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# Forty years of improvements in European air quality: the role of EU policy–industry interplay

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## Abstract

The EDGAR (Emissions Database for Global Atmospheric Research) v4.3 global anthropogenic emissions inventory of several gaseous ( $\text{SO}_2$ ,  $\text{NO}_x$ , CO, non-methane volatile organic compounds (NMVOCs) and  $\text{NH}_3$ ) and particulate ( $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , black and organic carbon (BC and OC)) air pollutants for the period 1970–2010 is used to develop retrospective air pollution emission scenarios to quantify the roles and contributions of changes in fuels consumption, technology, end-of-pipe emission reduction measures and their resulting impact on health and crop yields. This database presents changes in activity data, fuels and air pollution abatement technology for the past 4 decades, using international statistics and following guidelines for bottom-up emission inventory at the Tier 1 and Tier 2 levels with region-specific default values.

With two further retrospective scenarios we assess (1) the impact of the technology and end-of-pipe (EOP) reduction measures in the European Union (EU) by considering a stagnation of technology with constant emission factors from 1970 and with no further abatement measures and improvement in European emissions standards, but fuel consumption occurring at historical pace, and (2) the impact of increased fuel consumption by considering unchanged energy use with constant fuel consumption since 1970, but technological development and end-of-pipe reductions. Our scenario analysis focuses on the three most important and most regulated sectors (power generation, the manufacturing industry and road transport), which are subject of multi-pollutant EU Air Quality regulations.

If technology and European EOP reduction measures had stagnated at 1970 levels, EU air quality in 2010 would have suffered from 129 % higher  $\text{SO}_2$ , 71 % higher  $\text{NO}_x$  and 69 % higher  $\text{PM}_{2.5}$  emissions, demonstrating the large role of technology in reducing emissions in 2010. However, if fuel consumption had remained constant starting in 1970, the EU would have benefited from current technology and emission control standards, with reductions in  $\text{NO}_x$  by even 13 % more. Such further savings are not observed for  $\text{SO}_2$  and  $\text{PM}_{2.5}$ . If the EU consumed the same amount of fuels as in 1970

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but with the current technology and emission control standards, then the emissions of SO<sub>2</sub> and PM<sub>2.5</sub> would be 42 % respectively 10 % higher. This scenario shows the importance for air quality of abandoning heavy residual fuel oil and shifting fuel types (from, e.g., coal to gas) in the EU.

5 A reduced-form TM5-FASST (Fast Screening Scenario Tool based on the global chemical Transport Model 5) is applied to calculate regional and global levels of aerosol and ozone concentrations and to assess the impact of air quality improvements on human health and crop yield loss, showing substantial impacts of export of EU technologies and standards to other world regions.

## 10 **1 Introduction**

In the last few decades, air quality has become more and more important worldwide due to the fast pace of industrialization in many countries (Fenger, 2009). Air pollution negatively affects human health (Pope and Dockery, 2006; Anderson et al., 2012), influences climate, visibility and ecosystems (Monks et al., 2009; IPCC, 2013), and therefore has significant impacts on human life and the environment. Of high importance are the quantification and understanding of the impacts of primary and secondary anthropogenic air pollutants which are released into the atmosphere by large- and small-scale combustion, industrial processes, transportation, waste disposal, agriculture and forest and land-use change. Emission inventories have been developed in order to quantify total and sector-specific emissions at the country, regional and global levels (e.g., <http://www.emep.int/>, <http://www.epa.gov/ttn/chief/eiinformation.html>, <http://htap.org>, last access: May 2015).

For some industrialized countries, legislation has come into effect since the mid-1980s for the power generation sector, and since mid-1990s for road transport, while, for example, in China regulations have been implemented only more recently for these two sectors (CSC, 2013). Therefore, although developing countries have started regulating their air pollutant emissions, here we focus only on European air quality legis-

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lation, having a much longer history and affecting not only Europe, but several other countries elsewhere in the world (e.g., vehicle emissions in Japan and other Asian countries) (Crippa et al., 2015).

The European Union (EU), as well as the extended UNECE/CLTRAP/EMEP region (http://www.ceip.at/ms/ceip\_home1/ceip\_home/ceip\_unece/), has introduced several air quality protocols and legislation to reduce pollutant emissions in the atmosphere from anthropogenic activities (combustion processes, energy production, transportation, etc.). Figure 1 and Table S7.1 in the Supplement summarize most of European regulations for air quality and air pollutant emissions from after 1970, when the first air quality directive was introduced. European policies are classified into “air quality directives” when regulating pollutant concentrations in the air, “directives regulating air pollutants emissions from anthropogenic activities” when dealing with emission limits for specific activities, “EU standards on road vehicle emissions” and “fuel quality directives”. A broader air quality regulation framework is given by international conventions which were created to promote the improvement of global air quality, like the Convention on Long-range Transboundary Air Pollution (CLRTAP) created in 1979 and later extended by several protocols to address environmental issues and to identify measures and policies to lower anthropogenic air pollutant emissions, or the Gothenburg Protocol (GP) of 1999, recently revised in 2012.

In our work we make use of the EDGAR v4.3 emission data (Emissions Database for Global Atmospheric Research, http://edgar.jrc.ec.europa.eu/index.php) to compare the present situation with retrospective scenarios (1970–2010) developed with the aim of assessing the importance of the role of changes in fuel use and air pollution abatement technology in determining the trends of air pollutant emissions in Europe and around the world, and their impact on air quality, health and vegetation.

