

# Air quality and related health impact in the UNECE region: source attribution and scenario analysis

Claudio A. Belis<sup>1</sup>, Rita Van Dingenen<sup>1</sup>

<sup>1</sup> European Commission, Joint Research Centre, Via Fermi 2749, 21027 Ispra, Italy

5 *Correspondence to:* Claudio A. Belis (claudio.belis@ec.europa.eu)

**Abstract.** The TM5-FASST tool was used to study the influence of abatement policies within and outside the UNECEUnited Nations Economic Commission for Europe (UNECE) region on the exposure to O<sub>3</sub> and PM<sub>2.5</sub> and associated mortality in the UNECE countries. To that end, the impacts of pollutants deriving from different geographical areas and activity sectors were evaluated using ECLIPSE V6h air pollutant and greenhouse gases emission reduction scenarios. The mortalities were attributed to O<sub>3</sub> and PM<sub>2.5</sub> following the Global Burden of Disease approach and allocated to geographic areas (UNECE and non-UNECE) and activity sectors, including natural sources. In addition, a combination of runs designed for the purpose led to allocating exposure to O<sub>3</sub> and related mortality to two families of precursors: NO<sub>x</sub>-VOC and CH<sub>4</sub>. In this study the baseline scenario (CLE), ~~that~~which assumes that all air quality and greenhouse gas abatement measures adopted by 2018 are fully implemented, is compared with ~~more~~ ambitious scenarios (maximum feasible reduction, ~~(MFR))~~). The ~~conclusion~~findings from this comparison ~~is~~indicate that O<sub>3</sub> exposure within the UNECE area is more sensitive to measures outside the UNECE region, than PM<sub>2.5</sub> exposure, even though the latter leads to higher mortality than the former. In the “current legislation scenario” (CLE), the mortality associated with O<sub>3</sub> exposure in the UNECE region grows steadily from 2020 to 2050. The upward trend is mainly associated with the growing impact of CH<sub>4</sub> emissions from areas outside UNECE. Also, the mortality related to NO<sub>x</sub>-VOC emissions outside UNECE increases in the same period. By comparison, a measurable decrease (13%) is observed in the mortality attributable to NO<sub>x</sub>-VOC emissions ~~from~~within UNECE. In the same time window, the mortality associated with PM<sub>2.5</sub> exposure in the UNECE region ~~at first~~ decreases between 2020 and 2040 and then rises until 2050. The PM<sub>2.5</sub>-~~related~~ mortality in UNECE is mainly due to anthropogenic emissions within this region followed by natural sources (sea salt and dust) mainly located outside the UNECE region. Between 2020 and 2050, the impact of some UNECE anthropogenic sources on PM<sub>2.5</sub>-related mortality decreases progressively, in particular road transport, energy production and domestic combustion while others, namely agriculture and industry, show an upward trend. Finally, the analysis of MFR scenarios confirms that abatement measures in line with UN SDGsSustainable Development Goals (SDGs) and the Paris Agreement can lead to significant co-benefits between air quality and climate policies.

## 1. 1 Introduction

30 In 2019, 6.67 million deaths globally (equivalent to 12% of the total deaths) were attributed to air pollution exposure, mainly due to fine particles and ozone (HEI, 2020). Air pollution is the main environmental risk of premature death worldwide.

However, the gap between low- and medium-income countries (LMIC) and high income countries (HIC) has widened since the beginning of this century due to the increasing trend of PM<sub>2.5</sub>- related mortality in the former (Burnett and Cohen, 2020).

35 The Convention on Long-Range Transport of Air Pollution (also known as “the Air Convention”<sup>3</sup>) of the United Nations Economic Commission for Europe (UNECE) was adopted in 1979 and at present has ~~51 parties~~56 member States<sup>1</sup>, including the EU since 1982. It has eight protocols, four of which are active. The Gothenburg Protocol to abate acidification, eutrophication and ground-level ozone is under review and an evaluation is in progress to assess the adequacy of its obligations and provisions. One of the aspects under evaluation is the future trend for improvements in air quality, human health and ecosystems impacts linked to methane (CH<sub>4</sub>) emissions. Ground-level ozone (O<sub>3</sub>) concentrations in most of the UNECE region  
40 countries are also influenced by other factors in addition to the regional ozone precursors: e.g. climatic parameters, hemispheric transport and global CH<sub>4</sub> emissions (Butler et al., 2020). Global background levels of O<sub>3</sub>, PM<sub>2.5</sub> and their precursors, including CH<sub>4</sub> emissions, contribute significantly to air pollution within the UNECE region, with impacts on public health, ecosystems and biodiversity (Jonson et al., 2018; Lefohn et al., 2018). Projected trends in anthropogenic CH<sub>4</sub> emissions span a very wide range, depending on assumptions made about economic development and the use of emission control technology (Revell et al., 2015; Turnock et al., 2018).

The Air Convention protocols have contributed to reducing air pollution in UNECE countries. However, it is becoming more and more relevant to evaluate which pollutant levels are most affected/controlled by long-range transport of emissions outside the UNECE area, and to which extent new air quality guidelines can be achieved through emission reductions within UNECE only. ~~The aim of this~~This study aims to identify-investigate to what extent the abatement policies within the UNECE region  
50 and in the rest of the world (ROW) influence the exposure to O<sub>3</sub> and PM<sub>2.5</sub> and associated mortality in the UNECE countries. To that end, the impacts of pollutants deriving from different geographical areas and activity sources that contribute to air quality related mortality in the UNECE region are analysed under different air pollutants and greenhouse gas (GHG) emissions’ abatement scenarios. The emphasis is on quantifying the achievable benefits by analysing the gap between scenarios with high level of ambition and the baseline.~~different levels of ambition and the baseline.~~ In particular, one of the scenarios (MFR BASE)  
55 is mainly driven by technological development connected to air pollutant emissions combined with a basic set of climate-oriented policies (national determined contributions) while the other scenario (MFR-SDS) is an archetype of the potentially achievable reductions by implementing the UN sustainable development goals related to energy combined with ambitious climate-oriented policies.

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<sup>1</sup> Albania, Armenia, Austria, Azerbaijan, Belarus, Belgium, Bosnia and Herzegovina, Bulgaria, Canada, Croatia, Cyprus, Czech Republic, Denmark, Estonia, European Union, Finland, France, Georgia, Germany, Greece, Hungary, Iceland, Ireland, Israel, Italy, Kazakhstan, Kyrgyzstan, Latvia, Liechtenstein, Lithuania, Luxembourg, Malta, Monaco, Montenegro, Netherlands, North Macedonia, Norway, Poland, Portugal, Republic of Moldova, Romania, Russian Federation, Serbia, Slovakia, Slovenia, Spain, Sweden, Switzerland, Tajikistan, Türkiye, Turkmenistan, Ukraine, United Kingdom, United States of America and Uzbekistan.

## 2. Methods

### 60 2.1. Exposure and health impact assessment

The TM5-FAst Scenario Screening Tool (TM5-FASST) is a ~~simplified~~reduced-form air quality model based on linearised emission-concentration response sensitivities derived from (also called source-receptor coefficients). The emission-concentration responses to regional emission changes were pre-computed at 1°x1° grid resolution with the full ~~TM5 chemistry-transport (CTM) model TM5~~ (Krol et al., 2005), ~~to calculate~~ for 56 continental source regions, as well as for international shipping and aviation, for a 20% emission reduction in each of the relevant pollutant precursors (SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, BC, OC, NMVOC) and for each individual source region. The resulting deviation relative to the unperturbed case in ground level pollutant concentrations is assumed to scale linearly with the emission deviation relative to the unperturbed case. More details are given in the S.I.

The TM5-FASST model bypasses CPU-expensive explicit chemical and physical process computations, at the cost of accuracy, as documented by Van Dingenen et al. (2018). It worth mentioning that the model addresses impacts of air pollution globally. ~~The anthropogenic emissions under constant meteorological conditions (year 2001), and therefore does not consider~~ feedbacks of climate on photolysis rates, precursor residence times and deposition rates etc. This also implies that natural emissions of volatile organic components (including natural CH<sub>4</sub>), NO<sub>x</sub>, as well as natural PM<sub>2.5</sub>, are treated as fixed, constant contributions. Still, without claiming to be quantitatively equivalent to a full CTM, the model captures major features and implications of emission trends and has proven to be a useful screening tool in science-policy analysis (Van Dingenen et al., 2018).

A great advantage of a source-receptor model is that it keeps track of the contribution of each of the 56+2 source regions, as well as each individual precursor, to each receptor grid cell of the global domain, under the first-order assumption that all contributions can be added up linearly. This makes the model particularly useful for source attribution studies, which can be applied with a large flexibility on the definition of the receptor regions, the latter being a customisable aggregation of grid cells. In this study we consider as receptor region the UNECE domain, and we explore contributions of pollutant emissions outside and inside the UNECE region. Further detail in the attribution studies is obtained by breaking down the emissions by anthropogenic sector.

Health-relevant exposure metrics considered in the present study are the population-weighted PM<sub>2.5</sub> concentrations (as the sum of sulphate, nitrate, ammonium and primary PM<sub>2.5</sub>) and the seasonal daily maximum 8h ozone average (SDMA8h). We apply a sub-grid correction to account for the spatial correlation between population density and primary PM<sub>2.5</sub> associated with transport and household emissions, leading to a higher estimated exposure than the value based on a uniform PM<sub>2.5</sub> distribution across the 1°x1° grid. This is relevant where strong population gradients occur within a single grid (Van Dingenen et al., 2018). Details of the applied parametrisation are given in the S.I.

90 The mortality associated with ~~these~~exposure to outdoor pollutants is estimated according to the Global Burden of Disease (GBD) approach (Stanaway et al., 2018). ~~An overview of TM5 FASST~~The methodology is available to estimate air quality-

health impacts is given in the ~~supplementary material and~~ S.I. A complete description and validation of the TM5-FASST model is provided in Van Dingenen, et al. (2018) and Belis et al. (2022).

## 2.2. Sources

95 The contributions (or impacts) from anthropogenic sectors and natural emissions to PM<sub>2.5</sub> and O<sub>3</sub> exposure metrics in the UNECE region are estimated by the so called brute-force or emission reduction impact approach (Belis et al., 2020). The impact of the following anthropogenic activity sectors (11) was quantified: agriculture (AGR), agricultural waste burning (AWB), domestic and commercial combustion (DOM), energy production (ENE), industry (IND), use of solvents (SLV), road transport (TRA), gas flaring (FLR), waste management (WST), open biomass burning (BMB) and maritime (~~SHP~~ FireSHP).

