https://doi.org/10.5194/acp-2022-776
Preprint. Discussion started: 5 December 2022

Atmospheric Chemistry and Physics
Discussions

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

Claudio A. Belis[1], Rita Van Dingenen[1]

[1] European Commission, Joint Research Centre, Via Fermi 2749, 21027 Ispra, Italy

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 UNECE region on the exposure to O_3 and $PM_{2.5}$ 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 V6 air pollutant and greenhouse gases emission reduction scenarios. The mortalities were attributed to O_3 and $PM_{2.5}$ 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_3 and related mortality to two families of precursors: NO_X-VOC and CH_4. In this study the baseline scenario (CLE), that 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 from this comparison is that O_3 exposure within the UNECE area is more sensitive to measures outside the UNECE region, than $PM_{2.5}$ exposure even though the latter leads to higher mortality than the former. In the current legislation scenario (CLE), the mortality associated with O_3 exposure in the UNECE region grows steadily from 2020 to 2050. The upward trend is mainly associated with the growing impact of CH_4 emissions from areas outside UNECE. Also, the mortality related to NO_X-VOC emissions outside UNECE increases in the same period. By comparison, a measurable decrease is observed in the mortality attributable to NOx-VOC emissions from UNECE. In the same time window, the mortality associated with $PM_{2.5}$ exposure in the UNECE region at first decreases between 2020 and 2040 and then rises until 2050. The $PM_{2.5}$ 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 $PM_{2.5}$-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 SDGs and the Paris Agreement can lead to significant co-benefits between air quality and climate policies.

1. 1 Introduction

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.

https://doi.org/10.5194/acp-2022-776
Preprint. Discussion started: 5 December 2022

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 $PM_{2.5}$- related mortality in the former (Burnett and Cohen, 2020). The Convention on Long-Range Transport of Air Pollution (also known as the Air Convention) of the United Nations Economic Commission for Europe (UNECE) was adopted in 1979 and has 51 parties, 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_4) emissions. Ground-level ozone (O_3) concentrations in most of the UNECE region countries are also influenced by other factors in addition to the regional ozone precursors: e.g. climatic parameters, hemispheric transport and global CH_4 emissions (Butler et al., 2020). Global background levels of O_3, $PM_{2.5}$ and their precursors, including CH_4 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_4 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 study is to identify to what extent the abatement policies within the UNECE region and in the rest of the world (ROW) influence the exposure to O_3 and $PM_{2.5}$ 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 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.

2. Methods

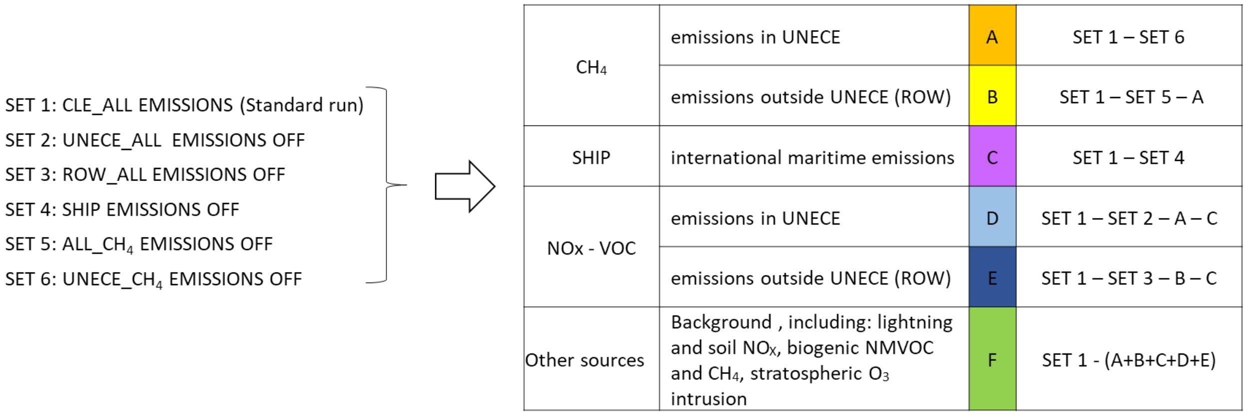
2.1. Exposure and health impact assessment

The TM5-FAst Scenario Screening Tool (TM5-FASST) is a simplified model based on linear emission concentration sensitivities derived from the full TM5 model (Krol et al., 2005), to calculate the impacts of air pollution globally. The exposure metrics are the population weighted PM2.5 concentrations and the seasonal daily maximum 8h ozone average (SDMA8h). The mortality associated with these pollutants is estimated according to the Global Burden of Disease (GBD) approach (Stanaway et al., 2018). An overview of TM5-FASST methodology is available in the supplementary material and a complete description is provided in Van Dingenen, et al. (2018) and Belis et al. (2022).

2.2. Sources

The contributions from anthropogenic sectors and natural emissions to $PM_{2.5}$ and O_3 exposure metrics in the UNECE region are estimated by 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 (SHIP). Fire emissions were added from SSP2-CMIP6 (projections) and van Marle et al. (2017), including large-scale biomass burning and savannah burning and excluding AWB emissions to avoid double counting. The resulting anthropogenic $PM_{2.5}$ concentration fields are overlaid with fixed natural $PM_{2.5}$ 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_3, the abovementioned sectoral attribution was complemented with runs separating the impact of VOC-NO_X and CH_4 precursor emissions from: UNECE (continental, anthropogenic), ROW (rest of the world: non-UNECE continental, anthropogenic), sources that cannot be separated between UNECE and ROW like international shipping (SHIP), and non-anthropogenic sources associated with biogenic and other sources according to the scheme described in Figure 1 .



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_4 EMISSIONS OFF
SET 6: UNECE_CH_4 EMISSIONS OFF

CH_4	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
NOx - 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_X, biogenic NMVOC and CH_4, stratospheric O_3 intrusion	F	SET 1 - (A+B+C+D+E)

Figure 1. Approach adopted to split O_3 concentrations by emission area (UNECE and non UNECE (ROW)) and by precursor (VOC-NO_X and CH_4).

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_X, 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).

In Appendix A, the $PM_{2.5}$ and O_3 source apportionment presented in this study is compared with similar studies in the literature.

The obtained shares for the $PM_{2.5}$ and O_3 exposure metrics are converted to total mortalities according to:

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

Where *EXPOSURE METRIC* $_{total}$ is the sum of all individual sources (*x*) shares.