Most literature on emission scenarios focuses on projecting actual emissions into the future to assess possible pathways of air quality and climate in view of new policies. So far, limited attention has been given to assess the real achievements in emission savings (reducing emissions) which the policy–industry interplay under existing air

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quality legislation and current technology brought in. The implementation of air quality directives with current technological means varies in effectiveness and is far from 100 % effective. Some works have analyzed past emissions trends for the most important air pollutants, but mainly focused on selected substances or specific regions (e.g., Klimont et al. (2013) for global  $\text{SO}_2$  or Kurokawa et al. (2013) for all pollutants in the Asian region). Historical global emission data sets for the past decades or century are compiled by combining data from several emission inventories, e.g., Lamarque et al. (2010) for 1850–2000 and Granier et al. (2011) for 1980–2010; however, an analysis of potential drivers of the emissions is difficult because of the heterogeneity and regional differences in the cause of variability of the original data that might show inconsistencies over the full time period and in global coverage. Amann et al. (2013) report the evolution of anthropogenic emissions of key air pollutants between 1990 and 2010 for several world regions as obtained from the GAINS (Greenhouse Gas Air Pollution Interactions and Synergies) model. The same system is used to provide scenarios of future emissions (up to 2050) depending on specific assumptions of air quality and climate policies (e.g., Cofala et al., 2007). A different approach is used by Rafaj et al. (2013), who aim to identify the factors (historical energy balances, population and economic growth, fuel mix, etc.) driving emission levels of air pollutants in Europe from 1960 to 2010, based on the RAINS (Regional Air Pollution and Simulation) and GAINS (<http://gains.iiasa.ac.at/models/>) models. They decomposed

the emissions to understand potential drivers of the trends. Here, we do not seek to analyze driver factors and further expectations of optimized reduction policies, but we want to take stock of the achievements based on consistently estimated emissions with reductions in the EU by technological progress and end-of-pipe measures and reported shifting of fuel types.

Our work focuses both on European and global historical emissions for a complete set of air pollutants, developing retrospective scenarios for the years 1970–2010 to assess the role and impact of the most important European air quality policies, identifying regional hotspots where these emission changes took place and estimating the corre-

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sponding impacts on health and vegetation. Historical emissions are provided for all countries by only one emission inventory (EDGAR v4.3) for several gaseous air pollutants ( $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{CO}$ , non-methane volatile organic compounds (NMVOCs) and  $\text{NH}_3$ ) and particulate matter ( $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , black and organic carbon (BC and OC)).

Two retrospective scenarios are developed from a European point of view in addition to the actual world “reference” emissions for 1970 and 2010, as schematically sketched in Fig. 2. Compared to future near-term scenarios 2010–2050, retrospective scenarios 1970–2010 have the advantage of using the well-known activity data time series. Obviously, no new policies can be proposed retrospectively, but the implementation of policies over time varied strongly and an evaluation of the savings and the further potential of the existing regulations might shed light on the needs for further strengthening the implementation of some policies.

The first retrospective scenario (STAG\_TECH) simulates the stagnation of technology with constant emission factors since 1970 and with no further abatement measures such as EU emissions standards, but uses the fuel consumption increase as historically reported. This scenario aims at assessing the benefit of a combination of technological progress and of EU emission quality standard policies on actual air quality. In this work, we do not disentangle the contributions of (EU) policy, of a globalizing industry with clean technology suppliers, or of a global fuel market. It is evident that the EU did not take emission reduction actions unilaterally, but mutually with other industrialized countries, using agreed efforts under the CLRTAP and the Gothenburg Protocol (GP). To model the stagnation of technology and of end-of-pipe measures in Europe, one has to assume constant emission factors representing the older technology deployed at a larger regional (or even global) scale but also assume no European control standard measure that is deployed either locally such as end-of-pipe abatement on industrial facilities or globally on vehicles.

The second retrospective scenario (STAG\_FUEL) assumes unchanged energy use with constant fuels consumption since 1970 while emission factors and abatements improve such as in the reference data for the historical emission time series. This

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scenario aims at assessing the offset of the emission savings with technology and end-of-pipe abatement by the increase in fuel consumption. In 2010 this scenario is expected to provide lower emissions than the 2010 reference scenario, mainly because of increasing per capita energy needs. This is most remarkably noted in the energy and transport sectors, which show at global levels a 3.6-fold and 2.6-fold increase over four decades, which is much more than the global population increase (1.8-fold). Even though energy efficiency increased, rebound effects were identified by, e.g., Barker et al. (2009) and Brännlund et al. (2007). In addition, the historical fuel consumption time series show shifts in the energy mix. The latter is country-specific and depends on natural reserves, the fuel price and stability, the energy stored per unit of fuel volume. Air quality and climate effects of a shifting of fuel types might be opposite, e.g., in the case of diesel cars (with lower CO<sub>2</sub> per terajoule but larger BC per terajoule emission factors) vs. petrol cars (with larger CO<sub>2</sub> per terajoule but lower BC per terajoule) as discussed in Clerides et al. (2008). Fuel mix choices are discussed in more detail at the European level, as well as the emission control standards, because of the European focus of this paper.

Finally, deploying the TM5-FASST (Fast Screening Scenario Tool based on the global chemical Transport Model 5) source–receptor model, (Tavoni et al., 2014), the impact of the different scenarios on health and crops, being primary objectives of policies, is screened. Improving air quality leads to positive effects on human health, reducing respiratory and cardiovascular diseases at reduced PM levels (Anenberg et al., 2012), and enhances crop yields at reduced O<sub>3</sub> levels (Shindell et al., 2011).

## 2 Methodology

In Sect. 2.1 an overview of the data and assumptions used to develop different emission scenarios is provided, addressing the EDGAR v4.3 methodology for the REF (reference), STAG\_Tech and STAG\_Fuel emissions. This is followed by a short descrip-

tion of the TM5-FASST model used here to screen the impact of the considered emission scenarios on pollutant concentrations, human health and crop yields in Sect. 2.2.

## 2.1 EDGAR v4.3 emission data and scenarios

The Emission Database for Global Atmospheric Research (EDGAR) version v4.3 (<http://edgar.jrc.ec.europa.eu/index.php>) is used as the reference inventory of anthropogenic emissions, providing global grid maps of sector-specific historical emission data from 1970 to 2010 for SO<sub>2</sub>, NO<sub>x</sub>, CO, NMVOCs (not chemically speciated, but as total), NH<sub>3</sub>, as well as particulate matter components, namely PM<sub>10</sub>, PM<sub>2.5</sub>, BC, OC. EDGAR v4.3 relies on international energy balances of IEA (2014, R. Quadrelli, personal communication, 2014) and agricultural statistics of FAO (2012) and regional or national information and assumptions on technology use and emission control standards. EDGAR v4.3 is one of the few global emission inventories with consistent emission time series covering 4 decades for the subject pollutants with high spatial resolution of 0.1 × 0.1° and consistent sector-specific breakdowns. Moreover, recent comparisons show the reliability of this emission inventory based on the good agreement between the EDGAR v4.3 2008 and 2010 emission data and the best estimates provided by official national data merged in the HTAP\_v2.2 data set (Janssens-Maenhout et al., 2015). Evaluation of the time series is performed by comparison of the data set with the AR5 data set of Lamarque et al. (2010) and the MACCity data set of Granier et al. (2011). A more detailed comparison between EDGARv4.3 and MACCity was possible at the country level and is documented in Sect. S5 in the Supplement, showing regional differences from few percent up to 50 %, which is within the range of uncertainties.