100 Historical fire emissions were added from ~~SSP2 CMIP6 (projections) and~~ van Marle et al. (2017) and projections from the harmonized CMIP6 SSP2 scenario (Feng et al., 2019), including large-scale biomass burning and savannah burning and excluding AWB emissions to avoid double counting with the ECLIPSE V6b emissions. The resulting anthropogenic PM<sub>2.5</sub> concentration fields are overlaid with fixed natural PM<sub>2.5</sub> sources dust (DUST) and sea salt (SS), taken as the average of the CAMS reanalysis for years 2000 to 2008 (<https://www.ecmwf.int/en/forecasts/dataset/cams-global-reanalysis>). For O<sub>3</sub>, the

105 abovementioned sectoral attribution was complemented with runs separating the impact of NO<sub>x</sub>-NMVOC (hereon NO<sub>x</sub>-VOC) ~~VOC-NO<sub>x</sub>~~ and CH<sub>4</sub> precursor emissions from: (1) UNECE (continental, anthropogenic), (2) ROW (rest of the world: non-UNECE continental, anthropogenic), ~~sources that cannot be separated between UNECE and ROW like~~ (3) international shipping (~~SHP~~ and non-anthropogenic sources associated with biogenic and hereon maritime), and (4) other sources, according to the scheme described in Figure 1 ~~Figure 1~~.

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SET 1: CLE\_ALL EMISSIONS (Standard run)  
 SET 2: UNECE\_ALL EMISSIONS OFF  
 SET 3: ROW\_ALL EMISSIONS OFF  
 SET 4: SHIP EMISSIONS OFF  
 SET 5: ALL\_CH<sub>4</sub> EMISSIONS OFF  
 SET 6: UNECE\_CH<sub>4</sub> EMISSIONS OFF

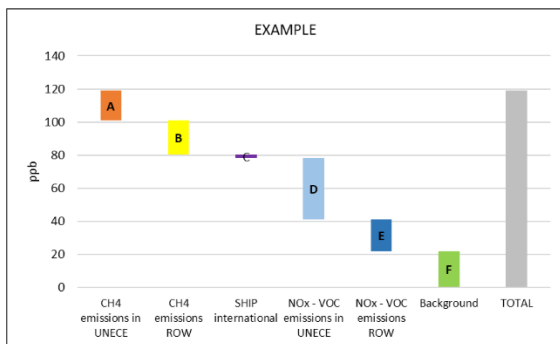


CH <sub>4</sub>	emissions in UNECE	A	SET 1 – SET 6
	emissions outside UNECE (ROW)	B	SET 1 – SET 5 – A
SHIP	international maritime emissions	C	SET 1 – SET 4
NO <sub>x</sub> - VOC	emissions in UNECE	D	SET 1 – SET 2 – A – C
	emissions outside UNECE (ROW)	E	SET 1 – SET 3 – B – C
Other sources	Background , including: lightning and soil NO <sub>x</sub> , biogenic NMVOC and CH <sub>4</sub> , stratospheric O <sub>3</sub> intrusion	F	SET 1 - (A+B+C+D+E)

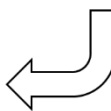
SET 1: CLE\_ALL EMISSIONS (Standard run)  
 SET 2: UNECE\_ALL EMISSIONS OFF  
 SET 3: ROW\_ALL EMISSIONS OFF  
 SET 4: SHIP EMISSIONS OFF  
 SET 5: ALL\_CH<sub>4</sub> EMISSIONS OFF  
 SET 6: UNECE\_CH<sub>4</sub> EMISSIONS OFF



Precursor or Special source	Geographic area	Code	Calculation
CH <sub>4</sub>	emissions in UNECE	A	= SET 1 – SET 6
	emissions outside UNECE (ROW)	B	= SET 1 – SET 5 – A
SHIP	international maritime emissions	C	= SET 1 – SET 4
NO <sub>x</sub> - VOC	emissions in UNECE	D	= SET 1 – SET 2 – A – C
	emissions outside UNECE (ROW)	E	= SET 1 – SET 3 – B – C
Other sources	Background , including: lightning and soil NO <sub>x</sub> , biogenic NMVOC and CH <sub>4</sub> , stratospheric O <sub>3</sub> intrusion	F	= SET 1 - (A+B+C+D+E)



TOTAL OZONE METRIC = A + B + C + D + E + F



**Figure 1. Approach adopted to split O<sub>3</sub> concentrations by emission area (UNECE and non UNECE (ROW)) and by precursor (NO<sub>x</sub>-VOC, VOC, NO<sub>x</sub> and CH<sub>4</sub>).**

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It should be noted that the TM5 FASST model does not include any feedbacks from changing chemical regimes when computations are performed switching off individual precursor emissions. However, since TM5 FASST ozone SRs are estimated with 20% simultaneous NO<sub>x</sub>, VOC emission reductions and only summer exposure is analysed, the chances of chemical regime changes are minimised. This is confirmed by comparisons with the original model TM5 that show an agreement within 5% for 6m DMA1 over the entire perturbation range (Van Dingenen et al., 2018).

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The standard simulations (Set 1) include the emissions in each of the three ECLIPSE V6b scenarios as described in section 02.3. Scenarios (CLE, MFR BASE, MFR-SDS). In addition, a series of perturbations (sets 2 to 6) were computed in which

125 the emissions of specific O<sub>3</sub> precursors (either NO<sub>x</sub>-VOC or CH<sub>4</sub>) were reduced worldwide or in specific areas (either UNECE  
or rest of the world) for each of the abovementioned scenarios. A total of 18 simulations were computed: one for each of the  
130 three scenarios in each of the six sets. Subsequently, sets 1 to 6 were conveniently subtracted as described in Figure 1 (right)  
to split the contributions/impacts of UNECE countries from those of the rest of the world and allocate O<sub>3</sub> to its two families of  
precursors: NO<sub>x</sub>-VOC or CH<sub>4</sub>. International maritime emissions are allocated into a stand-alone category as they are not  
attributed to any geographic area (UNECE nor ROW). The category “other sources” includes emissions not allocated to any  
specific area nor precursor (e.g. lightning and soil NO<sub>x</sub>, biogenic NMVOC and CH<sub>4</sub>, stratospheric O<sub>3</sub> intrusion). In the analysis  
of the results, the apportionment by region and precursor described here was combined with the information about  
anthropogenic sources described at the beginning of this section.

In Appendix A, the PM<sub>2.5</sub> and O<sub>3</sub> source apportionment presented in this study is compared with similar studies in the literature.  
The obtained shares for the PM<sub>2.5</sub> and O<sub>3</sub> exposure metrics are converted to total mortalities according to:

$$135 \quad MORTALITY_{source\ x} = MORTALITY_{total} \times \frac{EXPOSURE\ METRIC_{source\ x}}{EXPOSURE\ METRIC_{total}} \quad (4)$$

Where  $EXPOSURE\ METRIC_{total}$  is the sum of all individual sources ( $x$ ) shares.

### 2.3. Scenarios

140 This study evaluates a set of scenarios (Appendix A, Table A1) derived from the ECLIPSE dataset version 6b (Amman et al.,  
2011; Klimont et al., 2017) developed using the GAINS model (IIASA, 2022; Klimont et al., in preparation). ~~A detailed  
description of all the ECLIPSE scenarios used in this study is provided in Belis et al. (2022).~~

To assess different levels of ambition in the abatement policies from 2020 onwards the CLE is compared with ~~both maximum  
feasible reduction (MFR) scenarios (Appendix A, Table A1). In this study were used the gridded population projections from  
145 Jones and O’Neill (2016) which are in line with the Shared Socioeconomic Pathways (SSP) (Riahi, et al., 2017), two maximum  
feasible reduction (MFR) scenarios: MFR BASE and MFR-SDS (Appendix A, Table A1). For every macro-sector (e.g. energy,  
transport, industry, etc.), each scenario combines a set of cross-cutting measures with others specific for each region of the  
world. The CLE and MFR BASE scenarios are based on the International Energy Agency (IEA) new policy scenario (NPS;  
IEA, 2018) which includes measures that had been announced by 2018 and makes no assumptions about further evolution of  
150 these positions nor aims to achieving any particular outcome. The NPS includes European Union’s 2030 renewable energy  
and energy efficiency targets, the Chinese three-year action plan for cleaner air, the planned revision of the Corporate Average  
Fuel Economy standards in the United States, as well as the announced US Affordable Clean Energy rule. Moreover, it  
considers Japan’s revised basic energy plan and Korea’s 8th National Electricity Plan. The climate policy for both CLE and  
MFR BASE is the same and is specific for every country as it is based on the countries’ national determined contributions~~

155 (NDCs) under the Paris agreement (<https://unfccc.int/process-and-meetings/the-paris-agreement/nationally-determined-contributions-ndcs>). Example of cross-cutting measures in the NPS are: fuel sulphur standards of 10-15 ppm in the road transport sector, global cap of 0.5% on sulphur content in fuel in 2020 in the international shipping sector, and improving fuel efficiency by 2% per year until 2020 in the international aviation sector. The emission reduction in the MFR BASE scenario compared to the CLE are based on the introduction of best available technology with no cost limitations (Table A1).

160 Unlike the previous two, the MFR-SDS scenario is based on the IEA's Sustainable Development Scenario (IEA, 2018) which includes the main energy-related components of the Sustainable Development Goals, agreed by 193 countries in 2015 to keep the increase of global average temperature below 2 °C, achieving universal access to modern energy by 2030 and reducing dramatically the premature deaths due to energy-related air pollution. Examples of cross-cutting assumptions in the SDS are: staggered introduction of CO<sub>2</sub> prices, fossil fuel subsidies phased out by 2025 in net-importing countries and by 2035 in net-

165 exporting countries, and maximum sulphur content of oil products capped at 1% for heavy fuel oil, 0.1% for gasoil and 10 ppm for gasoline and diesel. A full description of the scenarios goes beyond the purposes of the present work. More details are available elsewhere (<https://iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv6.html>; IEA, 2018; Belis et al., 2022).

170 In this study were used the SSP gridded population projections from Jones and O'Neill (2016). The SSP2 projections were associated with CLE and MFR BASE while SSP1 were used with the MFR-SDS scenario.