2.3. Scenarios

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 Jones and O'Neill (2016) which are in line with the Shared Socioeconomic Pathways (SSP) (Riahi, et al., 2017). 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_3 and $PM_{2.5}$ precursors in all the studied scenarios are shown in Figure 2. In the **CLE scenario**, UNECE NO_X, VOC and $PM_{2.5}$ emissions decrease by 33%, 13% and 13%, respectively, between 2020 and 2050 while in ROW, NH_3 and CH_4 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_3, which after an initial decrease remains stable. Moreover, NH_3 is the only precursor with a distinct 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% ($PM_{2.5}$) and 35% (NH_3) lower than CLE while ROW emissions are between 80% ($PM_{2.5}$) and 37% (NH_3) lower than CLE. Despite MFR-SDS emissions follow similar trends, the reductions with respect to the CLE are higher, with the exception of NH_3 which is similar in both MFR scenarios.

https://doi.org/10.5194/acp-2022-776
Preprint. Discussion started: 5 December 2022

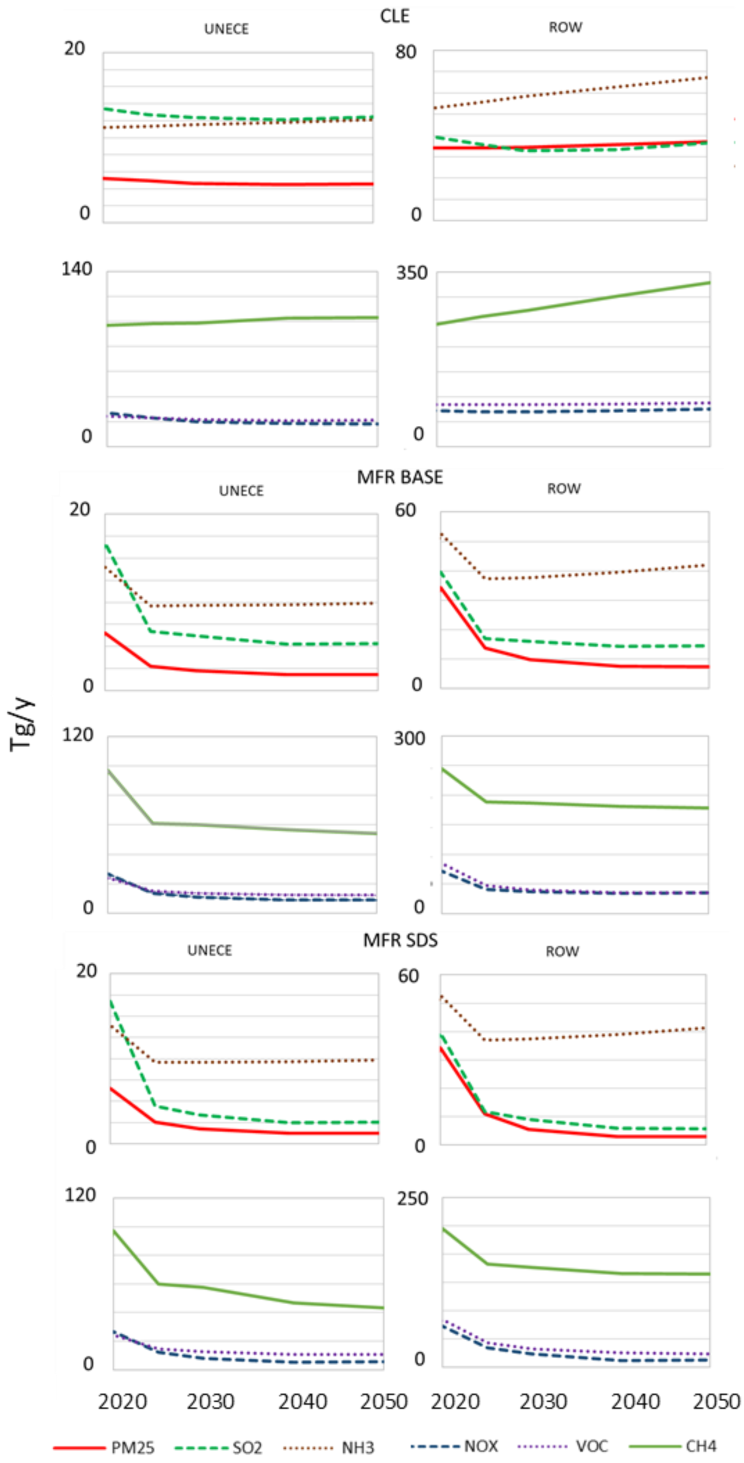


Figure 2. UNECE (left) and ROW (right) emission trends of main O_3 and $PM_{2.5}$ precursors in the studied ECLIPSE V6b scenarios

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 $PM_{2.5}$ (anthropogenic) and O_3 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 ROW (so called MFR UNECE scenario) (Figure 3).

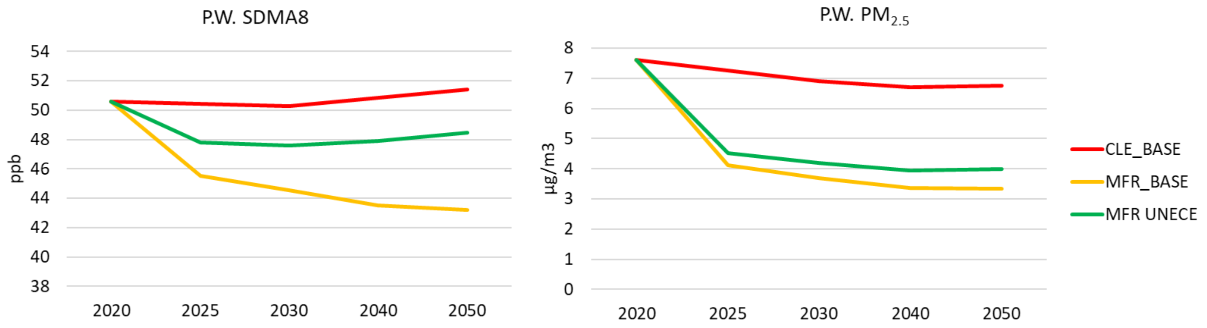


Figure 3. O_3 seasonal mean of 8hr (population weighted SDMA8h, left) and anthropogenic population weighted $PM_{2.5}$ (right) annual averages in UNECE region, average of countries, under different scenarios. CLE (current legislation), MFR BASE only in UNECE countries (MFR UNECE), MFR BASE in all countries (MFR BASE).