The EDGAR data sets are calculated under a consistent bottom-up approach with full time series of the activity data and allow straightforward implementation of scenario assumptions. We start from the calculation of the reference emissions of a specific pollutant  $x$  at time  $t$  due to activity data (AD) of sector  $i$  with technologies  $j$  and end-of-pipe measures  $k$ , in a country  $C$  as follows:

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$$EM_{C,i}(t, x) = \sum_{j,k} [AD_{C,i}(t) \cdot TECH_{C,i,j}(t) \cdot EOP_{C,i,j,k}(t) \cdot EF_{C,i,j}(t, x) \cdot (1 - RED_{C,i,j,k}(t, x))], \quad (1)$$

with TECH representing the penetration fraction of a specific technology in a sector, EOP the installed fraction of end-of-pipe measures, EF the uncontrolled emission factors and RED the emission reduction associated with the end-of-pipe control measures. We treat all 12 sectors of Table 1 separately and for each sector we provide a global grid map per pollutant and per month in 2010 as well as the time series 1970–2010 (annual grid maps) on <http://edgar.jrc.ec.europa.eu/pegasos/>. An overview of 2010 pollutant emissions is also reported in Sect. S2 for the three scenarios for 24 world regions.

We then focus on the main sectors that were effectively targeted by air quality measures imposed on the industrial sector by EU policies<sup>1</sup>. Firstly, the European power industry (IPCC category A) represents large national point sources, which continuously emit over a long period and since the 1980s have been equipped with additional end-of-pipe control measures, which is modeled at the Tier 2 level. The industrial combustion processes that are suitable to be regulated are the non-power generation industry (IPCC category 2A), in particular the manufacturing industry (cement, steel and non-ferrous metal industry, chemical<sup>2</sup> production, paper/food/textiles/wood and machinery production). This sector was subject to a much faster change in technology and market globalization, with a strong combined change in emission factors and (end-of-pipe) control measures, modeled at the Tier 1 level. Road transport is the third sector that has been effectively regulated in Europe since the 90s, with standards for the automobile industry modeled at the Tier 2 level. Detailed information about processes, technologies and abatement measures adopted for the three sectors of interest are summarized in Sect. S6.

<sup>1</sup>In 2010, the EU had 27 member states and is therefore defined as such in this study.

<sup>2</sup>Not including the petrochemical industry with oil production and refining.

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We modeled the STAG\_TECH scenario by assuming for the three sectors of interest constant emission factors to all technologies present in the emission model for the EU (specified mainly regionally, but few also globally). We further assumed in the STAG\_TECH scenario no implementation of European end-of-pipe control measures.

Other regional standards regulating end-of-pipe control, e.g., US power plant standards were not changed, because they fall outside the European scope of this study. Thus in STAG\_TECH, European energy is generated by the power plants of 1970 without additional end-of-pipe measures (in other words the emission reduction equals the lower limit<sup>3</sup>). A relatively large turnover of power plants was observed in the 1970s and 1980s (Platts database, 2007, <http://www.platts.com/>), resulting in a high share of power plants which reached 30–40 years of operational time in 2010. For the manufacturing industry, the effect of technology stagnation could only be reflected by keeping the emission factors, modeled at regional or global levels, constant. For road transport, the stagnation was mainly reflected by the removal of particle filters and catalysts of all the vehicles under EURO standards, mainly present in the fleet inside Europe but also in some Asian countries (or EURO standards 1 to 5 equalling the pre-EURO standards). The EURO standard penetration is shown as an example in Table S7.2 for diesel and petrol cars, light and heavy-duty vehicles and busses.

The change over time in technology and the implementation of the standard measures is assumed in EDGARv4.3 to start in the year the directive comes into force, but the timing of the implementation is subject to large uncertainty as it could be preempted by, e.g., striving towards newer technologies in the case of the manufacturing industry or delayed by, e.g., the slow penetration of vehicles with new standards in the national fleet. In this work we do not aim to analyze when exactly the emission reductions effectively took place, but instead we take stock of the achievements achieved by 2010, by comparing the reference emissions in 2010 with the STAG\_TECH emissions in the hypothetical situation of 2010 at 1970 technology and with 1970 emissions standards.

<sup>3</sup>This is the technological default of fly ash not passing through the stack.

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The second STAG\_FUEL scenario was modeled by assuming that these three sectors consumed the same amount of each fuel type as in 1970. Since the fuel market is to a large extent global, this scenario was implemented in all countries for the three selected sectors. All power plants, vehicles and industries with the reference 2010 emissions standards consume coal, gas and oil as in 1970. This scenario reveals two effects: the increase in energy consumption and the change in fuel mix. The comparison of the reference emissions in 2010 with the hypothetical 2010 STAG\_FUEL situation highlights the extra emissions due to the further increase of the economy with energy-intensive activities, partially offset by shifting of fuel types in some countries.

The shifts towards cleaner fuels (e.g., from coal to gas) are contrasted with the fuel shares fixed at 1970 levels and this effect is discussed in more detail for EU. It concerns, e.g., the penetration of gas in the power sector, or of the biofuels, mixed in the fuels for vehicles. It should be noted that the only variable in the STAG\_FUEL scenario is the fuel itself. The impact of coal washing or technological fuel preparation is not part of the STAG\_FUEL scenario but as it addresses technology, it is part of the STAG\_TECH scenario. Therefore, the fuel quality directives show their emission savings (mainly in sulfur) partially in the STAG\_FUEL scenario and partially in the STAG\_TECH scenario.