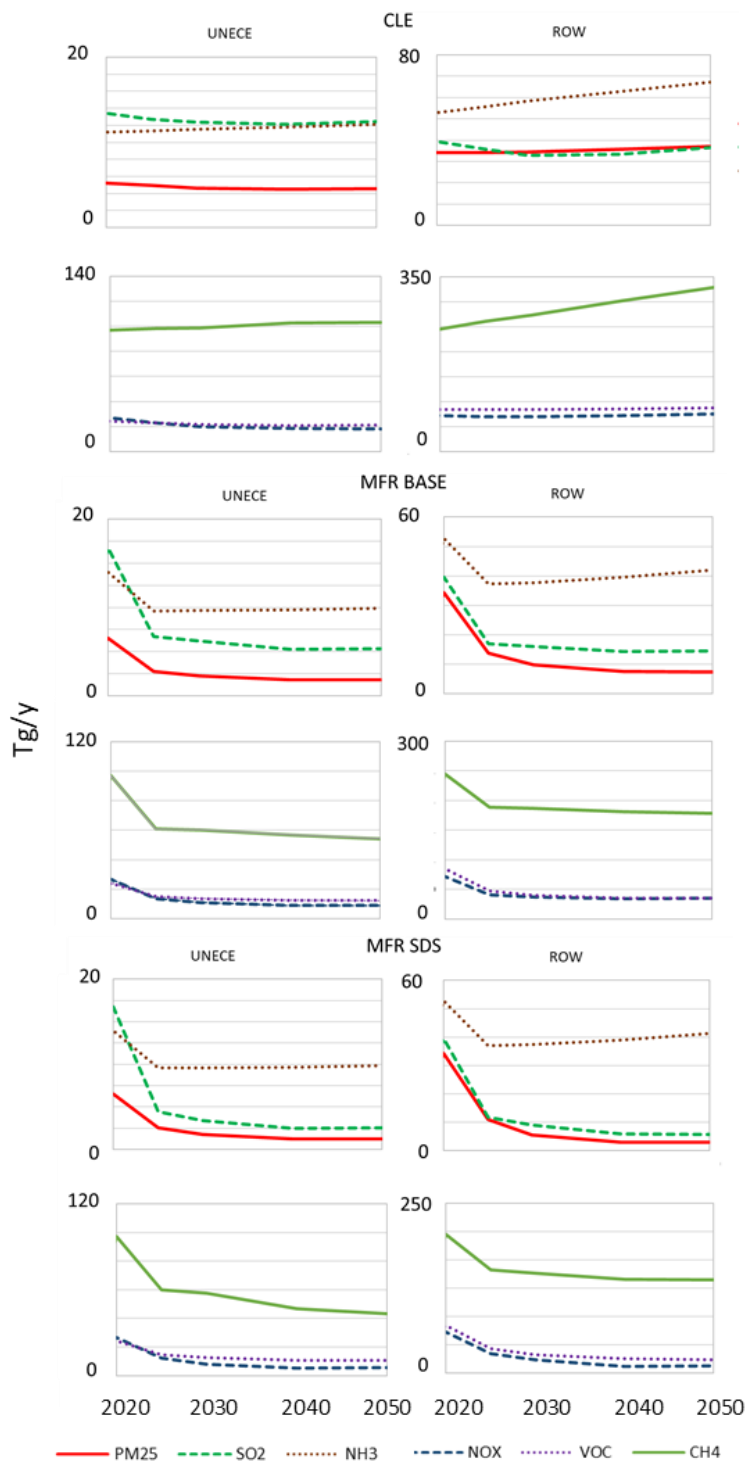
### 3. Results

#### 3.1. Emissions

The UNECE and ROW emission trends between 2020 and 2050 of O<sub>3</sub> and PM<sub>2.5</sub> precursors in all the studied scenarios are shown in ~~Figure 2~~Figure 2. In the **CLE scenario**, UNECE NO<sub>x</sub>, NMVOC and PM<sub>2.5</sub> emissions decrease by 33%, 13% and 13%, respectively, between 2020 and 2050 while in ROW, NH<sub>3</sub> and CH<sub>4</sub> grow by 27% and 34%, respectively.

In both MFR scenarios, UNECE emissions show a downward trend over the whole time window with the exception of NH<sub>3</sub>, which after an initial decrease remains stable. Moreover, in these scenarios NH<sub>3</sub> is the only precursor with a ~~distinct~~distinguishable upward emission trend between 2025 and 2050 in ROW while all the others show a downward trend.

In MFR BASE, UNECE emissions in 2050 are between 69% (PM<sub>2.5</sub>) and 35% (NH<sub>3</sub>) lower than CLE while ROW emissions are between 80% (PM<sub>2.5</sub>) and 37% (NH<sub>3</sub>) lower than CLE. Despite MFR-SDS emissions follow similar trends, the reductions with respect to the CLE are higher, with the exception of NH<sub>3</sub> which is ~~similar~~the same in both MFR scenarios.





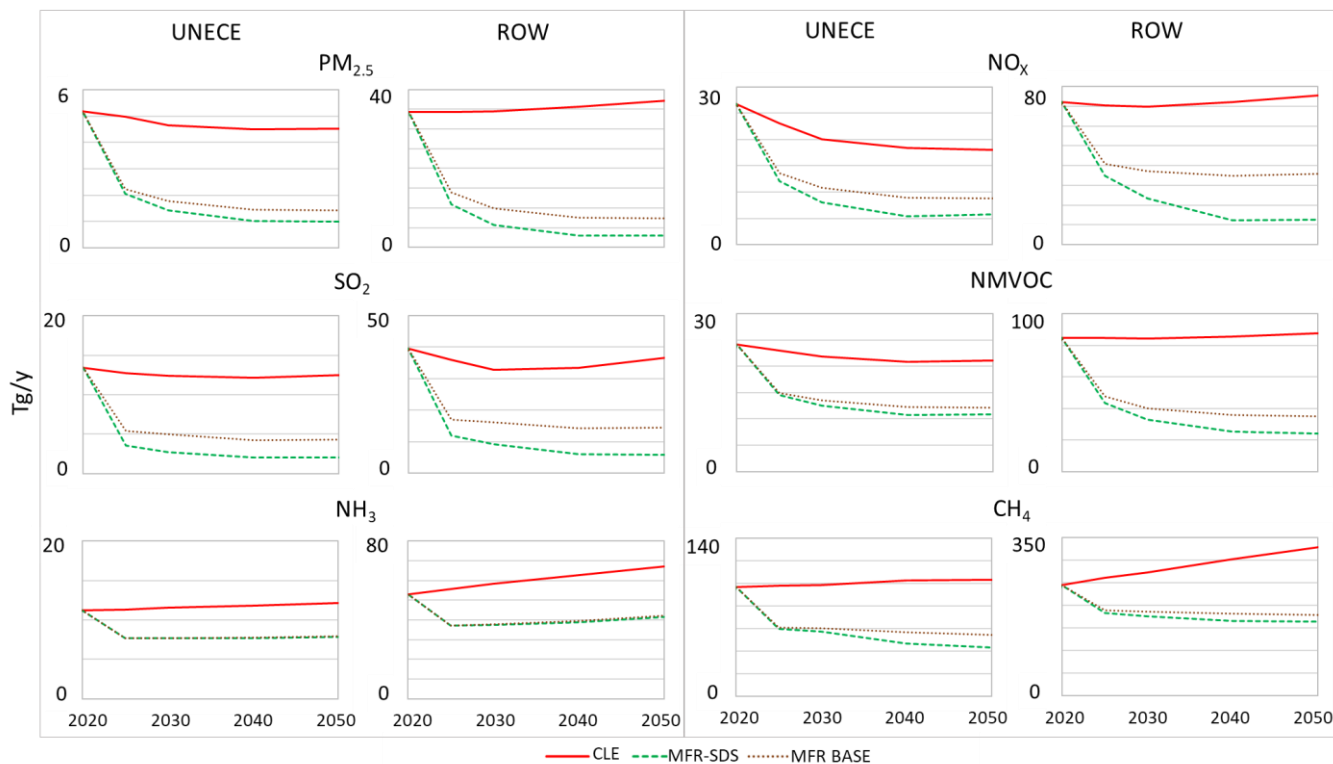
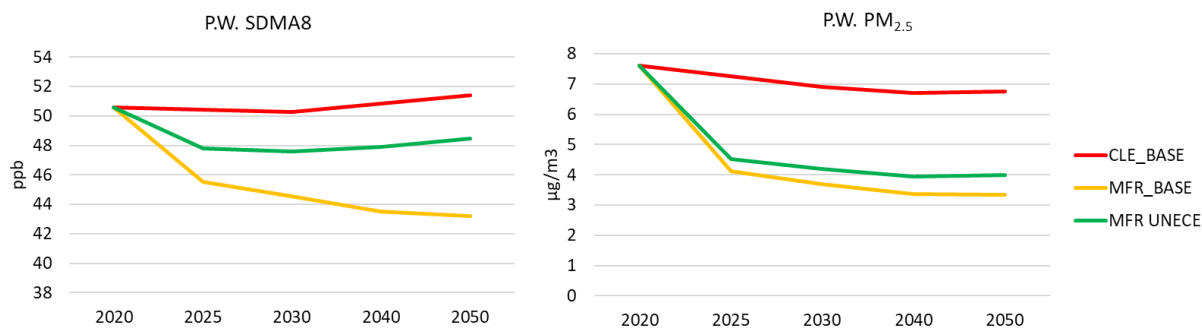


Figure 2. UNECE (left) and ROW (right) emission trends of main O<sub>3</sub> and PM<sub>2.5</sub> precursors in the studied ECLIPSE V6b scenarios

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### 3.2. Influence of ROW on UNECE

To assess the impact of air pollutant and GHG abatement measures outside the UNECE region (Rest of the World; ROW) on UNECE emission abatement policies, a regional source attribution exercise is discussed in this section. The exposure to PM<sub>2.5</sub> (anthropogenic) and O<sub>3</sub> in UNECE countries between 2020 and 2050 in the global baseline scenario (CLE) is compared with the MFR BASE scenario and with a scenario in which the emission reductions foreseen in the MFR BASE are applied only in the UNECE region while CLE emissions ~~apply only to~~ are kept in ROW (~~so-called~~ MFR UNECE scenario) (Figure 3~~Figure 3~~).



195 **Figure 3.** O<sub>3</sub> seasonal mean of 8hr (population weighted SDMA8h, left) and anthropogenic population weighted PM<sub>2.5</sub> (right) annual averages in UNECE region, average of countries, under different scenarios. CLE (current legislation), MFR BASE only applied in UNECE countries (MFR UNECE), MFR BASE in all countries (MFR BASE).

The O<sub>3</sub> exposure in CLE (red line) and MFR UNECE (green line) shows an upward trend from 2025 onwards. The abatement benefit, i.e. the difference between the O<sub>3</sub> exposure in CLE and MFR UNECE, over the considered time window is relatively small (5% to 6%) suggesting that applying emission reductions in UNECE countries only, leads to limited additional abatement in the O<sub>3</sub> exposure in UNECE countries relative to the baseline (CLE). By comparison, the O<sub>3</sub> exposure in MFR BASE (yellow line) follows a downward trend and the abatement benefit (delta CLE-MFR BASE) is twice as much as MFR UNECE (10% to 16%) indicating that implementing MFR worldwide would not only lead to higher abatement of exposure in UNECE but also reverses the trend from increasing to decreasing.