The O_3 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_3 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_3 exposure in UNECE countries relative to the baseline (CLE). By comparison, the O_3 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.

Unlike O_3, $PM_{2.5}$ 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 MFR BASE scenario globally leads to a relatively small marginal benefit (≤ 10% of CLE). In synthesis, for $PM_{2.5}$ abatement, UNECE is only slightly affected by ROW measures, while O_3 levels are strongly modulated by measures taken outside the UNECE region. This is obviously related to the longer (compared to $PM_{2.5}$) atmospheric lifetime of O_3 formed from its short-lived precursors NO_x and NMVOC, and of its other long-lived precursor CH_4 which contributes to global background O_3. The attribution of O_3 and $PM_{2.5}$ levels to precursor emissions in- and outside the UNECE region is further investigated in the following sections.

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

In this section, the O_3 exposure and related mortality inside 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_3 background share is estimated from total O_3 minus the sum of all anthropogenic sectors (see section 2.2).

The main single contributor to the O_3 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, little affected by anthropogenic emissions in the short term. In the 2020 – 2050 time window, the anthropogenic fraction of the O_3 exposure is worth 16 - 19 ppb.

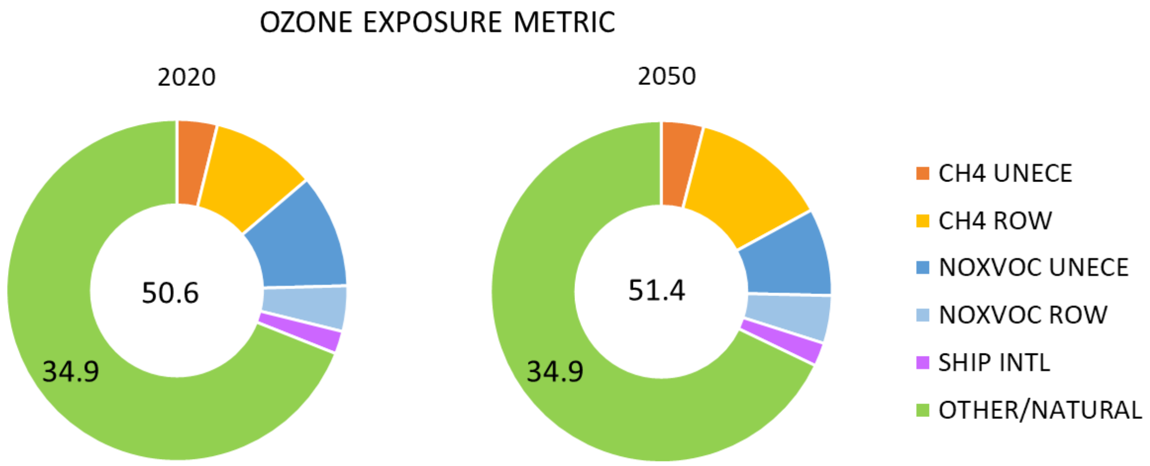


Figure 4. Allocation of the population weighted O_3 (SDMA8h) exposure in UNECE to geographic source areas (UNECE, ROW), precursors and other/natural sources. Units: ppb

In terms of precursors, in CLE there is a remarkable shift in the relative role of short-lived components (NO_X, NMVOC) versus CH_4 between 2020 and 2050. The initial dominant role of NO_X and NMVOC in anthropogenic ozone formation is replaced by CH_4 towards 2050. This is due to the combined decrease of UNECE NO_X and VOC emissions (while ROW emissions remain relatively constant) and increase of ROW CH_4 emissions (while UNECE emissions remain relatively constant). The overall O_3 exposure metric is stable along the observed time window because the decreasing impact of NO_X-VOC emissions from UNECE over time is largely compensated by the increasing impact of CH_4 emitted in ROW.

The overall share of O_3 exposure allocated to anthropogenic NO_X-VOC emissions is mainly associated with TRA, IND, SHP sources while the CH_4 emissions affecting this pollutant are mainly emitted from AGR, FLR and WST. ENE, another important anthropogenic source, presents similar shares of both precursor families (Figure S1).

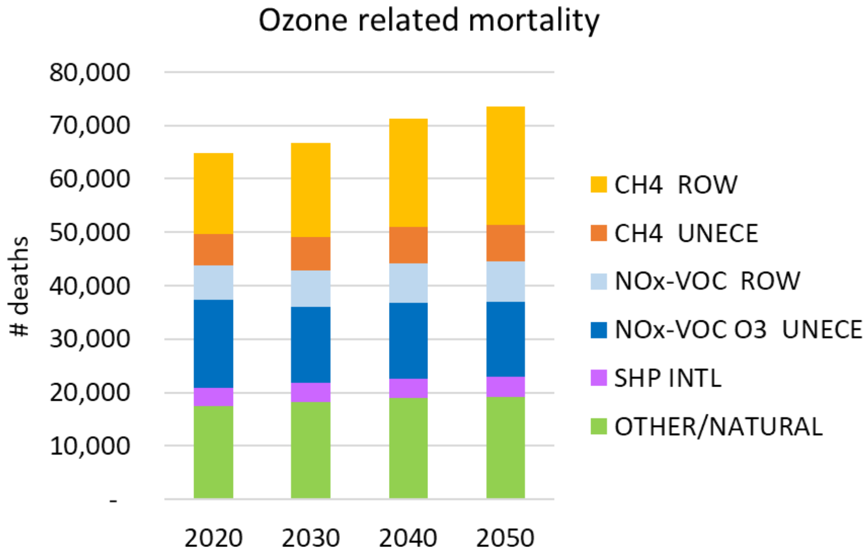


Figure 5. Mortality (UNECE total) associated with O_3 exposure in UNECE split by natural-background (only the fraction above the exposure threshold) and anthropogenic emissions.

In Figure 5 the premature mortality associated with O_3 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_4 emissions from ROW (+46 %, +7,000 deaths/year). Also the mortality related to anthropogenic NO_X-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_X-VOC emissions in UNECE which drops from 16,000 in 2020 to 14,000 in 2050.

