3c the total health impacts per million inhabitants. As such, Fig. 3 illustrates the imbalance of the geographic distribution of health impacts resulting from excess NO_x emissions and identifies the countries proportionally most affected.

Turning towards the ozone-related impacts, we find excess NO_x emissions released in Northwestern Europe and major urban areas in Portugal, Spain, Italy, and Greece to decrease surface ozone concentration. This is attributed to high background NO_x concentrations relative to volatile organic compounds (VOC) in these regions. This condition, known as NO_x-saturation (Seinfeld and Pandis, 2006), has been established for Europe by previous studies (Beekmann and

Table 3
Total impacts of excess on-road NO_x emissions in Europe attributed to each manufacturer. Mean values are presented, with 95% confidence intervals in parenthesis.

	Total number of attributable premature mortalities	Health costs, based on VSL (billion EUR)	Premature mortalities per 10 billion VKT	Premature mortalities per million cars	Premature mortalities per Gg (or kt) excess NO_x
BMW	60 (−1.8; 160)	0.2 (−0.00645; 0.56)	4.6 (−0.14; 12)	13 (−0.4; 35)	3.8 (0.54; 8.4)
Daimler	220 (7.4; 630)	0.77 (0.026; 2.2)	21 (0.69; 59)	49 (1.6; 140)	4.4 (0.8; 8.5)
Fiat	190 (19; 540)	0.66 (0.063; 1.9)	24 (2.3; 68)	25 (2.5; 71)	4.2 (0.87; 8.5)
Ford	160 (21; 380)	0.54 (0.07; 1.3)	12 (1.5; 28)	20 (2.6; 47)	3.7 (0.56; 7.3)
GM	430 (73; 980)	1.5 (0.24; 3.4)	36 (6; 81)	52 (8.9; 120)	4 (0.77; 7.8)
Hyundai	87 (18; 180)	0.3 (0.06; 0.61)	27 (5.7; 55)	41 (8.7; 83)	4.1 (0.85; 7.9)
PSA	240 (57; 540)	0.84 (0.19; 1.9)	9.6 (2.3; 21)	18 (4.2; 40)	4.3 (1.5; 8.1)
Renault	620 (130; 1500)	2.1 (0.46; 5.1)	33 (7.2; 78)	58 (13; 140)	4.6 (1.8; 8.6)
Toyota	79 (12; 190)	0.27 (0.041; 0.67)	15 (2.3; 37)	20 (3; 48)	4.1 (1; 7.9)
VW	590 (16; 1700)	2 (0.052; 5.8)	15 (0.4; 42)	32 (0.85; 89)	4.4 (0.98; 8.5)
Total	2700[a] (660; 5500)	9.2[a] (2.2; 19)	18[b] (4.4; 36)	33[b] (8; 67)	4[b] (1.1; 8)

[a] Sum of the above quantities.
[b] Average of the above quantities, weighted by manufacturers' shares of VKT, number of vehicles, and excess NO_x, respectively.