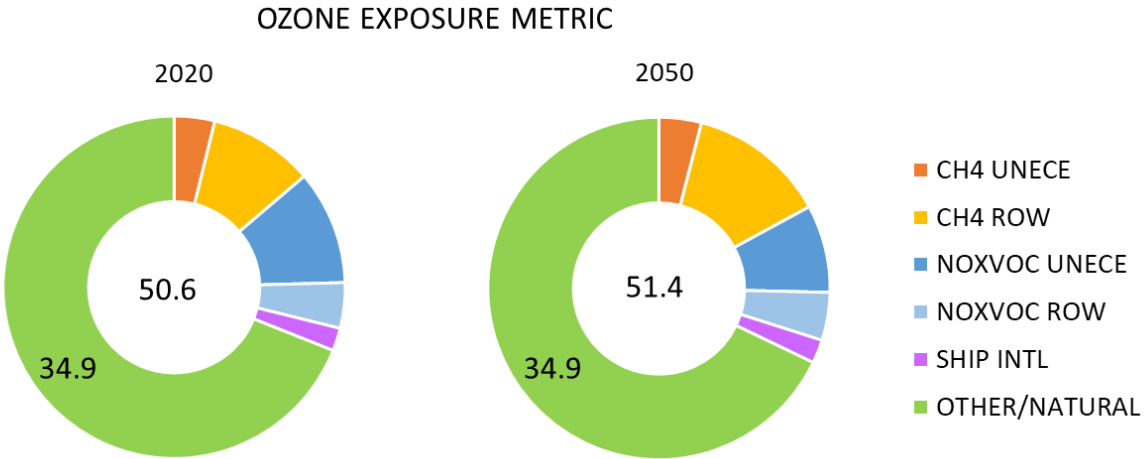
205 Unlike O<sub>3</sub>, PM<sub>2.5</sub> exposure shows a decreasing trend for the three scenarios. The abatement benefit (CLE - MFR UNECE) over the studied period is already high (-38% to -41%) and applying the MFR BASE scenario globally leads to a relatively small marginal benefit (≤ 10% of CLE). In synthesis, for PM<sub>2.5</sub> abatement, UNECE is only slightly affected by ROW measures, while O<sub>3</sub> levels are strongly modulated by measures taken outside the UNECE region. This is obviously related to the longer (compared to PM<sub>2.5</sub>) atmospheric lifetime of O<sub>3</sub> formed from its short-lived precursors NO<sub>x</sub> and NMVOC, and of its other long-lived precursor CH<sub>4</sub> which contributes to global background O<sub>3</sub>. The UNECE countries where the differences in O<sub>3</sub> and PM<sub>2.5</sub> exposure between MFR UNECE and MFR BASE are the highest (in the range 6 to 10 ppb and 1.5 to 2.4 µg/m<sup>3</sup>, respectively) are located at the boundary of the UNECE region and therefore more exposed to long-range pollution from the ROW (Figure S2). Some of these countries are in the Caucasus and central Asia (Armenia, Azerbaijan, Tajikistan, Kyrgyzstan and Turkmenistan) downwind highly polluted regions (e.g. southern Asia, Far East). The highest differences between these scenarios for both pollutants are observed in Israel which is a small country surrounded by an area of non-UNECE countries with high emissions. Some countries in the Atlantic coastal area (Portugal, Spain and Ireland) present high differences in the O<sub>3</sub> exposure between MFR UNECE and MFR BASE likely due to the influence of air masses circulating over the sea and mostly affected by emissions in ROW. A similar situation is observed in Malta which is mostly affected by the high background levels in the Mediterranean Sea.

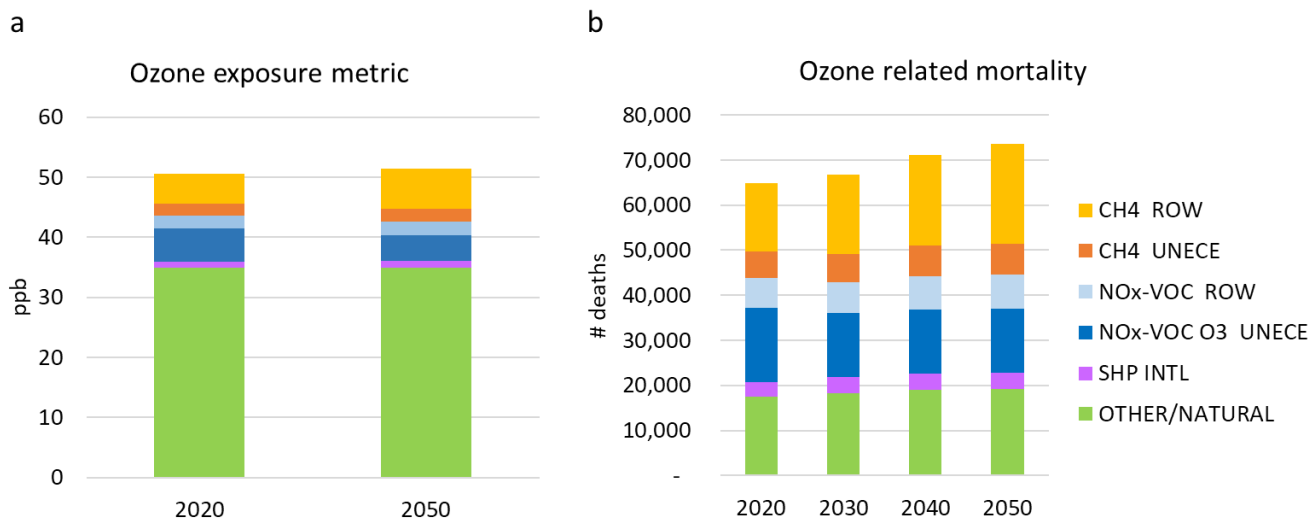
The attribution of O<sub>3</sub> and PM<sub>2.5</sub> levels to precursor emissions in- and outside the UNECE region is further investigated in the following sections.

220 **3.3. Source allocation of ozone exposure and premature mortality in UNECE in the baseline scenario (CLE)**

In this section, the O<sub>3</sub> exposure and related mortality inside within UNECE is broken down by (a) precursor (b) sector and (c) source region (UNECE vs. ROW) considering only the attribution runs of the CLE scenario. The other/natural O<sub>3</sub> background share is estimated from total O<sub>3</sub> minus the sum of all anthropogenic sectors (see section 2.2).

225 The O<sub>3</sub> background (OTHER/NATURAL), including biogenic and other unspecified sources (Figure 4a), is estimated by subtracting the sum of all anthropogenic sectors from total O<sub>3</sub> (see section 2.2) and is the main single contributor to the O<sub>3</sub> exposure. The main single contributor to the O<sub>3</sub> exposure is of non anthropogenic origin (OTHER/NATURAL), including biogenic and other unspecified sources (Figure 4). The impact of this “source” is approximately 35 ppb and remains relatively constant throughout the analysed time window (2020 – 2050). Despite its dominance, this component is not the main focus of the analysis since it is, by definition design, little affected by anthropogenic emissions in the short term. In the 2020 – 2050  
230 time window, the anthropogenic fraction of the O<sub>3</sub> exposure is worth 16 - 19 ppb.





235 **Figure 4. Allocation of the population weighted O<sub>3</sub> (SDMA8h) exposure in UNECE to geographic source areas (UNECE, ROW), precursors and other/natural sources. Units: ppb (a). Mortality (UNECE total) associated with O<sub>3</sub> exposure in UNECE split by natural-background (only the fraction above the exposure threshold) and anthropogenic emissions (b).**

In terms of precursors, in CLE there is a remarkable shift in the relative role of short-lived components (NO<sub>x</sub>, NMVOC) versus CH<sub>4</sub> between 2020 and 2050. The initial dominant role of NO<sub>x</sub> and NMVOC in anthropogenic ozone formation is replaced by CH<sub>4</sub> towards 2050. This is due to the combined decrease of UNECE NO<sub>x</sub> and NMVOC emissions (while ROW emissions remain relatively constant) and the increase of ROW CH<sub>4</sub> emissions (while UNECE emissions remain relatively constant).  
 240 The overall O<sub>3</sub> exposure metric is stable along the observed time window because the decreasing impact of NO<sub>x</sub>-VOC emissions from UNECE over time is largely compensated by the increasing impact of CH<sub>4</sub> emitted in ROW.

The overall share of O<sub>3</sub> exposure allocated to anthropogenic NO<sub>x</sub>-VOC emissions is mainly associated with ~~TRA, IND, SHP~~transport, industry and maritime sources while the CH<sub>4</sub> emissions affecting this pollutant are mainly emitted from ~~AGR, FLR~~agriculture, gas flaring and ~~WST, EN~~waste management, Energy production, another important anthropogenic source,  
 245 presents similar shares of both precursor families (Figure S1S3).

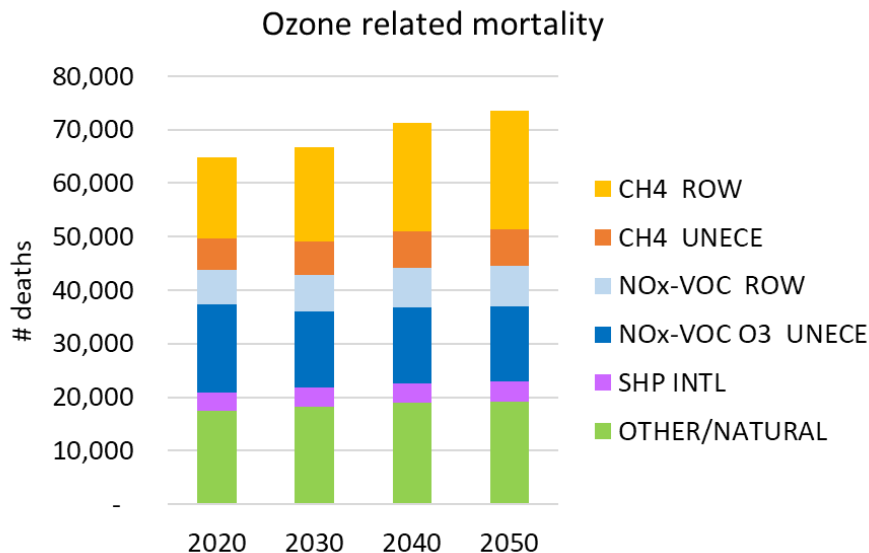


Figure 5. Mortality (UNECE total) associated with O<sub>3</sub> exposure in UNECE split by natural background (only the fraction above the exposure threshold) and anthropogenic emissions.

In Figure 5 In Figure 4b the premature mortality associated with O<sub>3</sub> exposure in the UNECE region estimated in the CLE is shown. The number of premature deaths grows steadily from 65,000 in 2020 to 74,000 in 2050. This upward trend in mortality is mainly associated with an increased impact of anthropogenic CH<sub>4</sub> emissions from ROW (+46 %, +7,000 deaths/year). Also the mortality related to anthropogenic NO<sub>x</sub>-VOC emissions in ROW increases by 17% in the same period (+1,000 deaths/year).

On the contrary, a measurable decrease is observed in the mortality attributable to anthropogenic NO<sub>x</sub>-VOC emissions in UNECE which drops from 16,000 in 2020 to 14,000 in 2050.