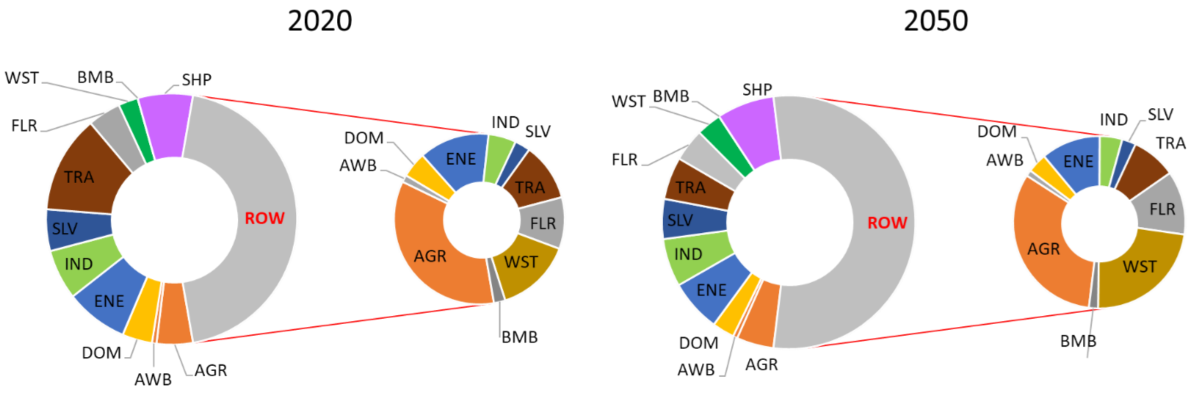


Figure 6. Allocation of O_3 exposure and related mortality (UNECE avg.) to anthropogenic sources under CLE. The overall impacts are represented in the main pie charts while the small pie charts to the left of them show the detail of ROW impacts only. AGR: agriculture, AWB: agricultural waste burning, 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 and ENE in 2020 to a one dominated by AGR, WST, TRA and ENE in 2050 (Figure 6). TRA, IND and SHP contribute to O_3 exposure only via NO_X-VOC precursors while AGR, FLR and WST contribute almost only via emissions of CH_4 (Figure S1).
The CH_4 impact of AGR, FLR, WST and ENE emissions from ROW on O_3 exposure in UNECE presents an upward trend between 2020 and 2050 (Figure S1). In the same time window, the NO_X-VOC contribution from TRA, ENE and DOM emissions from UNECE show a downward trend with the exception of IND which increases slightly. Although ENE is the only source which shares of O_3 exposure due to NO_X-VOC and CH_4 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 $PM_{2.5}$ exposure and premature mortality in UNECE in the baseline scenario

The UNECE anthropogenic emissions are the main responsible for $PM_{2.5}$ 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 7).

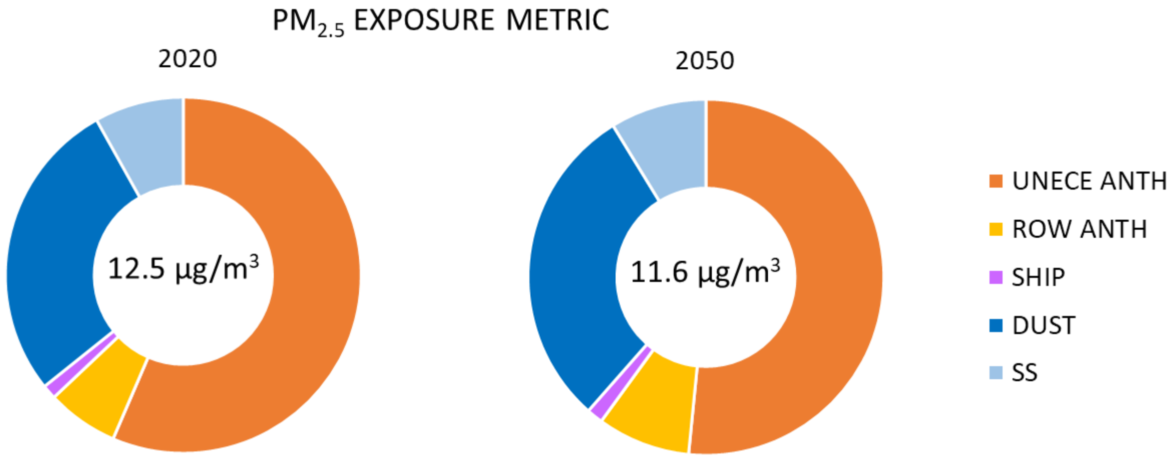


Figure 7. Allocation of the population weighted $PM_{2.5}$ exposure in UNECE to geographic source areas (UNECE, ROW) and natural sources under CLE.

The mortality associated with $PM_{2.5}$ 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).

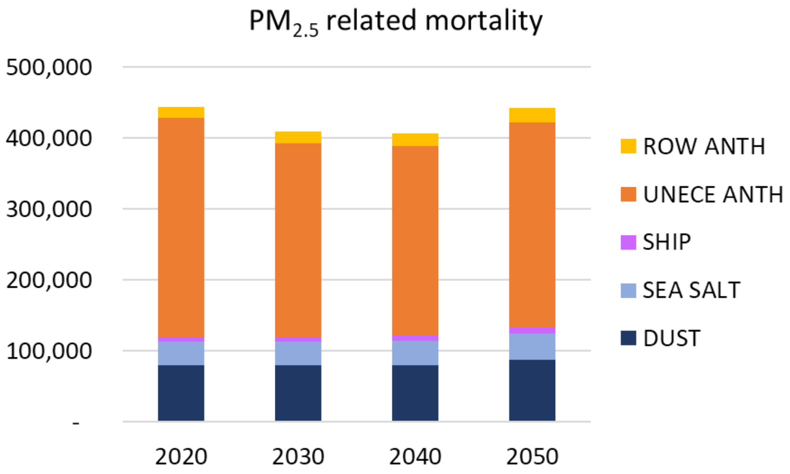


Figure 8. Mortality (UNECE avg.) associated with PM$_{2.5}$ exposure attributable to both anthropogenic and natural sources under CLE.

The main anthropogenic contributors within UNECE are: AGR, IND, DOM, ENE and TRA (Figure 9). An overall downward trend in the impact of DOM, ENE and TRA from UNECE and an increasing role of IND and AGR from this region are observed. The share of SHP, a contributor which is not geographically allocated in this analysis, is stable from 2020 onwards. In 2050, there is an increase in the PM$_{2.5}$ exposure mainly due to a rise in the impact of AGR, TRA, FLR and WST emissions from ROW and AGR and IND emissions from the UNECE region.