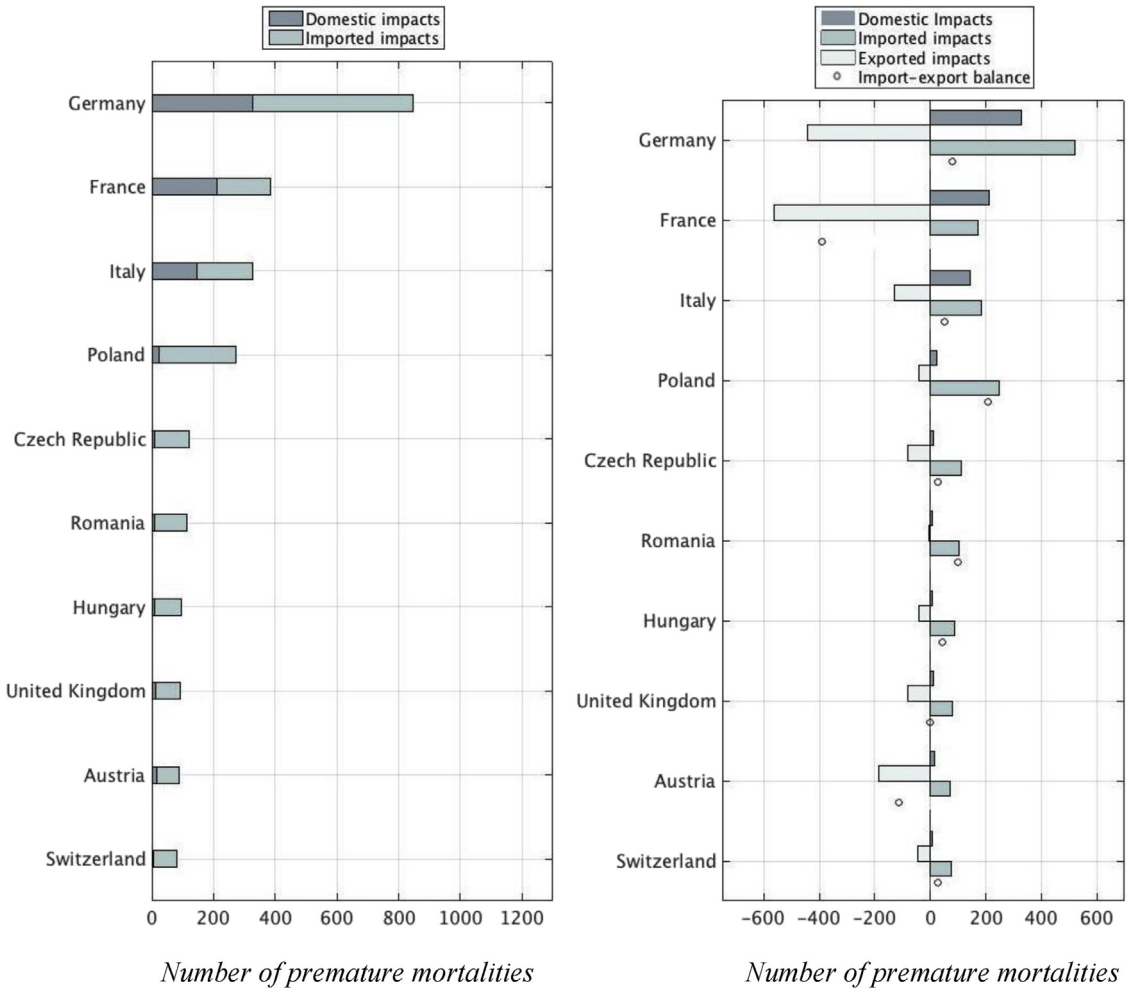

Fig. 2. Health impacts by country for the ten most affected countries. 2*a (left)* Relative share of domestic emissions in average national total premature mortalities. 2*b (right)* Balance of transboundary effects by country (mean number of premature mortalities are shown). Domestic impacts are shown for reference. Exported impacts are counted negatively.

Vautard, 2010; Martin et al., 2004; Colette et al., 2011; Akritidis et al., 2014; Stevenson et al., 2004; Afshar-Mohajer and Henderson, 2017) and causes a reduction in ozone concentrations with increases in NO_x emissions. Having said this, we find that in some places such as the Netherlands, Portugal and Greece, ozone reduction effects dominate the impacts of local $PM_{2.5}$ production. This suggests that $PM_{2.5}$ resulting from domestic excess emissions in these areas is rapidly removed by precipitation or carried away by winds, while ozone reduction effects are more local. Imported impacts however lead the mean estimate for the total number of premature mortalities in these countries to be above zero. This is further discussed in Appendix A.