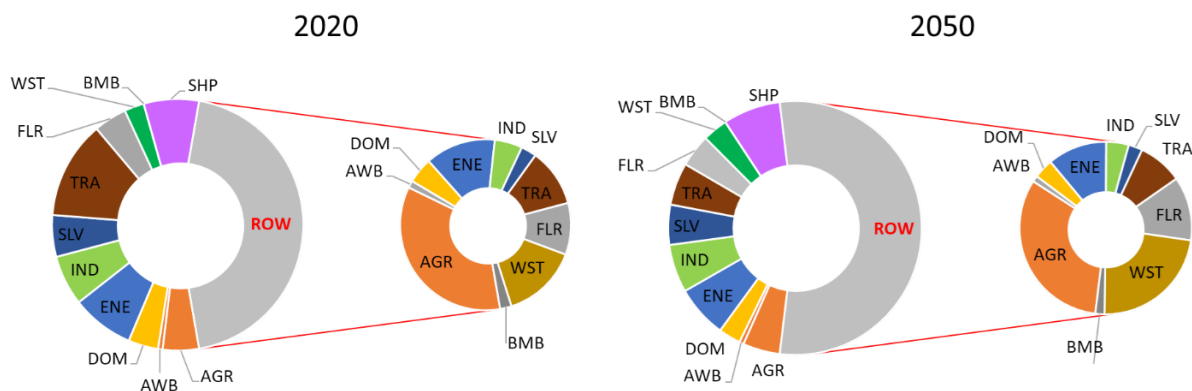


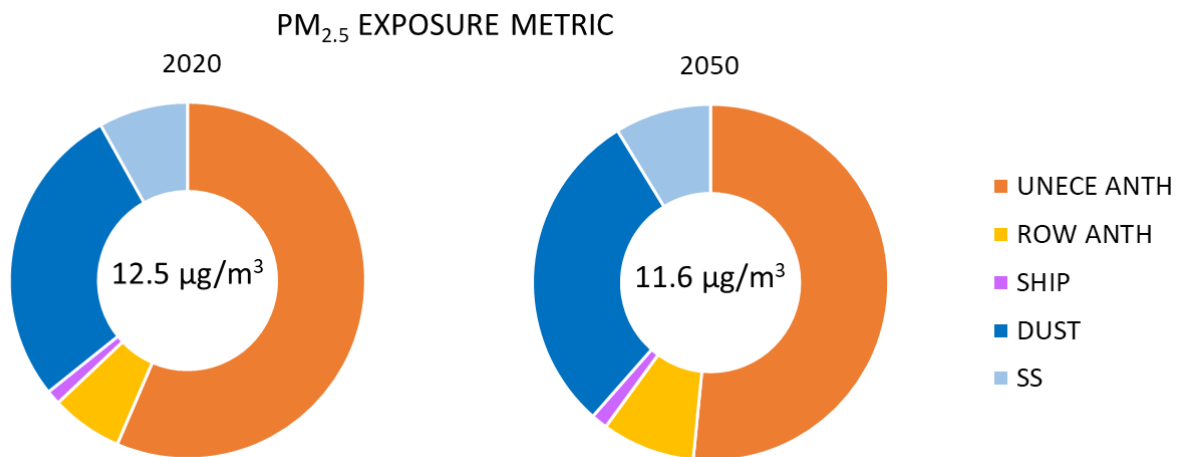
Figure 56. Allocation of O<sub>3</sub> exposure and related mortality (UNECE avg.) to anthropogenic sources under CLE. The overall impacts are represented in the main pie charts while the small pie charts to the left of them show the detail of ROW impacts only. The data are also available in Table S1. AGR: agriculture, AWB: agricultural waste burning, DOM: domestic and commercial combustion, ENE: energy production, IND: industry, SLV: use of solvents, TRA: road transport, FLR: gas flaring, WST: waste management, BMB: open biomass burning and SHP: maritime.

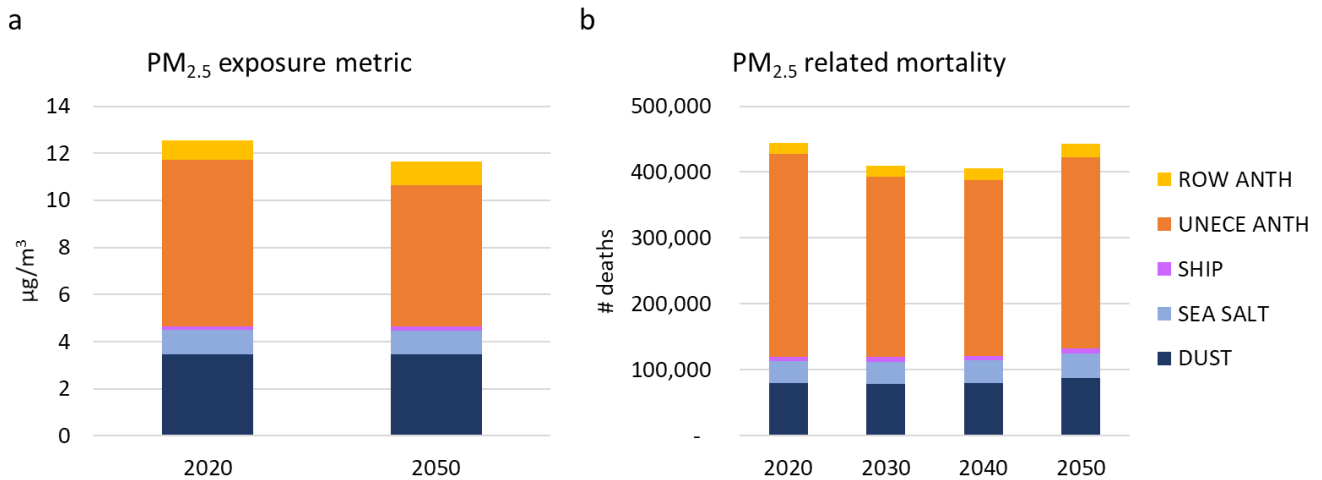
The contributing sectors change their relative importance evolving from a mix dominated by TRA, AGR transport, agriculture and ENE energy production in 2020 to a one dominated by AGR, WST, TRA agriculture, waste management, transport and ENE energy production in 2050 (Figure 5, Figure 6, Table S1). TRA, IND transport, industry and SHP maritime contribute to O<sub>3</sub> exposure only via NO<sub>x</sub>-VOC precursors while AGR, FLR agriculture, gas flaring and WST waste management contribute almost only via emissions of CH<sub>4</sub> emissions (Figure S1, S3).

The CH<sub>4</sub> impact of AGR, FLR, WST agriculture, gas flaring, waste management and ENE energy production emissions from ROW on O<sub>3</sub> exposure in UNECE presents an upward trend between 2020 and 2050 (Figure S1, S3). In the same time window, the NO<sub>x</sub>-VOC contribution from TRA, ENE transport, energy production and DOM domestic emissions from UNECE show a downward trend with the exception of IND industry which increases slightly. Although ENE energy production is the only source which shares of O<sub>3</sub> exposure due to NO<sub>x</sub>-VOC and CH<sub>4</sub> are comparable, the balance between these two components evolves along the studied time window towards an increase in the share of the latter.

### 3.4. Source allocation of PM<sub>2.5</sub> exposure and premature mortality in UNECE in the baseline scenario

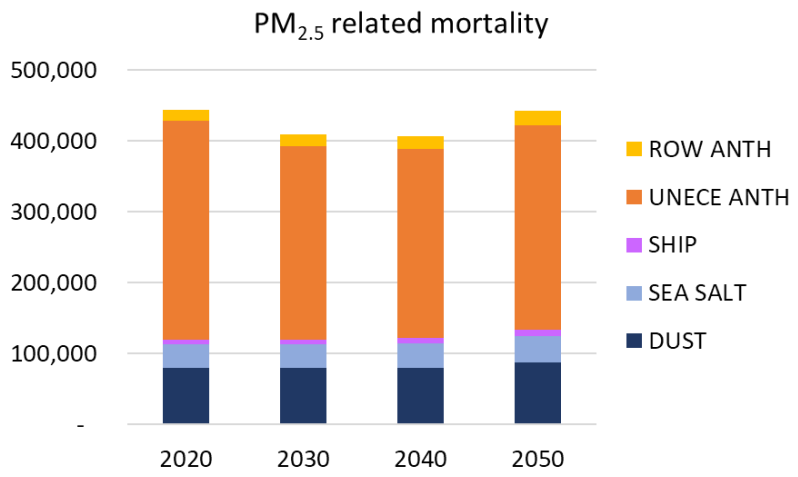
The UNECE anthropogenic emissions are the main responsible for PM<sub>2.5</sub> exposure in UNECE, with a decreasing trend between 2020 and 2050, while those from ROW have a minor role which increases slightly over the observed time window (Figure 6, Figure 7).





280 **Figure 67.** Allocation of the population weighted PM<sub>2.5</sub> exposure in UNECE to geographic source areas (UNECE, ROW) and natural sources under CLE (a). Mortality (UNECE avg.) associated with PM<sub>2.5</sub> exposure attributable to both anthropogenic and natural sources under CLE (b).

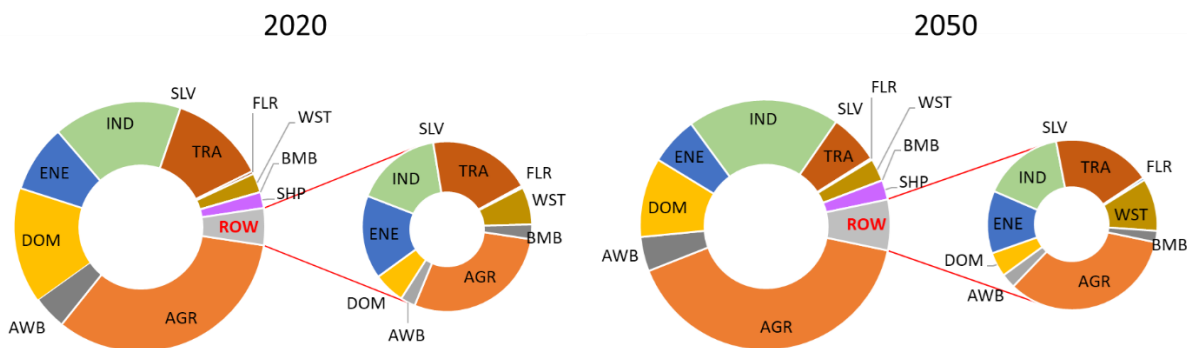
The mortality associated with PM<sub>2.5</sub> exposure in the UNECE region (including both natural and anthropogenic sources) is 444,000 cases in 2020. It shows a downward trend between 2020 and 2030 and a subsequent rise between 2040 and 2050 when it reaches 443,000 units (Figure 8, Figure 6b).



~~Figure 8. Mortality (UNECE avg.) associated with PM<sub>2.5</sub> exposure attributable to both anthropogenic and natural sources under CLE.~~

290 The main anthropogenic contributors within UNECE are: ~~AGR, IND, DOM, ENE and TRA~~ agriculture, industry, domestic, energy production and transport (Figure 7, Figure 9). An overall downward trend in the impact of ~~DOM, ENE and TRA~~ domestic, energy production and transport from UNECE and an increasing role of ~~IND~~ industry and ~~AGR~~ agriculture from

295 this region are observed. The share of SHPmaritime, a contributor which is not geographically allocated in this analysis, is stable from 2020 onwards. In 2050, there is an increase in the PM<sub>2.5</sub> exposure mainly due to a rise in the impact of AGR, TRA, FLRagriculture, transport, gas flaring and WSTwaste management emissions from ROW and AGRagriculture and INDindustry emissions from the UNECE region.