## 2.2 Fast screening of impacts with TM5-FASST

The TM5-FASST model (Fast Scenario Screening Tool) is a linearized source–receptor model derived at a resolution of  $1^\circ \times 1^\circ$  in the main source regions from the global chemical transport model TM5-CTM (Tracer model 5, chemistry transport model) (Krol et al., 2005) for gaseous and particulate matter atmospheric pollutants. It considers 56 world regions with more detail over Europe. Detailed information on the FASST model can be found in dedicated works (Van Dingenen et al., 2009, 2015), while here we summarize its basic working principle and assumptions. The concentration of a substance  $x$  at time  $t$ , caused by the emission of a precursor  $l$  with source strength EM (emission) in source region  $S1, \dots, 56$  and received in receptor region  $R1, \dots, 56$  is calculated by the addition of a background concentration background (BG) and the contribution of

the linearized matrix function for each precursor (Eq. 2):

$$\begin{bmatrix} \text{CONC}_{R1} \\ \dots \\ \text{CONC}_{R56} \end{bmatrix} (t, x) = \begin{bmatrix} \text{BG}_{R1} \\ \dots \\ \text{BG}_{R56} \end{bmatrix} (t, x) + \sum_l \begin{bmatrix} \alpha_{R1S1}(t, x, l) & \dots & \alpha_{R56S56}(t, x, l) \\ \dots & \dots & \dots \\ \alpha_{R56S1}(t, x, l) & \dots & \alpha_{R56S56}(t, x, l) \end{bmatrix} \cdot \begin{bmatrix} \text{EM}_{S1}(t, l) \\ \dots \\ \text{EM}_{S56}(t, l) \end{bmatrix}, \quad (2)$$

where  $\alpha_{R1S1}, \dots, \alpha_{R56S56}$  are the source receptor coefficients for precursors  $l$  to substance  $x$ .

5 For each source region, the TM5-FASST model requires the annual emissions of  $\text{PM}_{2.5}$  to be specified<sup>4</sup> as BC, primary organic matter and other  $\text{PM}_{2.5}$ , and the precursors ( $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{CO}$ , NMVOCs and  $\text{NH}_3$ ) in order to estimate the corresponding  $\text{PM}_{2.5}$  and ozone concentrations in the receptor regions. Making use of source–receptor relationships, it converts the emissions from certain source regions to pollutant concentrations at the receptor regions, simulating meteorological and chemical processes. Only anthropogenic emissions are input to this model and the considered chemical reactions include the formation of secondary inorganic aerosol species (ammonium nitrate and sulfate) from gaseous precursors ( $\text{SO}_2$ ,  $\text{NO}_x$  and  $\text{NH}_3$ ), while no estimation of SOA (secondary organic aerosols) is performed. Secondary organic aerosol from biogenic sources is included in the reference simulation following the AEROCOM recommendations in Dentener et al. (2006), but not for anthropogenic SOA, for which no source–receptor relationships are calculated. Ozone formation is simulated through the reactions involving  $\text{CO}$ , VOCs and  $\text{NO}_x$ . Once  $\text{O}_3$  and  $\text{PM}_{2.5}$  concentrations, as well as its chemical composition, are simulated, the impacts of such concentrations on health and crops and vegetation are evaluated.

<sup>4</sup>For simplicity, “primary organic matter” is assumed to be  $1.3 \cdot \text{OC}$  emissions and for the “other  $\text{PM}_{2.5}$ ” the default of BC plus  $0.3 \cdot \text{OC}$  emissions is assumed.

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Tropospheric ozone and particulate matter negatively affect human health, increasing respiratory and cardiovascular diseases, lung cancer, etc. (WHO, 2013). Through parameterizations relating pollutant concentrations and exposed population (Anenberg et al., 2009; Jerrett et al., 2009; Burnett, 2014), TM5-FASST estimates the premature mortality for a population older than 30 years of age, exposed to O<sub>3</sub> and PM<sub>2.5</sub> concentrations. Moreover, ozone is a toxic compound for plants, reducing crop productivity (especially for wheat) and affecting plant diversity (UNEP/WMO, 2011). Following the procedure developed by Van Dingenen et al. (2009), the TM5-FASST model can quantify the loss yield due to crop exposure to O<sub>3</sub> for four types of crops (wheat, maize, rice and soy) at global and regional levels.

### 3 Emission scenarios results

We first compare the reference emission levels in 1970 and 2010 (REF(1970) and REF(2010)), and then the REF(2010) to the two retrospective scenarios (STAG\_TECH and STAG\_FUEL) at the global scale in Fig. 3. With the scenarios we focus on the three selected sectors: power industry (or “energy”), non-power industrial combustion of the manufacturing industry (or “industry”) and road transport (or “road”). We then evaluate the changes at the EU level in Fig. 4.

#### 3.1 Global emission trends

Figure 3 presents the air pollutants and aerosols trends at the global level. SO<sub>2</sub> emissions do not show a significant trend from 1970 (at 100 Tg) to 2010 (102 Tg), because the pre-combustion preparation of fuel (e.g., coal sulfur wash) and the post-combustion exhaust treatment with SO<sub>2</sub> emission reductions (e.g., by flue gas desulfurization (FGD) units) were counterbalanced by the increased use of fuel (in particular of coal) worldwide. The power sector contributed mostly in the 1970s to total sulfur emissions, and in the case of no technological progress for sulfur emission reductions

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taking place in the EU, global emissions would have been 1.4 times higher as shown by the STAG\_TECH scenario. However, if we consumed with current technology the same amount of fuel as in 1970 for the power, industry and road transport sectors, global SO<sub>2</sub> emissions would be only two-thirds of the 2010 emissions (STAG\_FUEL).