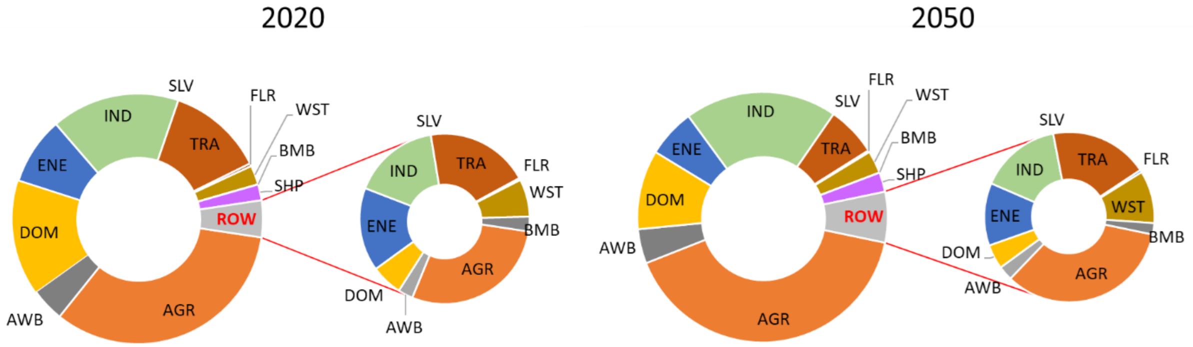


Figure 9. Allocation of PM$_{2.5}$ exposure and related mortality (UNECE avg.) to anthropogenic sources under CLE. The overall impacts are represented in the main pie charts while the small pie charts to the left of them show the detail of ROW impacts only. . AGR: agriculture, AWB: agricultural waste 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

This section evaluates the trends of the O_3 and $PM_{2.5}$ 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 10). In 2050, the MFR BASE and MFR-SDS O_3 exposure is 16% and 20% lower than CLE, respectively, while the $PM_{2.5}$ (anthropogenic) exposure in the abovementioned scenarios is 51 % and 59% below CLE, respectively.

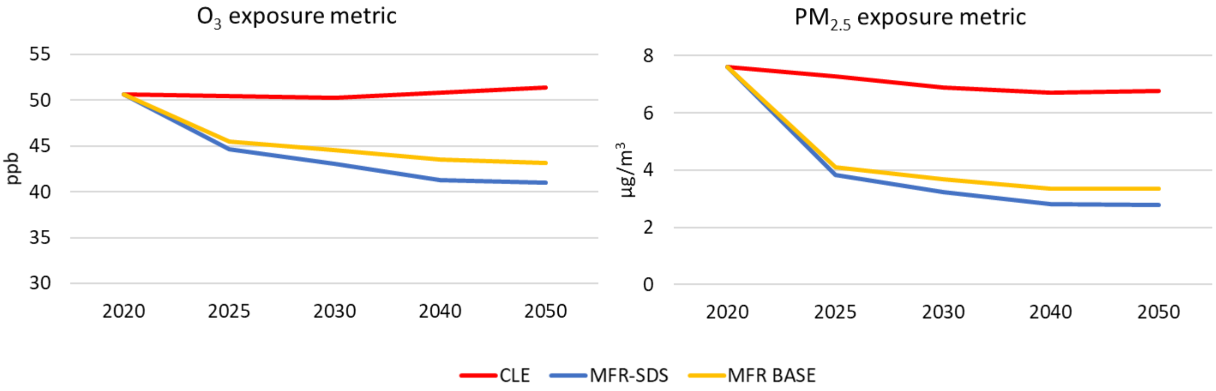


Figure 10. O_3 and anthropogenic $PM_{2.5}$ exposure metrics (UNECE avg.) computed with TM5-FASST according to the ECLIPSE V 6b scenarios: CLE, MFR BASE and MFR-SDS.

In the period 2025 – 2050, the main anthropogenic contributor to O_3 exposure and mortality in both MFR scenarios is by far AGR due to CH_4 emissions in ROW (Figure S2).

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_3 exposure (Figure 11 top left). Such improvement is mainly associated with NO_X-VOC emission reductions in the UNECE region and reductions of CH_4 in ROW, the role of which increases considerably between 2025 and 2050 (Figure 11 top left). A more detailed analysis of the MFR BASE reveals that the main UNECE NO_X-VOC emission reductions in 2050 are associated with ENE, IND and TRA sectors while those of CH_4 in ROW are mainly due to abatement of FLR and ENE in 2025 with dramatic abatement increase in the WST sector between this year and 2050 (Figure 12 top).

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_X-VOC emissions in both UNECE and ROW (Figure 11 bottom left). Such abatement of O_3-related mortality in the MFR SDS scenario is associated with emission reductions in the TRA sector in 2050 in both UNECE and ROW compared to 2020 (Figure 12 bottom).

https://doi.org/10.5194/acp-2022-776
Preprint. Discussion started: 5 December 2022