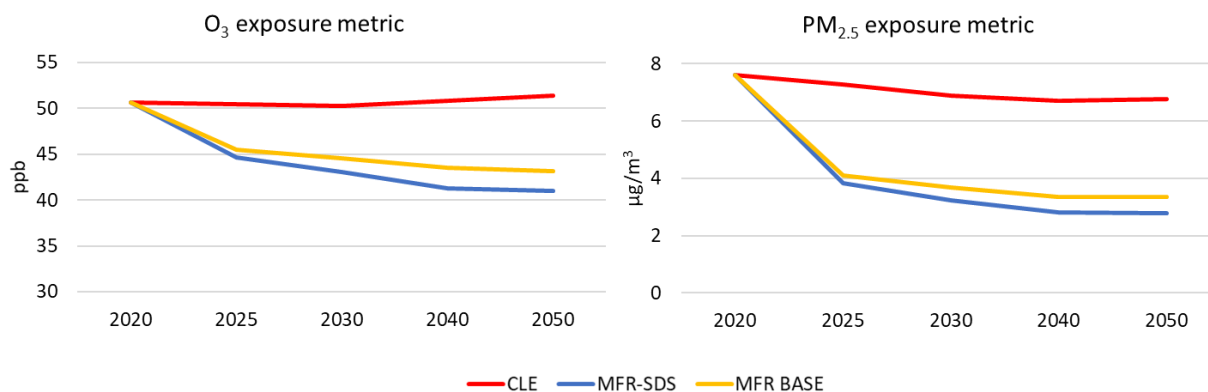


300 **Figure 79.** Allocation of PM<sub>2.5</sub> exposure and related mortality (UNECE avg.) to anthropogenic sources under CLE. The overall impacts are represented in the main pie charts while the small pie charts to the left of them show the detail of ROW impacts only. **The data are also available in Table S2.** AGR: agriculture, AWB: agricultural waste burning, DOM: domestic and commercial combustion, ENE: energy production, IND: industry, SLV: use of solvents, TRA: road transport, FLR: gas flaring, WST: waste management, BMB: open biomass burning and SHP: maritime.

### 3.5. Source allocation of exposure to air pollutants in UNECE in MFR scenarios

305 This section evaluates the trends of the O<sub>3</sub> and PM<sub>2.5</sub> exposure in UNECE between 2020 and 2050 computed with TM5-FASST using the ECLIPSE V-6b MFR BASE and MFR-SDS emission scenarios (Table A1; Figure 8Figure 10). In 2050, the MFR BASE and MFR-SDS O<sub>3</sub> exposure is 16% and 20% lower than CLE, respectively, while the PM<sub>2.5</sub> (anthropogenic) exposure in the abovementioned scenarios is 51 % and 59% below CLE, respectively.





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**Figure 810.** O<sub>3</sub> and anthropogenic PM<sub>2.5</sub> exposure metrics (UNECE avg.) computed with TM5-FASST according to the ECLIPSE V 6b scenarios: CLE, MFR BASE and MFR-SDS.

In the period 2025 – 2050, the main anthropogenic contributor to O<sub>3</sub> exposure and mortality in both MFR scenarios is by far AGRagriculture due to CH<sub>4</sub> emissions in ROW (Figure S2S4).

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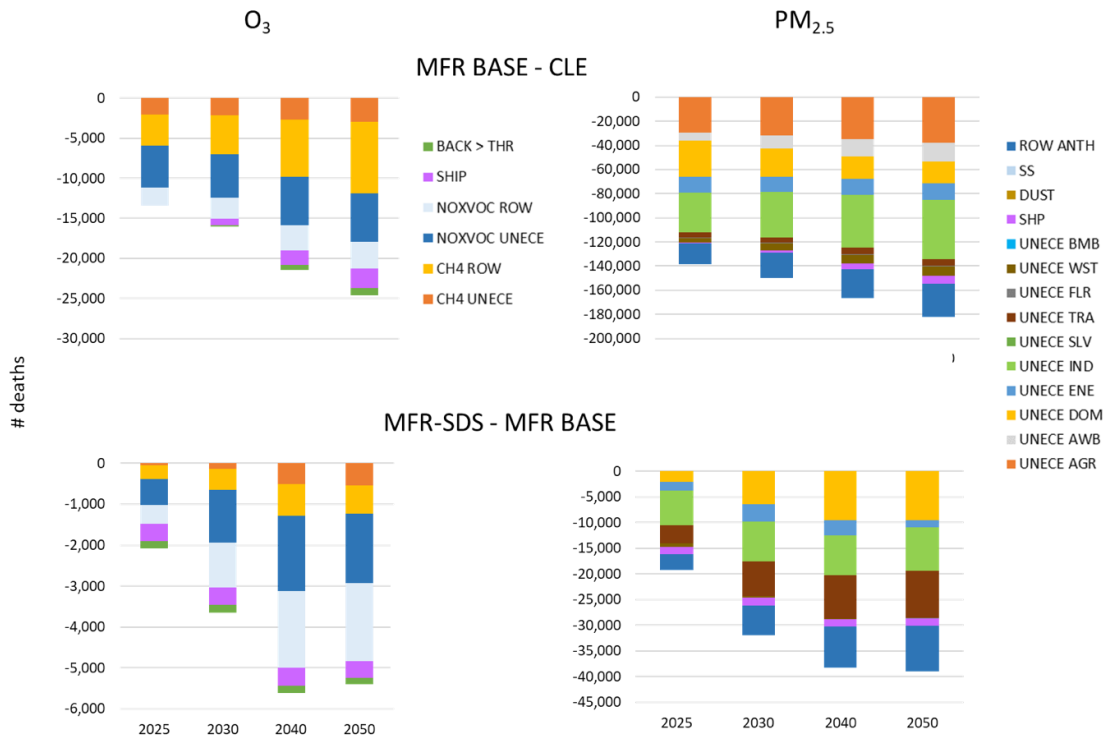
In the MFR BASE scenario, which is mainly based on the implementation of best available technologies (BATs) and Paris Agreement NDCs, the delta mortality in UNECE compared to CLE ranges from -13,000 cases (-21%) in 2025 to -24,000 cases (-34%) in 2050 due to lower O<sub>3</sub> exposure (Figure 9Figure 11 top left). Such improvement is mainly associated with NO<sub>x</sub>-VOC emission reductions in the UNECE region and reductions of CH<sub>4</sub> in ROW, the role of which increases considerably between 2025 and 2050 (Figure 9Figure 11 top left). A more detailed analysis of the MFR BASE reveals that the main UNECE NO<sub>x</sub>-

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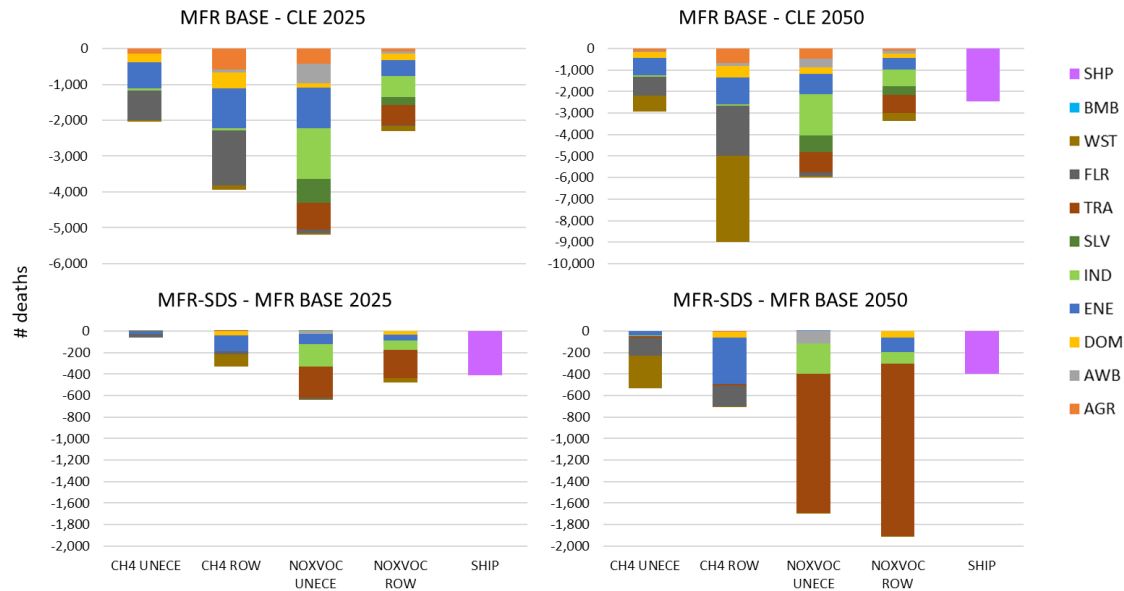
VOC emission reductions in 2050 are associated with ENE, INDenergy production, industry and TRAtransport sectors. By comparison, -while those of CH<sub>4</sub> in ROW are mainly due to abatement of FLRgas flaring and ENEenergy production in 2025 with dramatic abatement increase in the WSTwaste management sector between this year and 2050 (Figure 10Figure 12 top).

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The additional improvement compared to the MFR BASE from the most ambitious MFR-SDS scenario, in line with energy related SDGs and global temperature increase containment, ranges between ca. -2,000 cases (-4%) in 2025 and -5,500 (-11%) cases in 2050 and is mainly due to the reduction of NO<sub>x</sub>-VOC emissions in both UNECE and ROW (Figure 9Figure 11 bottom left). Such abatement of O<sub>3</sub>-related mortality in the MFR SDS scenario is associated with emission reductions in the TRAtransport sector in 2050 in both UNECE and ROW compared to 2020 (Figure 10Figure 12 bottom).



330 **Figure 944.** Delta MFR BASE - CLE and MFR-SDS – MFR BASE of O<sub>3</sub> (left) and PM<sub>2.5</sub> (right) associated mortality (UNECE total) split by precursor and main emission areas. For O<sub>3</sub> we only consider the fraction of ‘OTHER/NATURAL’ exceeding the zero effect threshold of 29.1 ppb.



335 **Figure 1012. Delta MFR BASE – CLE (top) and MFR-SDS – MFR BASE (bottom) of UNECE O<sub>3</sub> associated mortality in 2025 and 2050 split by source sectors.**

In MFR BASE the delta mortality in UNECE due to PM<sub>2.5</sub> exposure compared to the CLE ranges from ca. -137,000 cases (-33%) in 2025 to ca. -187,000 cases (-41%) in 2050 (Figure 9Figure 11 top right). Such improvement is mainly due to abatement of emissions in the AGRagriculture and INDindustry sectors in UNECE. In this region, the abatement of DOMemissions in the domestic sector shows a decreasing importance between 2025 and 2050 while the opposite is true for AWBagricultural waste burning and the sum-of anthropogenic emissions in ROW. By comparison, the MFR-SDS scenario leads to an additional reduction in mortality compared to the MFR BASE of ca. -19,000 cases (-7%) in 2025 that reaches ca. -40,000 cases (-15%) in 2050 (Figure 9Figure 11 bottom right). In this case, the reduction is associated with INDindustry emissions abatement, relatively constant throughout the observed period, and an increasing abatement along the studied time window in DOMdomestic and TRAtransport sectors from UNECE and anthropogenic emissions in ROW (Figure 9Figure 11 bottom right).