5 For the other pollutants, like NO<sub>x</sub>, CO and NMVOCs, we see a significant increase from 1970 to 2010, which reflects mainly the increase in fuel consumption in the energy, industry and road sectors (e.g., NO<sub>x</sub> emissions from power generation tripled globally, from 10.5 to 31.4 Tg). If the same amount of fuel had been used in these three sectors (STAG\_FUEL), emissions would have remained constant over the 4 decades, as shown  
10 in the STAG\_FUEL scenario. For these air pollutants, the largest emission reduction is obtained in the road transport sector. At the global scale, road emissions increased by a factor of ca. 4 (except for NH<sub>3</sub>) due to the increased traffic volumes in many industrialized and emerging countries (see also to the STAG\_FUEL to REF(2010) ratio in Fig. 3b), which has only a limited share in the total. When comparing the STAG\_TECH  
15 to the REF(2010) case, it is found that road emissions were globally reduced by 88 % for SO<sub>2</sub> and 60 % for NO<sub>x</sub>, CO and NMVOCs and 50 % for PM, which clearly illustrates the development of more efficient vehicles and introduction of more stringent abatement measures. A shift from diesel to petrol could have caused another reduction but the opposite took place: diesel vehicles represented only ca. 20 % of the global fleet  
20 in 1970 compared to ca. 75 % in 2010. Other sectors, such as residential combustion, also make important contributions to total emission but is shared among many more individuals. For the aerosols (PM<sub>10</sub>, PM<sub>2.5</sub> and BC) the increase in emission level from 1970 to 2010 is even stronger, although the three selected sectors are not amongst the largest contributors (a major contribution comes from residential combustion but it was  
25 not included in our scenarios). However, reductions of more than 99 % are obtained for road, 60 % for industry and 40 % for energy.

More information on the ratios between each retrospective scenario and the reference case are given in Tables S1.1 and S1.2. In the subsections below, we focus on the three sectors separately for the historical trend, STAG\_FUEL and STAG\_TECH cases.

## 3.2 European emission trends

### 3.2.1 EU power industry (“energy”)

Figure S4.1 in the Supplement presents the evolution over the years 1970–2010 of SO<sub>2</sub> and PM<sub>10</sub> power plant emissions in Europe, highlighting the role played over time in actual emission levels (blue area) by the introduction of abatement measures (EOP, red area), and by the change in emission factors and technology (green area). The concurrent effects due to the change in fuel quality (directive 98/70/EC, 1998, as well as international conventions like CLRTAP and GP) and the introduction of abatement measures (non-regenerative dry/semidry and wet FGD), following the directive regulating emissions from large combustion plants (2001/80/EC), determined the actual 2010 emission levels. Concerning particulate matter (here represented by PM<sub>10</sub>, but the same results apply also to PM<sub>2.5</sub> and its carbonaceous components), in 1970 power plants were already equipped with some abatement measures (e.g., cyclones), which became more and more used especially in the nineties (using finer filters) not only due to law restrictions but also to increase their efficiency (decreasing the variable cost of fuel input) and robustness (protect delicate components from corrosive air pollutant gases and to shorten outages). Therefore, the major reduction in PM<sub>10</sub> emissions is due to the application of abatement measures.

In our STAG\_TECH scenario we only take into account the European regulations on power plants in addition to the complete stagnation of technologies at the global level, although emerging countries like China and India have been implementing their own policies just in the last decade to regulate air pollutant emissions and improve air quality. For the power generation sector, the impact of European legislation can be seen only in the European context, but this does not mean that other countries have not progressed in the development of air quality regulations. This represents the effect of “policies outside of Europe” which are not taken into account here.

Figure 4 presents a closeup on the EU, and shows clearly that the emissions decreased from 1970 to 2010 for all air pollutants and aerosols. This reduction was ob-

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tained through the introduction of end-of-pipe measures primarily, but not exclusively. For the STAG\_FUEL scenario, which uses the fuel consumption of 1970 but the current emission reduction measures one would expect a lower value than the REF(2010) case, as observed globally. For SO<sub>2</sub> and PM<sub>10</sub>, the STAG\_FUEL case results in a 15 and 8 % higher value even though one-third less energy is consumed with the current technology and emission reduction measures. This is due to the fuel mix, which contains in 1970 a significant share of heavy residual fuel oil and coal (both rich in sulfur), and was partially substituted with cleaner fuels, to a large extent natural gas, from 1970 to 2010 as illustrated in Fig. 5. In 2010, coal-related fuels represented ca. 49 % of EU fuel consumption, compared to 65 % in 1970, while the oil contribution decreased from 25.5 to 9 %. The EU phased out the use of heavy residual fuel oil in the energy sector. The EU energy sector increased its fuel consumption by adding natural gas, in particular in UK, Sweden, and Spain (IEA, 2014). Moreover Poland and Germany increased the share of lignite over bituminous coal in their power industry (IEA, 2014), leading to lower BC emissions. Contrastingly, we observe a higher share of clean S fuels, such as natural gas and wood, in 2010 than in 1970 (35 vs. 9 % and 7 vs. 0.5 %, respectively). As such, SO<sub>2</sub> emissions from power plants were drastically reduced in Europe from 1970 to 2010 due to the shift to cleaner fuels and due to technological treatments of both fuels (to lower sulfur content) and flue gas (to lower SO<sub>2</sub> emission) following EU policies, consistent with the analysis of Rafaj et al. (2013).

At the European level, the STAG\_TECH scenarios produced 50 % higher NO<sub>x</sub> emissions compared to the REF(2010) when assuming technology stagnation and not yet optimized combustion processes (lower efficiency, lower air–fuel mix) (see Table S1.2 and Fig. 4b). Interestingly, NH<sub>3</sub> emissions are higher in the STAG\_TECH scenario compared to the reference case due to the fuel shift from oil to gas which emits much less NH<sub>3</sub> than the oil. European legislation has been also successful in effectively abating PM<sub>10</sub> emissions from power plants, reducing them by a factor of more than 3 compared to the STAG\_TECH scenario. Similar conclusions can be drawn for PM<sub>2.5</sub> emissions and its components (BC and OC).



### 3.2.2 EU manufacturing industry (“industry”)

The primary emissions of industrial activities, including all manufacturing activities, are SO<sub>2</sub>, NO<sub>x</sub>, CO and PM. NMVOC emissions are to a large extent due to the use of solvents and specific chemical processes. When comparing STAG\_FUEL to the REF(2010) emissions for Europe, we observe, contrary to global-scale emissions doubling, a decrease of a factor of 1.5–4 depending on the pollutant (see Fig. 4a and Table S1.1). The emissions from the manufacturing industry were affected by the shift to cleaner fuels from 1970 to 2010 (see Fig. 5) and, in particular, there was a considerable reduction in the use of heavy residual fuel oil. From 1970 to 2010 the relative fuel usage in manufacturing industries changed from 26.9 to 16.9 % for coal, from 18.8 to 52.4 % for gas, from 53.6 to 18.9 % for oil and from 0.7 to 11.9 % for wood. More details about non-EU countries are discussed in Sect. S3.