On the contrary, excess NO_x emissions released in the Mediterranean basin (with the exception of major urban areas) are estimated to increase ozone concentrations. This is in line with previous studies (Beekmann and Vautard, 2010; Martin et al., 2004; Colette et al., 2011; Akritidis et al., 2014), which established these opposite regimes between Northwestern and Mediterranean Europe using independent models. On average and since population is concentrated in

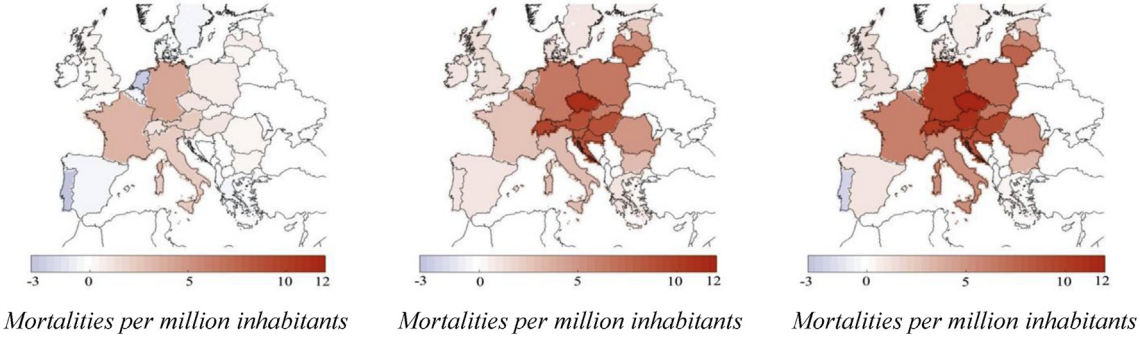

Fig. 3. Average normalized impacts (premature mortalities per million inhabitants) by country. *3a (left)*. Mean number of premature mortalities due to domestic excess emissions (domestic impacts) per million inhabitants. 3b *(middle)*. Mean number of premature mortalities due to emissions released abroad (imported impacts) per million inhabitants. 3b *(right)*. Total mean number of premature mortalities per million inhabitants.

urban areas, which tend to have higher background NO_x concentrations, and in Northwestern Europe, excess NO_x emissions are estimated to reduce ozone-related premature mortalities in Europe. However, the detrimental effects of increased $PM_{2.5}$ concentrations outweigh the beneficial effects of ozone reductions by a ratio of 6 to 1: 3300 premature mortalities (95% CI: 1700 to 6100) are attributed to increased $PM_{2.5}$ concentrations while ozone concentration reductions are associated with a 600 (95% CI: −3 to 2000) decrease in premature mortalities in Europe, yielding to a net impact of 2700 (95% CI: 660 to 5500) premature mortalities.

The health impacts from 2015 emissions are dominated by Euro 5 and pre-Euro 5 vehicles (44% and 47% of the excess NO_x, respectively). In future years, older vehicles will be retired and replaced by newer, Euro 6 vehicles. Among the Euro 6 vehicles currently on sale in Europe, Renault and Fiat show the highest emissions with modeled mean estimates (accounting for the sample vehicles' share in new sales) of 953 and 923 mg NO_x/km, which are 24 and 38% higher, respectively, than the estimated mean emissions of Renault and Fiat vehicles on the road in 2015 (comprised of Euro 6, Euro 5 and pre-Euro 5 vehicles). Ford, PSA, and BMW vehicles also show increased emissions from their Euro 6 vehicles compared to the mean emissions from Ford, PSA, and BMW vehicles currently on the road, even though their Euro 6 on-road NO_x emissions per kilometer remain significantly lower than those of their Euro 6 counterparts from Renault and Fiat (50% lower for Ford and PSA on average, and 70% lower for BMW). For other manufacturers, mean Euro 6 vehicle emissions are significantly lower than those of their vehicles currently on the road. For example, modeled mean on-road NO_x emissions from Volkswagen and Toyota Euro 6 cars are 191 mg/km and 257 mg/km, respectively, which are 29 and 23% lower than the emissions of their current vehicles currently on the road.

Considering only the net change in mortality across the region, our mean estimate of mortality attributable to excess NO_x emissions light-duty diesel vehicles is approximately 45% lower than that from an earlier study by Jonson et al. (2017). This can be explained by several methodological differences. First, the emissions inventory developed for this work accounts for manufacturer-specific excess emissions and country-specific fleet mixes and fleet usage. This yields smaller VKT and NO_x estimates, most notably in Eastern European countries. Second, we apply cause-specific concentration response functions which typically yield lower mortality estimates than the all-cause concentration response functions used by Jonson et al. Finally, this study assumes that NO_x emissions factors from pre-Euro 5 vehicles are well represented by on-road measurements for Euro 4 vehicles, while Jonson et al. use emission standard-specific emissions factors, which are up to 30% higher for earlier standards.