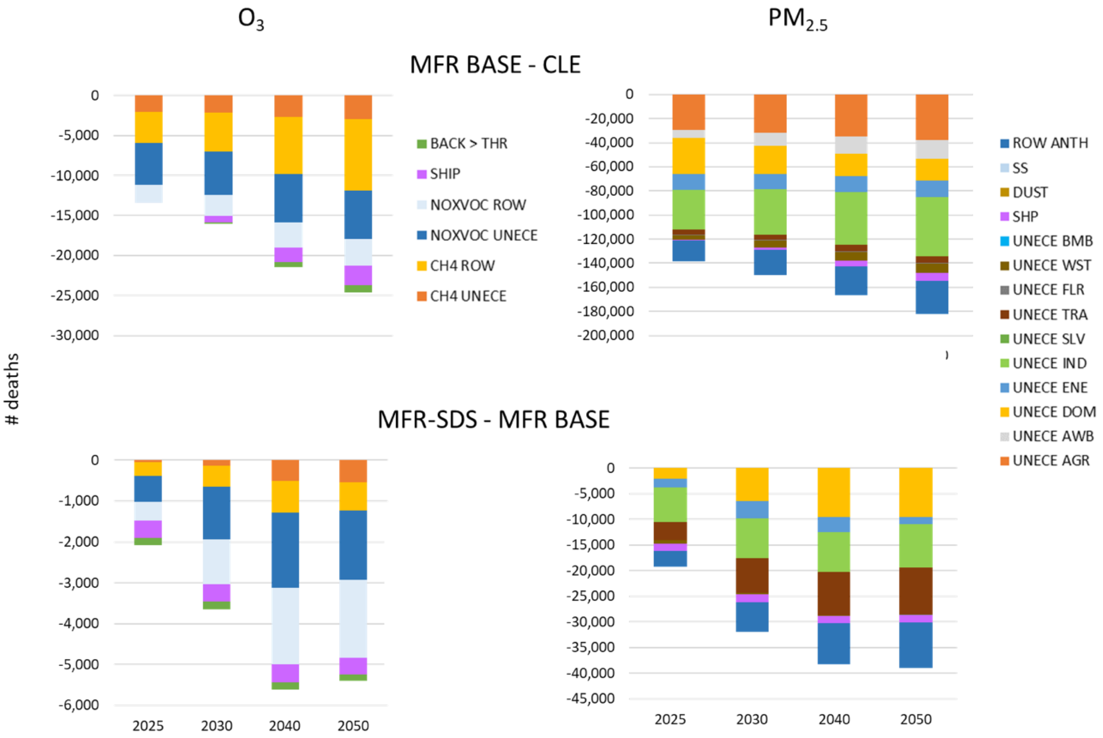


Figure 11. Delta MFR BASE - CLE and MFR-SDS – MFR BASE of O_3 (left) and $PM_{2.5}$ (right) associated mortality (UNECE total) split by precursor and main emission areas. For O_3 we only consider the fraction of 'OTHER/NATURAL' exceeding the zero effect threshold of 29.1 ppb.

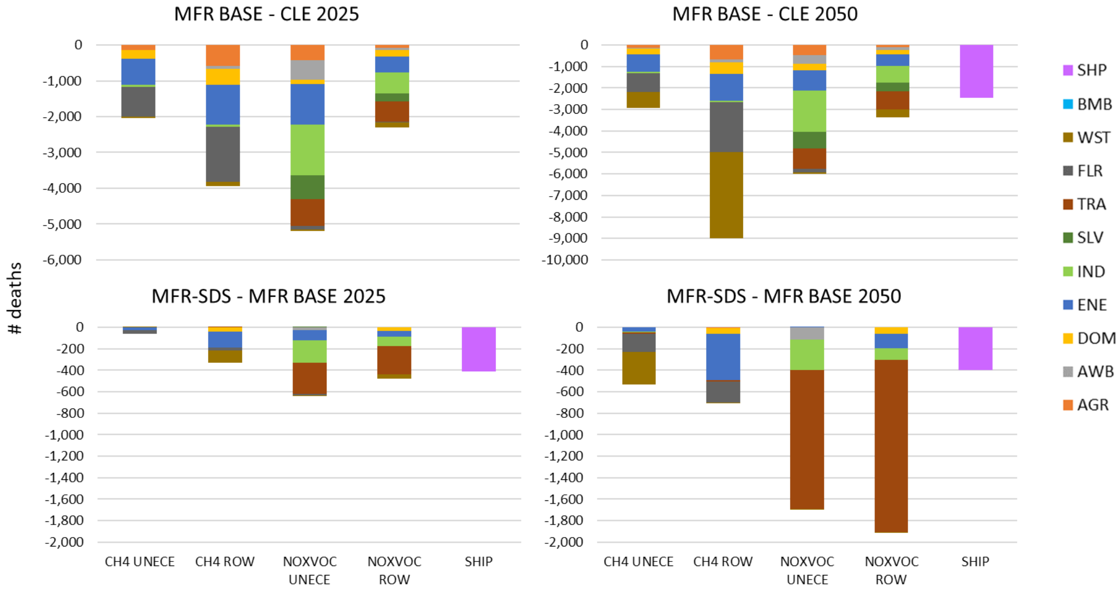


Figure 12. Delta MFR BASE – CLE (top) and MFR-SDS – MFR BASE (bottom) of UNECE O_3 associated mortality in 2025 and 2050 split by source sectors.

In MFR BASE the delta mortality in UNECE due to $PM_{2.5}$ exposure compared to the CLE ranges from ca. -137,000 cases (-33%) in 2025 to ca. -187,000 cases (-41%) in 2050 (Figure 11 top right). Such improvement is mainly due to abatement of emissions in the AGR and IND sectors in UNECE. In this region, the abatement of DOM emissions shows a decreasing importance between 2025 and 2050 while the opposite is true for AWB 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 11 bottom right). In this case, the reduction is associated with IND emissions abatement, relatively constant throughout the observed period, and an increasing abatement along the studied time window in DOM and TRA from UNECE and anthropogenic emissions in ROW (Figure 11 bottom right).

4. Main findings

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_3 than for $PM_{2.5}$.

In CLE, The main single contributor to the O_3 exposure in the UNECE region is non-anthropogenic O_3 (OTHER/NATURAL), including biogenic and other unspecified sources (mainly soil-derived NO_X, 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_3 exposure is equivalent to 16 - 19 ppb. TRA, IND, SHP contribute to this fraction mainly via NO_X-VOC precursors' emissions while AGR, FLR and WST contribute mostly via emissions of the CH_4 precursor. ENE is the only source affecting O_3 exposure with similar shares for both precursor families.

The overall upward trend in the O_3 related mortality in the UNECE region over the studied time window is mainly associated with the increasing share of CH_4 emissions from ROW. The O_3 exposure shares of AGR, WST, FLR and ENE CH_4 emissions from ROW shows an upward trend along the simulated time window while the one of TRA, ENE and DOM NO_X-VOC emissions from UNECE shows the opposite trend.

Unlike O_3, anthropogenic UNECE emissions are the main source of $PM_{2.5}$ 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 $PM_{2.5}$ exposure metric over the simulated time window.

As a whole, the MFR BASE leads to 34% and 41 % mortality reductions with respect to the CLE scenario in 2050 for O_3 and $PM_{2.5}$ exposure, respectively, while the MFR-SDS leads to a total abatement of mortality in 2050 with respect to CLE of 41% and 50% for O_3 and $PM_{2.5}$ 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

https://doi.org/10.5194/acp-2022-776
Preprint. Discussion started: 5 December 2022

precursor emissions. Nevertheless, the estimated levels and source allocation is 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.