The impact of technological development and deployment of pollutant abatement measures on the industrial sector is depicted in Fig. 4b. When comparing the STAG\_TECH and REF(2010) cases, we find that only emissions from SO<sub>2</sub>, CO, NH<sub>3</sub> and PM components are slightly affected in Europe with ratios ranging from 1.2 to 2.6, except for NH<sub>3</sub> where the ratio is 3.9 due to higher emissions from oil combustion than from gas. Therefore, even in Europe, reference emissions from the manufacturing industry sector are generally higher in absolute terms compared to those from the power sector (see Fig. 4) because of the deployment of less clean fuels and less efficient technologies, as well as the lack of stringent effective abatement measures. Just recently, the European legislature introduced the 2010/75/EU directive to regulate emissions from industrial activities; therefore, the implementation of these new standards and reduction measures will require some years before we are able to quantify the impact of this policy on industrial emissions.

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### 3.2.3 EU road transport (“road”)

The largest effects of technology and end-of-pipe control measures is observed in the road sector in the EU. The fuel quality directive reduced SO<sub>2</sub> emissions by 2 orders of magnitude. With the optimization of combustion technology (motor inside flow and combustion, common rail fuel injection, preheating) the CO and NMVOC emissions have also been reduced. With the adoption of EURO standards, particulate filters have penetrated national car fleets, and PM at the exhaust has been reduced by more than a factor of 4. The efficiency of the particulate filters is much higher, but emissions become more determined by a relatively small fraction of vehicles with lower emissions standards or even those super-emitting. Moreover, particulate filters do not work when the S content in the fuel is larger than 50 ppm. Furthermore, EURO standards reduced NO<sub>x</sub> emissions, at the expense of increasing NH<sub>3</sub> emissions (which is the only substance that is increased in emission under the STAG\_TECH scenario). European NO<sub>x</sub> and BC increased in 2010 by a factor of 3.2 and 5.2, respectively, compared to the STAG\_FUEL scenario (see Fig. 4), not only due to the increased fuel consumption but also to the shift from petrol to diesel, thus emitting more NO<sub>x</sub> and particulate matter, for passenger cars in the EU, as depicted in Fig. 6.

Figure S4.2 shows the change in road transport emissions over time for SO<sub>2</sub> and PM<sub>2.5</sub> in Europe. Already in the 1970s, Europe was moving towards the use of cleaner fuels, strengthened by the introduction of the CLRTAP conventions and GP, thus reducing SO<sub>2</sub> road emissions. In 1999 the European Union directive 1999/32/EC required the improvement of petrol and diesel fuel quality, lowering their S content. On the other hand, the deployment of cleaner fuels did not reduce primary particulate matter emissions (e.g., PM<sub>2.5</sub> as shown in Fig. S4.2); however, only with the gradual introduction in the 1990s of EURO standards for vehicles (from EURO1 in 1992 to EURO5 in 2009 (see Table S7.2)), imposing the application of particle filters, were PM road transport emissions abated. This exemplifies the policy response to different types of pollutants and sources through the implementation of new policies.

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As shown in Fig. 4b, comparing REF(2010) vs. STAG\_TECH, the reduction in the sulfur fuel content reduced SO<sub>2</sub> emissions in the EU by a factor of 160. Lower SO<sub>2</sub> emissions in 2010 are mainly associated with the implementation of EU fuel quality directives and international conventions, the shift to cleaner fuels, and much less by the presence of EOP measures. Low ratios of REF(2010) to STAG\_TECH data are observed for Europe, ranging from 0.41 and 0.17 for NO<sub>x</sub> and CO, respectively, due to the advancement in combustion technology for diesel cars highly deployed in Europe. NH<sub>3</sub> emissions increased with the implementation of catalysts on gasoline vehicles, leading to a decrease of 70 % in NH<sub>3</sub> European emissions when catalysts are not considered (Fig. 4b, STAG\_TECH scenario). The effectiveness of abatement measures can be quantified looking at PM emissions, which decreased by a factor of 4–5, owing to the application of particulate filters in Europe. Figure 7 reports road transport PM<sub>2.5</sub> emissions for the year 2010 and for the STAG\_TECH scenario for world regions (details about region classifications can be found in Sect. S6.4). A decrease of ca. 50 % of PM<sub>2.5</sub> road emissions (0.91 Tg) is observed globally due to the implementation of the EURO standards on vehicles. This reduction is almost equally attributed to the impact of EU standards in Europe (0.47 Tg) as well as outside of the EU (0.44 Tg). Major impact of EURO standards outside Europe is found in China, Southeast Asia, India, the Middle East, Indonesia, Japan, Oceania, etc., while a smaller impact is seen in North America due to the deployment of standards not affected by the STAG\_TECH scenario (UT1, UT2, UT3, PH1 and PH2). Further analysis about the spillover of the EURO standards outside Europe are presented in Crippa et al. (2015).

### 3.3 European hotspots: avoided emissions for the year 2010

Figure 8 shows the spatial distribution in Europe (grid maps with 0.1 × 0.1° resolution) of the difference in emissions of the STAG\_TECH and REF scenarios for selected pollutants (SO<sub>2</sub>, NO<sub>x</sub>, CO, PM<sub>10</sub> and BC). These maps can be interpreted as the avoided emissions in Europe due to the implementation of European legislation on the power generation industry and road transport, together with the change in emission factors

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and technologies which also affected the industrial sector. The avoided SO<sub>2</sub> emissions are mainly located in western European urban areas (e.g., Paris, Madrid, London, Rome, Berlin and the Benelux region) due to the co-location of several emission activities while many point sources (power plants and industries) are spread over Europe representing the reduction in SO<sub>2</sub> emissions due to the switch to cleaner fuels (shifting of fuel types and lower sulfur content). A different spatial distribution of the avoided emissions is found for NO<sub>x</sub> and CO, where in addition to urban areas, a big reduction is observed for road transport (road tracks are visible for both pollutants). Interestingly, Italy, Germany, the United Kingdom and the Benelux region have strongly reduced CO emissions more uniformly compared to other European regions (e.g., France and Spain). PM<sub>10</sub> and BC grid maps highlight the effectiveness of EURO standards on road vehicles especially in western European countries, representing a successful example to be followed by eastern European regions. Finally, the implementation of particulate filters on power plants and industries was also effective in very industrialized areas (e.g., Benelux) and other major conurbations.