4. Sensitivity analysis and limitations

To determine the sensitivity of our outcomes to our specific choice of concentration response function, we repeated our analysis using some alternative CRFs based on different epidemiological studies. First, we repeated our analysis of the impacts of $PM_{2.5}$, using a log-linear CRF adapted from a meta-analysis of epidemiological studies (Hoek et al., 2013) which relates exposure to $PM_{2.5}$ to an increased risk of cardiovascular premature mortality. The central relative risk for cardiovascular disease mortality is 1.11 (95% CI: 1.05 to 1.16) per 10 μg m^{-3} increase in $PM_{2.5}$ exposure. Using this CRF, $PM_{2.5}$-attributable mortality decreases from 3300 (95% CI: 1700 to 6100) to 3200 (95% CI: 1000 to 8300) on average. This corresponds to a 3% lower mean estimate than reported earlier using the Burnett et al. (2014) integrated exposure response function.

For ozone, we repeated our analysis using the results of a study by Jerrett et al. (2009). They relate 1-hour daily maximum (MDA1) ozone exposure during the local ozone season (usually summer) to premature mortality due to exacerbation of both asthma and chronic obstructive pulmonary disease (COPD). The central relative risk for these outcomes is 1.04 (95% CI: 1.01 to 1.067) per a 10 ppb increase in ozone-season MDA1 ozone concentration. Applying a log-linear CRF with the Jerrett et al. (2009) relative risk in place of the Turner et al. (2015) CRF described earlier, total ozone-attributable mortality goes from −600 (95% CI: −2000 to 2.8) to −18 (95% CI: −78 to 11). Total premature mortality in this case increases from 2700 (95% CI: 660 to 5500) to 3300 (95% CI: 1700 to 6100). The averaging period (ozone season in the case of Jerrett et al. (2009), full year in the case of Turner et al. (2015)) explains this difference, as the exposure to ozone season 1-hour maximum ozone concentrations is found to be 30% higher than exposure to the annual average of the 8-hour maximum ozone concentrations, and approximately 70% less sensitive to NO_x emissions in a NO_x-saturated environment.

Increased exposure to NO_2 has also been correlated with increased all-cause premature mortality, based on studies of urban areas (Hoek et al., 2013). The WHO HRAPIE project (WHO, 2013b) recommends applying to adult populations (over 30 years old) a linear concentration-response function associating increased NO_2 exposure to an increase in all-cause mortality, corresponding to a relative risk of 1.055 (95% CI: 1.031 to 1.08) per 10 μg m^{-3} annual average NO_2 in excess of 20 μg m^{-3}. Using this method, we estimate that an additional 5700 (95% CI: 3000 to 9400) premature mortalities result from excess diesel NO_x emissions. However, significant uncertainty exists with regard to the extent that NO_2 is directly responsible for the increased mortality reported in the literature, as opposed to other by-products of combustion such as $PM_{2.5}$ or ozone (WHO, 2013a). As such we do not include premature mortality due to NO_2 exposure in our estimates of aggregate impact.

We note that the resolution of the air quality model used in this study does not allow to capture urban-scale impacts of direct exposure to NO_2, so that our estimates should be considered lower-end estimates of the actual health impacts of excess NO_x. This is because Denby et al. (2011) find that regional chemical transport models, when compared to results obtained at higher resolution, typically (i) underestimate NO_2 health impacts by 44±4% (ii) underestimate PM_{10} by 15±4% and (iii) overestimate ozone by approximately 13%. Furthermore, Thompson et al. (2014) find that $PM_{2.5}$ impacts vary by ±10% when increasing resolution from 36 km to 4 km.