#### 4. **Main findings and discussion**

Implementing more stringent air quality and GHG emission abatement policies only in the UNECE region (MFR UNECE scenario) leads to limited benefits in the air pollution exposure in this region because their effect is partially offset by the unabated emissions from non-UNECE countries, when similar measures are not implemented there as well. Such effect is more pronounced for O<sub>3</sub> than for PM<sub>2.5</sub>.

In CLE, ~~The~~ the main single contributor to the O<sub>3</sub> exposure in the UNECE region is non-anthropogenic O<sub>3</sub> (OTHER/NATURAL), including biogenic and other unspecified sources (mainly soil-derived NO<sub>x</sub>, lightning and stratospheric intrusion), which remains relatively constant at ca. 35 ppb throughout the entire time window (2020 – 2050). In this scenario, the anthropogenic fraction of the O<sub>3</sub> exposure is equivalent to 16 - 19 ppb. TRA, IND, SHPTransport, industry and maritime sectors contribute to this fraction mainly via NO<sub>x</sub>-VOC precursors' emissions while AGR, FLR and WSTagriculture, gas flaring and waste management contribute mostly via emissions of the CH<sub>4</sub> precursor. ENEEnergy production is the only source affecting O<sub>3</sub> exposure with similar shares for both precursor families.

The overall upward trend in the O<sub>3</sub> related mortality in the UNECE region over the studied time window is mainly associated with the increasing share of CH<sub>4</sub> emissions from ROW. The O<sub>3</sub> exposure shares of AGR, WST, FLRagriculture, waste management, gas flaring and ENEenergy production CH<sub>4</sub> emissions from ROW shows an upward trend along the simulated time window while the one of TRA, ENEtransport, energy production and DOMdomestic NO<sub>x</sub>-VOC emissions from UNECE shows ~~the~~ an opposite trend.

Unlike O<sub>3</sub>, anthropogenic UNECE emissions are the main source of PM<sub>2.5</sub> exposure and related mortality in UNECE countries. However, due to a reduction in the share of UNECE emissions and an increase in that from ROW, the importance of the former decreases from 70% to 65% of the total PM<sub>2.5</sub> exposure metric over the simulated time window.

As a whole, the MFR BASE leads to 34% and 41 % mortality reductions with respect compared to the CLE scenario in 2050 for O<sub>3</sub> and PM<sub>2.5</sub> exposure, respectively, while the MFR-SDS leads to a total abatement of mortality in 2050 with respect compared to CLE of 41% and 50% for O<sub>3</sub> and PM<sub>2.5</sub> exposure, respectively.

~~One of the limitations of the adopted methodology is that secondary organic aerosol chemistry is not considered. In addition, the TM5-FASST model does not include feedbacks from changing chemical regimes when switching off individual precursor emissions. Nevertheless, the estimated levels and source allocation is~~  
The applied methodology, based on a reduced form model, has several limitations we discuss here. Some of the limitations are inherited from the parent TM5 CTM. This is the case for secondary organic aerosol chemistry which is not considered and leads to a conservative estimate of PM<sub>2.5</sub> exposure and consequently of the benefits from controls. The omission of secondary organic PM in TM5 is estimated to introduce a low bias in the PM<sub>2.5</sub> concentration of the order of 0.1 µg/m<sup>3</sup> as global mean. However, regional levels in central Europe and China can reach up to 1 µg/m<sup>3</sup> in areas where average levels of primary organic matter are 20 µg/m<sup>3</sup> (Van Dingenen et al., 2018). In addition, the TM5-FASST model does not include non-linear responses due to changing chemical regimes when switching off individual precursor emissions, nor does it consider impacts of future climate change on photolysis rates and on natural emissions that may affect ozone chemistry. Although an evaluation of climate-chemistry interactions is beyond the capabilities and the scope of the TM5-FASST model, we briefly discuss their possible impacts on our conclusions. In terms of changing meteorology, a warmer climate is expected to cause higher surface ozone production due to higher photolysis rates and more stagnant conditions which would call for more stringent controls than anticipated under present climate in order to meet limit levels. The climate penalty on summertime surface ozone concentrations is estimated to be in the range 1 – 10 ppb, with highest impacts in polluted conditions (Jacob and Winner, 2008). In terms of emissions, a warming climate will increase CH<sub>4</sub> emissions from wetlands, the major natural CH<sub>4</sub> source (Gedney et al., 2004). Also natural VOC emissions from vegetation are expected to increase with increasing temperature – up to a critical temperature after which emissions decrease again (e.g. 38°C for isoprene). The impact of increased natural VOC (including methane) on the O<sub>3</sub> response to NO<sub>x</sub> emission changes depends on the chemical regime. In NO<sub>x</sub>-saturated (VOC-limited) conditions, the climate-driven increased VOC emissions will increase the natural component of O<sub>3</sub> formation, and drive the chemical regime more towards the NO<sub>x</sub>-limited region, implying a higher response of O<sub>3</sub> to anthropogenic NO<sub>x</sub> emission changes. This situation is only characteristic of strongly polluted urban areas. Under the more common conditions of VOC-saturation (NO<sub>x</sub>-limitation), the O<sub>3</sub> response to NO<sub>x</sub> is only weakly dependent on the VOC concentrations (Akimoto and Tanimoto, 2022).

The applied TM5-FASST methodology, not including these climate-chemistry feedbacks, is likely to underestimate the natural component of O<sub>3</sub> formation in a future, warmer climate, as well as the O<sub>3</sub> response to NO<sub>x</sub> reductions in specific polluted conditions. However, this does not compromise our conclusion that control of anthropogenic CH<sub>4</sub> emissions can play a prominent and increasing role in the coming decades.

The estimated levels and source allocation in our study are comparable with those obtained in studies with similar scope. However, using previous studies as reference is not straightforward due to different underlying methodological assumptions and aggregation of the output data. This is particularly true when comparing the source apportionment with brute-force or emission reduction impact approach (used in this study) with the one resulting from tagged method studies- (Appendix A, Figure A2).

## 5. Conclusions

The scenario analysis presented in this study assesses the exposure to O<sub>3</sub> and PM<sub>2.5</sub> and associated mortality between 2020 and 2050 in the UNECE countries. To that end, a baseline scenario in which the air quality and GHG abatement measures adopted by 2018 are implemented (CLE) is compared with other scenarios with increasing degree of ambition. The adopted methodology for the identification of geographical origin with sectoral anthropogenic sources and precursors detail led to an in-depth understanding of the impact that different measures may have on mortality in the UNECE region in the medium and long-term.

The study demonstrates that applying emission reductions only in UNECE countries leads to a limited abatement in the O<sub>3</sub> exposure in UNECE countries with respect to the baseline (CLE) and that the implementation of BATs worldwide would not only lead to higher abatement of exposure in UNECE countries but also to a trend reversal, from increasing to decreasing. Moreover, the study shows that the overall upward trend in the O<sub>3</sub>-related mortality in the UNECE region over the studied time window is mainly associated with the growing share of CH<sub>4</sub> emissions from ROW. This is mostly related to the relatively long atmospheric lifetime of O<sub>3</sub> (compared to PM<sub>2.5</sub>), formed from its short-lived precursors NO<sub>x</sub> and NMVOC, and to the one of its other-long-lived precursor CH<sub>4</sub> which contributes to global background O<sub>3</sub>. On the contrary, PM<sub>2.5</sub> related mortality in UNECE appears to be mainly affected by domestic~~its own~~ emissions.

Controlling O<sub>3</sub> exposure in UNECE countries is necessary to prevent the CLE projected increase in annual mortality from ca. 65,000 in 2020 to ca. 73,500 in 2050 (+9,000 deaths/year), while acting on PM<sub>2.5</sub> is a high priority to avoid the considerable mortality attributed to this pollutant turning back in 2050 to the same levels of 2020 (ca. 444,000 units). The analysis of the CLE scenario suggests the opportunity to act on CH<sub>4</sub> sources AGR, ENE, FLR and WST~~agriculture, energy production, gas flaring and waste management~~ beyond the UNECE region (ROW) in order to prevent an increase in O<sub>3</sub> exposure and related mortality in the UNECE countries from 2030 onwards- (in addition to the benefits for the ROW region). On the contrary, to significantly reduce the PM<sub>2.5</sub> exposure and related mortality in the UNECE region beyond the CLE measures in the long term (2050), the main focus should be on the anthropogenic emissions from AGR~~agriculture~~ and IND~~industry~~ sectors within the UNECE region.

In MFR-SDS, the abatement of some of the most critical CH<sub>4</sub> sources identified in the analysis of CLE (ENE, FLR~~energy production, gas flaring~~ and WST~~waste management~~) plus the reduction of NO<sub>x</sub>-VOC from IND~~industry~~ and TRA~~transport~~ globally and SHP~~those of the maritime sector~~ lead to a 30% - 41% drop of O<sub>3</sub>-related mortality with respect to CLE in 2030

430 and 2050 (equal to ca. 20,000 – 30,000 avoided premature deaths/year), respectively. Moreover, the abatement of the most critical UNECE PM<sub>2.5</sub> emissions identified in the analysis of CLE (i.e. AGRagriculture and INDindustry) plus DOMemissions in the domestic sector complemented by reductions in natural sources (DUST and SS) lead to a -44% - 50% drop in the PM<sub>2.5</sub> related mortality compared to CLE in 2030 and 2050 (equal to ca. 182,000 – 221,000 avoided premature deaths/year)-, respectively.

435 The analysis of MFR-SDS scenario confirms that the measures in line with UN SDGs concerning energy sources can lead to significant benefits. It also shows the potential co-benefits of joint air quality and GHG abatement policies in line with Paris Agreement ambition of keeping the global average temperature increase below 2°C. However, considering the impact of AGRagriculture, an important NH<sub>3</sub> contributor, on the two studied pollutants in the CLE scenario, more ambitious reductions of this source should be explored considering that the abatement of NH<sub>3</sub> in the MFR scenarios compared to CLE is modest (-  
440 32 % to -35% in UNECE in the studied time window).

The conclusions of this study are relevant for the revision of the UNECE's Air Convention Gothenburg protocol-~~under~~ progress.

#### Code/Data availability

Code and Data are available on request

445 Author contribution

Both authors contributed equally to all the phases of the work and manuscript drafting.

#### Competing interests

The authors declare they have no conflict of interest.