## 4 Corresponding impacts on air quality, health and crops

### 4.1 Concentration and composition changes

Energy, industry and road emissions data from the considered scenarios have been used in the TM5-FASST model to derive the corresponding PM<sub>2.5</sub> and O<sub>3</sub> concentrations for main world regions. As shown in Fig. 9, we first compare the impact of the emissions changes from 1970 to 2010 (REF(1970) vs. REF(2010)) on PM<sub>2.5</sub> and O<sub>3</sub> concentrations, and then the differences of the other two scenarios (STAG\_FUEL and STAG\_TECH) with 2010 reference data. Delta concentrations are calculated as the difference from each scenario and the REF\_2010 values, so a positive delta means that concentrations for a certain scenario were higher than the reference 2010 data.

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PM<sub>2.5</sub> concentrations decreased from 1970 to 2010 by 6.1 and 9.7  $\mu\text{g m}^{-3}$  in North America and Europe, respectively, while increased concentrations in 2010 are especially observed for Asian countries (35.3  $\mu\text{g m}^{-3}$ ), Africa (2.3  $\mu\text{g m}^{-3}$ ) and Latin America (2  $\mu\text{g m}^{-3}$ ). A similar pattern is also observed for O<sub>3</sub> – industrialized countries had higher concentrations in 1970 compared to 2010 (delta O<sub>3</sub> equal to 4 ppb for North America and to 1.2 ppb for Europe), while developing countries reached higher concentrations in 2010 (39.7, 20.8, 18.7 and 11.3 ppb for Asia, Latin America, Russia and Africa, respectively). Similarly to the trend observed from 1970 to 2010, comparing the STAG\_FUEL scenario to the REF(2010) case, the population weighted PM<sub>2.5</sub> and O<sub>3</sub> concentrations show opposite patterns for industrialized and emerging countries; however, the largest impact is observed for Asia where the stagnation of fuel consumption and mixture at 1970 levels would have produced much lower concentrations (delta PM<sub>2.5</sub> = 39.9  $\mu\text{g m}^{-3}$  and delta O<sub>3</sub> = 18.4 ppb) compared to real 2010 levels. A completely different pattern is observed for the STAG\_TECH vs. REF(2010) scenarios since the stagnation of technologies at 1970 levels applied to present-day consumption would have produced enhanced emissions for all world regions, especially for Europe (delta PM<sub>2.5</sub> = 9.5  $\mu\text{g m}^{-3}$  and delta O<sub>3</sub> = 5.3 ppb), Asia (delta PM<sub>2.5</sub> = 10.1  $\mu\text{g m}^{-3}$  and delta O<sub>3</sub> = 8 ppb) and Russia (delta PM<sub>2.5</sub> = 5.6  $\mu\text{g m}^{-3}$  and delta O<sub>3</sub> = 15.7 ppb). As shown in more detail in Fig. S8.1, the implementation of EU air quality legislation for industrial facilities, in particular power plants and of the EURO standards for road vehicles, coupled with the change in technology and fuel quality, led to on average 4–5  $\mu\text{g m}^{-3}$  lower concentrations in Europe as PM<sub>2.5</sub> (including both the primary and secondary particulate components simulated by TM5-FASST). Power plant PM<sub>2.5</sub> concentrations decreased by 1.9 and 2.9  $\mu\text{g m}^{-3}$  in western and central Europe (with a more coal-fired power industry). However, effects of power plant emission reductions (STAG\_TECH scenario) in other regions (Japan, China, USA, India, etc. with delta PM<sub>2.5</sub> values between 1.5 and 3.7  $\mu\text{g m}^{-3}$ ) are significant. For Japan and China, large contribution in concentration reduction is seen from the road sector (1.3 and 1.1  $\mu\text{g m}^{-3}$  respectively), and are related to an export of EURO standards via market globalization. The impacts of this

specific sector is studied in more detail in a separate publication by Crippa et al. (2015). For the USA, the large reduction in PM<sub>2.5</sub> concentration is due to the energy sector. Although the end-of-pipe measure implementation on US power plants is ascribed to US air quality legislation (and not analyzed in the STAG\_TECH scenario), both Europe and the USA profited equally from fuel quality improvement.

Looking into the detailed chemical composition of the PM<sub>2.5</sub> changes for the energy, road transport and industrial sectors (Fig. S8.2), we gain further insights into the sectors and processes that contributed to the concentration reductions. Power plant emissions typically consist of aerosol precursor gases and particulate matter (also including fly ash); therefore, its delta PM<sub>2.5</sub> chemical composition is mainly formed by secondary inorganic components (nitrates, sulfates and ammonium) as well as other PM<sub>2.5</sub>. Similar aerosol chemical composition changes are found for industrial sources with less secondary particulate sulfates as heavy residual fuel oil is phased out. A different PM<sub>2.5</sub> chemical composition response is found for road transport, which consists of primary organic matter, BC, as well as ammonium nitrate particles formed by the chemical reaction of NO<sub>x</sub> and NH<sub>3</sub> emitted by this sector. The delta particulate SO<sub>4</sub> mainly represents the impact of the change in the sulfur fuel content in worldwide regions due to the implementation of EU fuel quality directives as well as international conventions and globally it corresponds to ca. 0.5 μg m<sup>-3</sup> less for the year 2010 on average (1.5 μg m<sup>-3</sup> for USA, 1.3 μg m<sup>-3</sup> for China, 1.1 μg m<sup>-3</sup> for central Europe and 0.9 μg m<sup>-3</sup> for western Europe).