In addition, the non-linear shape of the $PM_{2.5}$ exposure-response function (Burnett et al., 2014) might lead us to underestimate health impacts from $PM_{2.5}$, as our baseline simulation already includes some excess NO_x emissions, as discussed in Section 2.3. This in turn leads us to overestimate background $PM_{2.5}$ concentrations on top of which health impacts are computed. We test the magnitude of this effect by repeating our analysis with 10% lower background $PM_{2.5}$ concentrations. We find that the calculated health impacts with the modified background match those calculated with our baseline background with an error of 7%. Since our median estimate for excess on-road NO_x emissions results in increases of $PM_{2.5}$ concentrations that are smaller than 6% across the whole grid, we do not expect this non-linearity to significantly affect our results.

Differences in activity across different car segments are not taken into account. Our VKT estimate for 2015 is assumed to depend only on the age of a given vehicle. Nonetheless, we note that the TRACCS project (Papadimitriou et al., 2013) gathered data for four vehicle segments (small, lower-medium, upper-medium, executive) and found no significant difference in activity (less than 5% for the average vehicle). We also assume that the population distribution remains constant over a 20-year time frame. Our assumption that on-road NO_x emissions from pre-Euro 5 vehicles follow the same distribution as emissions from Euro 5 vehicles may also lead us to underestimate the total amount of excess NO_x emissions (Hoek et al., 2013; Jerrett et al., 2009). In addition, we do not account for the fact that NO_x emissions indices may increase with vehicle age. This is consistent with the fact that this study focuses on the health impacts of NO_x emissions in excess of the Euro standards, which are applicable only to new cars. The health

impacts of additional NO_x emissions due to vehicles aging are therefore outside the scope of this study. We however note that Euro 5 and Euro 6 vehicles are less than 6 years old in 2015, and Chen and Borken-Kleefeld (2016) found no significant increase in NO_x emissions index with age for Euro 4 diesel cars. For pre-Euro 4 vehicles, they find an increase of the NO_x emissions index of about 40% throughout the vehicle's lifetime, but pre-Euro 4 vehicles represent less than 25% of vehicles on the road in 2015.

Differential toxicity between the constituents of $PM_{2.5}$ is also not accounted for. The chemical composition of fine particulate matter is thought to influence its toxicity (Hoek et al., 2013; WHO, 2013a). Given that NO_x emissions affect ammonium nitrate more strongly than other $PM_{2.5}$ species, any differential toxicity between this compound and others is not captured by the CRFs. In addition, this analysis focuses on inorganic species and primary organic species and does not include a complete mechanism for secondary organic aerosol (SOA) formation. We however note that Afshar-Mohajer and Henderson (2017) find that a 10_x increase in aircraft NO_x emissions over the course of a month caused the total secondary organic aerosol burden to vary by a maximum of 1.6%, while the ozone burden was found to vary by up to 8%. Understanding the effect of excess NO_x on SOA is an important area for future research. Finally, our results do not include morbidity impacts, although these are expected to be small relative to mortality impacts (US EPA, 2011).

5. Conclusion

This study establishes that excess NO_x emissions from diesel passenger vehicles constitute a Europe-wide public health issue which cannot be fully addressed at the national level as 70% of all impacts are found to be trans-boundary. These results, therefore, suggest that any policy response will have to be implemented at the EU level. In addition, we find that total health impacts by manufacturers vary by a factor of 4.5 on a per-vehicle basis and 8 on a per-kilometer driven basis, with the lowest emitting manufacturer on a per-km basis approaching emission standard values. This shows that it is currently technologically feasible to reduce on-road emissions to the emission standard values. The geographic location of excess NO_x emissions within Europe only explains a small part of these discrepancies, as impacts per unit NO_x emitted vary by only a factor of 1.2 among manufacturers, while the majority of differences in health impacts is driven by differences in tailpipe emissions between manufacturers.

Finally, even the newest Euro 6 cars from several major manufacturers show higher emissions under real driving conditions than those set by the Euro 6 standard. This implies that, in the absence of additional regulatory or technological measures, excess emissions and associated negative health impacts will continue even as Euro 6 cars reach a higher penetration in the vehicle fleet.

Data availability

All datasets used in this study are publicly available, with the exception of Landscan population dataset. Data and scripts produced in this study are either in the paper or available upon request from the corresponding author.

Declarations of interest

None.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.06.047.

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