5. Conclusions

The scenario analysis presented in this study assesses the exposure to O_3 and $PM_{2.5}$ 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 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_3 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_3-related mortality in the UNECE region over the studied time window is mainly associated with the growing share of CH_4 emissions from ROW. This is mostly related to the relatively long atmospheric lifetime of O_3 (compared to $PM_{2.5}$) formed from its short-lived precursors NO_x and NMVOC, and the one of its other long-lived precursor CH_4 which contributes to global background O_3. On the contrary, $PM_{2.5}$ related mortality in UNECE appears to be mainly affected by domestic emissions.

Controlling O_3 exposure in UNECE counties 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 $PM_{2.5}$ 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_4 sources AGR, ENE, FLR and WST beyond the UNECE region (ROW) in order to prevent an increase in O_3 exposure and related mortality in the UNECE countries from 2030 onwards. On the contrary, to significantly reduce the $PM_{2.5}$ 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 and IND sectors within the UNECE region.

In MFR-SDS, the abatement of some of the most critical CH_4 sources identified in the analysis of CLE (ENE, FLR and WST) plus the reduction of NO_X-VOC from IND and TRA globally and SHP lead to a 30% - 41% drop of O_3-related mortality with respect to CLE in 2030 and 2050 (equal to ca. 20,000 – 30,000 avoided premature deaths/year), respectively. Moreover, the abatement of the most critical UNECE $PM_{2.5}$ emissions identified in the analysis of CLE (i.e. AGR and IND) plus DOM complemented by reductions in natural sources (DUST and SS) lead to a 44% - 50% drop in the $PM_{2.5}$ related mortality compared to CLE in 2030 and 2050 (equal to ca. 182,000 – 221,000 avoided premature deaths/year) , respectively.

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 AGR, an important NH_3 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_3 in the MFR scenarios compared to CLE is modest (-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.

Acknowledgements

The authors are grateful to Julian Wilson for reviewing the English style of the manuscript.

References

Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F., and Winiwarter, W.: Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications, Environmental Modelling & Software, 26, 1489-1501, https://doi.org/10.1016/j.envsoft.2011.07.012, 2011.

Belis, C. A., Pernigotti, D., Pirovano, et al.: Evaluation of receptor and chemical transport models for PM10 source apportionment, Atmospheric Environment: X, 5, 100053, https://doi.org/10.1016/j.aeaoa.2019.100053, 2020.

Belis, C. A., Van Dingenen, R., Klimont, Z., and Dentener, F.: Scenario analysis of PM2.5 and ozone impacts on health, crops and climate with TM5-FASST: A case study in the Western Balkans, Journal of Environmental Management, 319, 115738, https://doi.org/10.1016/j.jenvman.2022.115738, 2022.

Burnett, R., and Cohen, A.: Relative Risk Functions for Estimating Excess Mortality Attributable to Outdoor PM2.5 Air Pollution: Evolution and State-of-the-Art, in: Atmosphere, 6, 2020.

Butler, T., Lupascu, A., and Nalam, A.: Attribution of ground-level ozone to anthropogenic and natural sources of nitrogen oxides and reactive carbon in a global chemical transport model, Atmos. Chem. Phys., 20, 10707-10731, 10.5194/acp-20-10707-2020, 2020.

HEI: State of Global Air 2020, Health Effects Institute, Boston, USA, 2020.

IIASA, Global emission fields of air pollutants and GHG: https://iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv6.html, 2021.

IEA: World Energy Outlook 2018, International Energy Agency 2018.

Jonson, J. E., Schulz, M., Emmons, L., Flemming, J., Henze, D., Sudo, K., Tronstad Lund, M., Lin, M., Benedictow, A., Koffi, B., Dentener, F., Keating, T., Kivi, R., and Davila, Y.: The effects of intercontinental emission sources on European air pollution levels, Atmos. Chem. Phys., 18, 13655-13672, 10.5194/acp-18-13655-2018, 2018.

Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., and Schöpp, W.: Global anthropogenic emissions of particulate matter including black carbon, Atmos. Chem. Phys., 17, 8681-8723, 10.5194/acp-17-8681-2017, 2017.

Klimont, Z., Höglund-Isaksson, L., Heyes, C., Rafaj, P., Schöpp, W., Cofala, J., Purohit, P., Borken-Kleefeld, J., Kupiainen, K., Kiesewetter, G., Winiwarter, W., Amann, M, Zhao, B., Wang, S.X., Bertok, I., Sander, R. Global scenarios of air pollutants and methane: 1990-2050. In preparation.

Krol, M., Houweling, S., Bregman, B., van den Broek, M., Segers, A., van Velthoven, P., Peters, W., Dentener, F., and Bergamaschi, P.: The two-way nested global chemistry-transport zoom model TM5: algorithm and applications, Atmos. Chem. Phys., 5, 417-432, 10.5194/acp-5-417-2005, 2005.

Lefohn, A. S., Malley, C. S., Smith, L., Wells, B., Hazucha, M., Simon, H., Naik, V., Mills, G., Schultz, M. G., Paoletti, E., De Marco, A., Xu, X., Zhang, L., Wang, T., Neufeld, H. S., Musselman, R. C., Tarasick, D., Brauer, M., Feng, Z., Tang, H., Kobayashi, K., Sicard, P., Solberg, S., and Gerosa, G.: Tropospheric ozone assessment report: Global ozone metrics for climate change, human health, and crop/ecosystem research, Elementa, 6, 10.1525/elementa.279, 201

McDuffie, E. E., Martin, R. V., Spadaro, J. V., Burnett, R., Smith, S. J., O'Rourke, P., Hammer, M. S., van Donkelaar, A., Bindle, L., Shah, V., Jaeglé, L., Luo, G., Yu, F., Adeniran, J. A., Lin, J., and Brauer, M.: Source sector and fuel contributions to ambient PM2.5 and attributable mortality across multiple spatial scales, Nature Communications, 12, 3594, 10.1038/s41467-021-23853-y, 2021.

Revell, L. E., Tummon, F., Stenke, A., Sukhodolov, T., Coulon, A., Rozanov, E., Garny, H., Grewe, V., and Peter, T.: Drivers of the tropospheric ozone budget throughout the 21st century under the medium-high climate scenario RCP 6.0, Atmos. Chem. Phys., 15, 5887-5902, 10.5194/acp-15-5887-2015, 2015.