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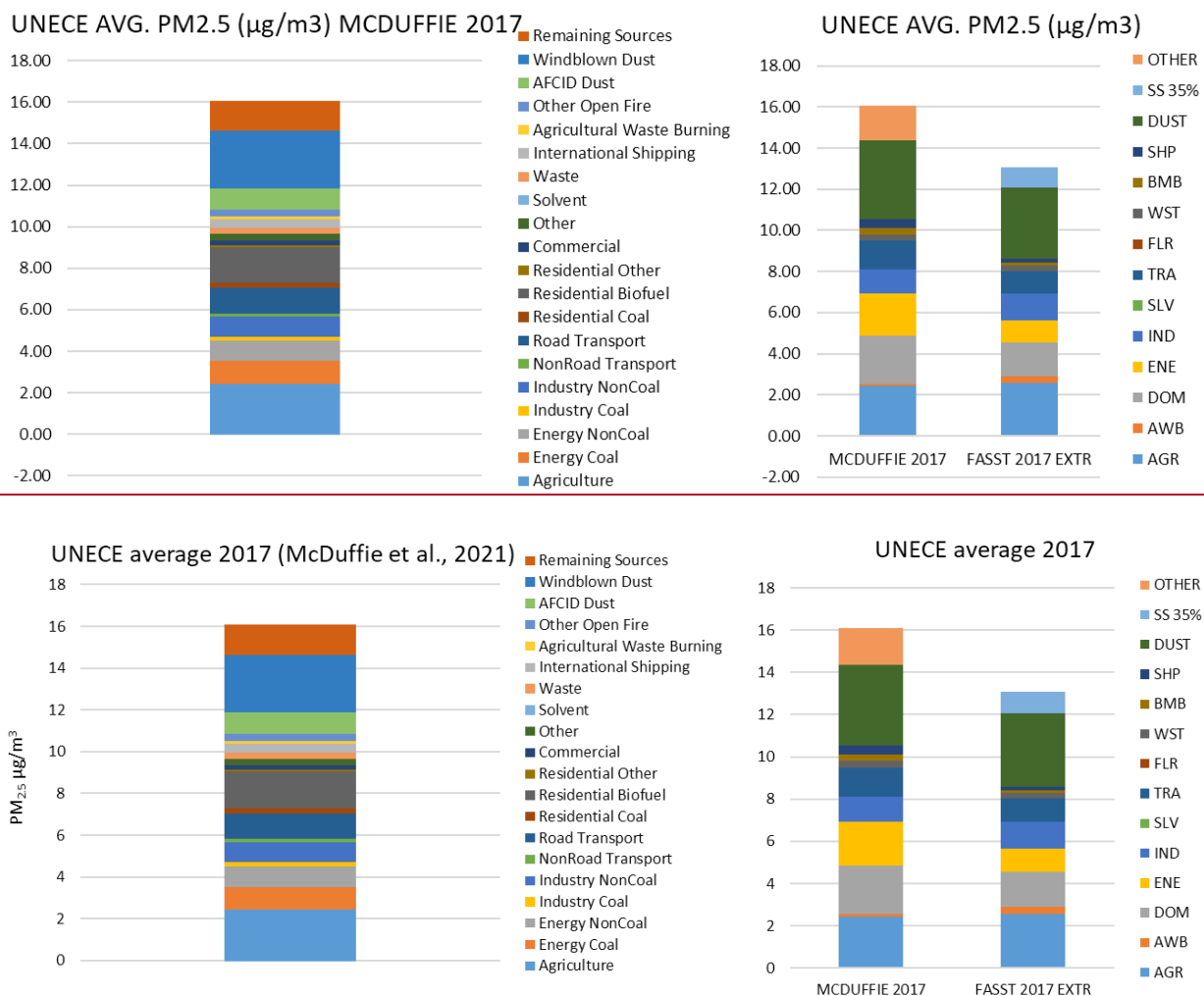


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Comparison with other studies

The source allocation of average PM<sub>2.5</sub> exposure in UNECE described in the present study is comparable with the one reported by Mc Duffie et al. (2021) for all world countries in 2017 on the basis of a combination of satellite data, chemical transport models and ground based observations. The UNECE average population weighted PM<sub>2.5</sub> split in 20 source categories including fuel details obtained from the country averages reported in the abovementioned study is shown in Figure A1 left. Such categories are merged using the same categories as the present study for comparison with the estimations obtained with TM5-FASST (~~present study~~) extrapolated for 2017 (Figure A1 right).

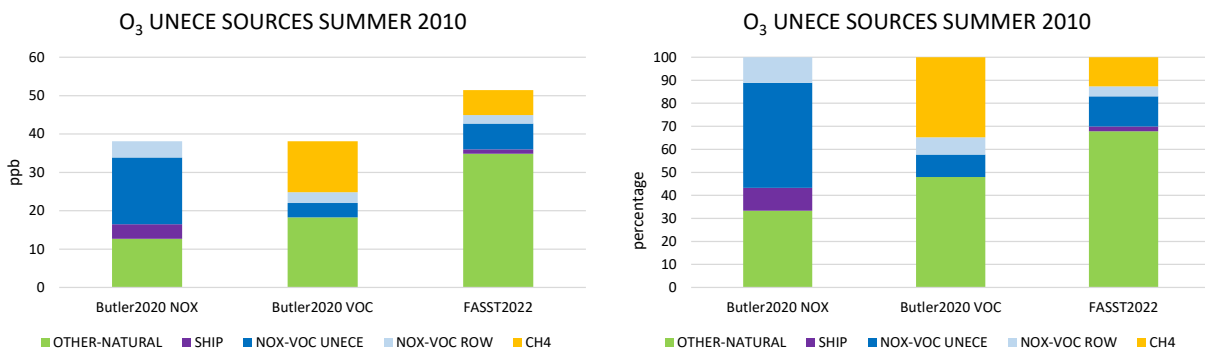


535 Figure A1. UNECE average population weighted PM<sub>2.5</sub> split by source categories. Left: Original source categories (Mc Duffie et al., 2021); Right: comparison of PM<sub>2.5</sub> source apportionment of the present study with the one ~~on~~ by Mc Duffie et al. 2021 ~~the left~~ using the same source categories.

540 The average UNECE population weighted PM<sub>2.5</sub> from TM5-FASST is 2.4 µg/m<sup>3</sup> (-18%) lower than the one obtained from the country values reported by Mc Duffie and co-authors likely due to the use of data fusion in the latter. The population weighted PM<sub>2.5</sub> allocated by TM5-FASST to ENEenergy production and DOMdomestic is lower than the one reported in the abovementioned study (-47% and -29%, respectively). On the contrary,- the higher while AWBagricultural waste share-is higher in TM5-FASST (+160%) has been attributed to the incorporation of forest fires under this category in this model than the one reported in the abovementioned study (Figure A1 right). This is likely due to the incorporation of forest fires under this category in TM5 FASST.

545 The UNECE O<sub>3</sub> source allocation in the 2010 warm season (April-September) obtained in this study with TM5-FASST based on a perturbation approach was compared with the one reported by Butler et al. (2020) using a tagging approach (hereon Butler2020). Comparing the two outputs is, however, not straightforward because Butler2020 splits the total O<sub>3</sub> concentrations in two alternative ways either by NO<sub>x</sub> precursors or by VOC precursors while TM5-FASST splits them to NO<sub>x</sub>-VOC and CH<sub>4</sub> both precursors at once. Moreover, in Butler2020 Central Asia (CAS) VOC contributions as well as those from Israel are included in ROW while in this study these countries have been accurately attributed to included in the UNECE region.

550 The O<sub>3</sub> concentrations are higher in TM5-FASST compared to Butler2020 likely due to the use of maximum daily 8h averages instead of monthly averages as Butler2020 (Figure A2). The share of O<sub>3</sub> produced by NO<sub>x</sub>-VOC emitted in UNECE according to TM5-FASST (6 ppb, 13%) lies in-between the estimations obtained by Butler2020 for the contribution of NO<sub>x</sub> (17 ppb, 45%) and NMVOC (4 ppb, 10%) emissions in this region. By comparison, the share of O<sub>3</sub> deriving from NO<sub>x</sub>-VOC emissions from ROW provided by TM5-FASST (2 ppb, 4%) is slightly lower than the estimations by Butler2020 for NO<sub>x</sub> (4 ppb, 11 %) and VOC (3 ppb, 7%), respectively.



**Figure A2. UNECE average O<sub>3</sub> split by sources categories using a tagged approach (Butler et al., 2020) and a perturbation approach (TM5-FASST, this study) expressed as concentrations (left) and percentages (right).**

560 Butler2020 links the CH<sub>4</sub>-related O<sub>3</sub> only to VOC emissions and does not associate this precursor to any specific geographic area while TM5-FASST allocates CH<sub>4</sub>-related O<sub>3</sub> to its geographic source regions and precursors. In this analysis the TM5-FASST aggregated share of O<sub>3</sub> associated with CH<sub>4</sub> (6 ppb, 13%) is considerably lower than the one attributed by Butler2020 to this fraction (13 ppb, 35%). Also the contribution of shipping to O<sub>3</sub> concentrations estimated by Butler2020 (4 ppb, 10%)

is higher than the share reported by TM5-FASST in this study (1 ppb, 2%). By comparison, the role of Other-Natural source is higher in TM5-FASST (35 ppb, 67%) compared with the one attributed by Butler2020 (13 ppb, 33% for NO<sub>x</sub> and 18 ppb, 48% for VOC source allocation, respectively).

**Description-Brief description of scenarios**

The scenarios used in this study are summarised in Table A1.

570 **Table A1. Description of ECLIPSE version 6b global scenarios used in this study (IIASA, 2021; Klimont et al., in preparation).**

Scenario	abbreviation	Air quality policy	Climate policy
Current legislation (baseline)	CLE	Assumes the implementation of the future commitments included in the air quality legislation in force by 2018. Current baseline projections according to the IEA World Energy Outlook 2018 New Policy Scenario (NPS) which includes EU 2030 renewable energy and energy efficiency targets and announced energy policies by China, USA, Japan and Korea.	Incorporates only commitments made in the national determined contributions (NDC) under the Paris Agreement.
Maximum technical reduction baseline	MFR BASE	Stringent policy assuming introduction of best currently available technology and no cost limitations. However, no further technological improvements are foreseen. Same activity drivers as CLE following NPS.	Incorporates only commitments made in the NDCs under the Paris Agreement.
Maximum technical reduction sustainable development	MFR-SDS	Similar to MFR BASE. However, relies on the most ambitious IEA sustainable development scenario (SDS). Includes outcomes of energy-related SDGs: reducing	Aligned with Sustainable Development Goal #13 and Paris Agreement goal of holding global average temperature increase below 2 °C.

		dramatically premature deaths due to energy-related air pollution and universal access to modern energy by 2030.	
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The current legislation baseline (CLE) scenario considers fuel consumption from IEA (International Energy Agency), agriculture data from FAO (UN Food and Agriculture Organisation) and IFA (International Fertilizer Organization), and statistics on industry, waste, shipping, etc., from other sources (IEA, 2018).