Riahi, K., van Vuuren, D. P., Kriegler, E., et al.: The Shared Socioeconomic Pathways and their energy, land use, and greenhouse gas emissions implications: An overview, Global Environmental Change, 42, 153-168, https://doi.org/10.1016/j.gloenvcha.2016.05.009, 2017.

Stanaway, J. D., Afshin, A., Gakidou et al., C. J. L.: Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks for 195 countries and territories, 1990–2017: a systematic analysis for the Global Burden of Disease Study 2017, The Lancet, 392, 1923-1994, https://doi.org/10.1016/S0140-6736(18)32225-6, 2018.

Turner, M. C., Jerrett, M., Pope, C. A., Krewski, D., Gapstur, S. M., Diver, W. R., Beckerman, B. S., Marshall, J. D., Su, J., Crouse, D. L., and Burnett, R. T.: Long-Term Ozone Exposure and Mortality in a Large Prospective Study, American Journal of Respiratory and Critical Care Medicine, 193, 1134-1142, 10.1164/rccm.201508-1633OC, 2015.

Turnock, S. T., Wild, O., Dentener, F. J., Davila, Y., Emmons, L. K., Flemming, J., Folberth, G. A., Henze, D. K., Jonson, J. E., Keating, T. J., Kengo, S., Lin, M., Lund, M., Tilmes, S., and O'Connor, F. M.: The impact of future emission policies on tropospheric ozone using a parameterised approach, Atmos. Chem. Phys., 18, 8953-8978, 10.5194/acp-18-8953-2018, 2018.

Van Dingenen, R., Dentener, F., Crippa, M., Leitao, J., Marmer, E., Rao, S., Solazzo, E., and Valentini, L.: TM5-FASST: a global atmospheric source–receptor model for rapid impact analysis of emission changes on air quality and short-lived climate pollutants, Atmos. Chem. Phys., 18, 16173-16211, 10.5194/acp-18-16173-2018, 2018.

van Marle, M. J. E., Kloster, S., Magi, B. I., Marlon, J. R., Daniau, A. L., Field, R. D., Arneth, A., Forrest, M., Hantson, S., Kehrwald, N. M., Knorr, W., Lasslop, G., Li, F., Mangeon, S., Yue, C., Kaiser, J. W., and van der Werf, G. R.: Historic global biomass burning emissions for CMIP6 (BB4CMIP) based on merging satellite observations with proxies and fire models (1750–2015), Geosci. Model Dev., 10, 3329-3357, 10.5194/gmd-10-3329-2017, 2017.

Appendix A

Comparison with other studies

The source allocation of average $PM_{2.5}$ 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 $PM_{2.5}$ 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 for comparison with the estimations obtained with TM5-FASST (present study) extrapolated for 2017 (Figure A1 right).

Figure A1. UNECE average population weighted $PM_{2.5}$ split by source categories. Left: Original source categories (Mc Duffie et al., 2021); Right: comparison of $PM_{2.5}$ source apportionment of the present study with the one on the left using the same source categories.

The average UNECE population weighted $PM_{2.5}$ from TM5-FASST is 2.4 µg/m^3 (-18%) lower than the one obtained from the country values reported by Mc Duffie and co-authors. The population weighted $PM_{2.5}$ allocated by TM5-FASST to ENE and DOM is lower (-47% and -29%, respectively) while AWB is higher (+160%) 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.

The UNECE O_3 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_3 concentrations in two alternative ways either by NO_X precursors or by VOC precursors while TM5-FASST splits them to 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 the UNECE region.

The O_3 concentrations are higher in TM5-FASST likely due to the use of maximum daily 8h averages instead of monthly averages as Butler2020 (Figure A2). The share of O_3 produced by NO_X-VOC emitted in UNECE according to TM5-FASST (6 ppb, 13%) lies in-between the estimations obtained by Butler2020 for the contribution of NO_X (17ppb, 45%) and NMVOC (4 ppb, 10%) emissions in this region. By comparison, the share of O_3 deriving from NO_X-VOC emissions from ROW provided by TM5-FASST (2 ppb, 4%) is slightly lower than the estimations by Butler2020 for NO_X (4 ppb, 11 %) and VOC (3 ppb, 7%), respectively.

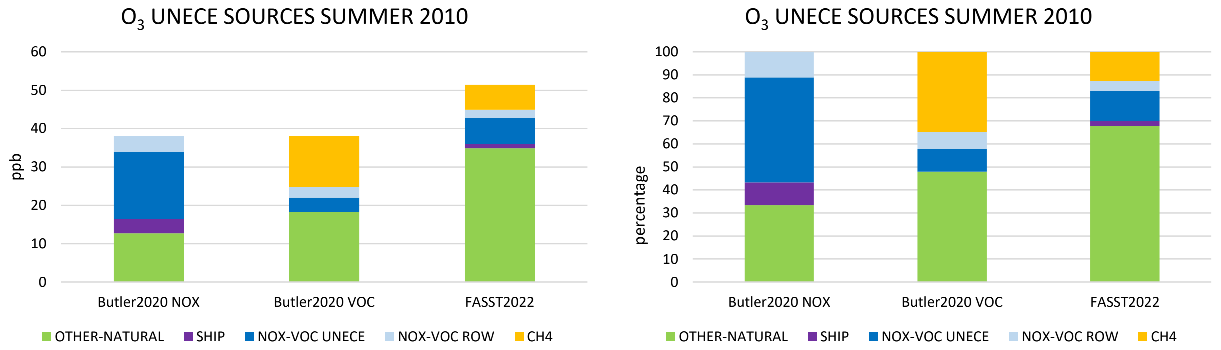


Figure A2. UNECE average O_3 split by sources categories using a tagged approach (Butler et al., 2020) and a perturbation approach (TM5-FASST, this study) expressed as concentrations (left) and percentages (right).

Butler2020 links the CH_4-related O_3 only to VOC emissions and does not associate this precursor to any specific geographic area while TM5-FASST allocates CH_4-related O_3 to its geographic source regions and precursors. In this analysis the TM5-FASST aggregated share of O_3 associated with CH_4 (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_3 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_X and 18 ppb, 48% for VOC source allocation, respectively).

Description of scenarios

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

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	Incorporates only commitments made in the national determined contributions (NDC) under the Paris Agreement.

		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.	
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 dramatically premature deaths due to energy-related air pollution and universal access to modern energy by 2030.	Aligned with Sustainable Development Goal #13 and Paris Agreement goal of holding global average temperature increase below 2 °C.

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). 425