While the impact on PM<sub>2.5</sub> concentrations due to technology and emission reductions (STAG\_TECH scenario) are mostly found in the source region with emission change (Europe, Japan, China), longer-range effects are found for ozone. This is an important result because it represents the need of having continental-scale policies for some pollutants. O<sub>3</sub> formation is driven by the reaction of the precursors NO<sub>x</sub>, CO and NMVOCs, derived mostly from the road sector and strongly abated over the past two decades with the EURO standards. The avoided annual and regional average O<sub>3</sub> concentrations ranges between 0.5 and 6 ppb, which significantly affects current O<sub>3</sub> levels

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ranging from 30 to 50 ppb and which are most present in the hot arid regions of North Africa, the Middle East and Turkey.

## 4.2 Health and crop impacts of improved air quality

Modeled delta  $PM_{2.5}$  and  $O_3$  concentrations are used in TM5-FASST to estimate the impact of the specific scenario assumptions on human health and crop yield (see Fig. 9). An increase of  $PM_{2.5}$  is reflected by a reduction of life expectancy, since enhanced PM concentrations are the main harmful components impacting human health. So, comparing the 1970 situation with the 2010 one, globally a loss in life expectancy is observed mainly for North America (6 months) and Europe (9.2 months), while increased life expectancy is found for developing countries (14.7 months for Asia). Moreover, considering the STAG\_FUEL case, enhanced life expectancy would have been reached by most of the world's regions (17.6 months for Asia, 4.6 months for Russia, 4.7 months for Africa and 3.4 months for Latin America), while the opposite situation is observed for the STAG\_TECH vs. REF(2010) scenarios.

As expected, significant impact on life expectancy is observed for western and central European countries (4–5 months, see also Fig. S8.3), where the impact of emission reduction measures is largest. Life expectancy increases also in other industrialized countries like Japan and USA (gaining 3.5 and 2 months, respectively, in life expectancy) and only to minor extent in developing and emerging countries. Therefore, major health benefits from emission reduction measures are observed in highly populated areas where PM and ozone changes are large.

$O_3$  concentrations negatively influence crop growth, so the reduction in  $O_3$  concentrations observed for the REF(1970) vs. REF(2010) and STAG\_FUEL vs. REF(2010) is reflected in a net positive crop yield (global gain equal to 15.5 and 31.1 million Mt for the two scenarios, respectively). Conversely, as shown in Fig. 9 and more in detail in Fig. S8.3, the emission control measures on vehicles were mostly responsible for mitigating impacts on crop yields. The introduction of vehicle EURO standards led to reduction of worldwide ozone levels due to its atmospheric transport, corresponding to

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a crop yield benefit up to 8.3 million Mt of avoided crop loss, representing 0.3 % of world production of maize, wheat, rice and soy. Specifically, the reduction of road transport emissions allowed the production of an additional 2 Mt of crops in China in 2010, 1.4 in western Europe, 1 in India, etc.

## 5 Conclusions

The interplay of European air quality policies and technological advancement to reduce anthropogenic emissions in Europe and the world over the last 40 years has been investigated. This period encompasses from 1970, when the first European air quality directive was introduced until 2010, the last year with reliable statistical data availability. We mainly focus on two retrospective scenarios for 1970–2010 in order to analyze separately the impact of concurrent factors on today's emission levels, like the change in fuel consumption and fuel composition (STAG\_FUEL), the implementation of abatement measures, the improvement in fuel quality and the shift to cleaner fuels and the change in technologies (STAG\_TECH). The scenarios allow us to assess the role and impact of EU legislation on air quality, of technology development and of fuel use. These scenarios can also serve as an example for designing efficient air pollution abatement policies in emerging economies. Here we focus on the most relevant pollutant ( $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{CO}$ , NMVOCs and  $\text{NH}_3$ ) emissions, and particulate matter ( $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , BC, OC) affecting air quality at global and European levels. At the global scale, emissions of most components stabilized or increased, due to the increased growth of activities in particular of countries with emerging economies, despite emission control measures such as implemented in Europe (comparing 1970 and 2010, we note that global emissions of some pollutants increased by up to ca. 80 %) and other industrialized countries. For example, European  $\text{SO}_2$  emissions were reduced by 80 % from 1970 to 2010 while there was almost no change at the global level. Looking at the European situation, we identify the relevant factors leading to the 2010 emission levels. For the power and manufacturing industry sectors, the increased fuel consump-

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tion was coupled with a shift towards cleaner fuels (from coal-related fuels to gas), and supported by the implementation of fuel quality directives, regulating the sulfur fuel content as well as end-of-pipe emission control measures under, e.g., large combustion plant directives. Despite a strong increase in traffic, the overall emissions were strongly reduced due to the implementation of abatement measures following the EURO standards for vehicles (especially for NO<sub>x</sub>, CO, NMVOCs and PM), the use of cleaner fuels (with lower S content) and the shift from petrol to diesel passenger cars (emitting more particles but less CO). Therefore, from our study we have determined that a variety of EU air quality policies since 1970 have avoided a dramatic deterioration of air quality in Europe and beyond. For example, the EURO norms for vehicles, and fuel quality directives (sulfur), were among the most influential policies impacting air quality globally (e.g., 88% reduction of SO<sub>2</sub> and 50% reduction of PM<sub>2.5</sub> from global road transport exhaust emissions).

To complete the assessment, the TM5-FASST model was used to estimate the impact on PM<sub>2.5</sub> and O<sub>3</sub> concentrations, human health and crop production of the considered scenarios compared to the reference case. PM<sub>2.5</sub> concentrations were reduced by 4.5 and 5 μg m<sup>-3</sup> in central and western Europe respectively, as well as in Japan, China, USA, India (range 1.5–3.7 μg m<sup>-3</sup>); moreover, ozone concentrations were reduced by 3–9 ppb in several world regions also due to atmospheric transport. We estimate that EU policies increased life expectancy of people not only in Europe, but also in Japan and the USA by several months (e.g., 5 months in Europe and 3.5 months in Japan). In addition, the introduction of EURO standards led to the reduction of worldwide ozone levels, with up to 8.3 million Mt avoided crop loss, which corresponds to 0.3% of the present world production of maize, wheat, rice and soy.

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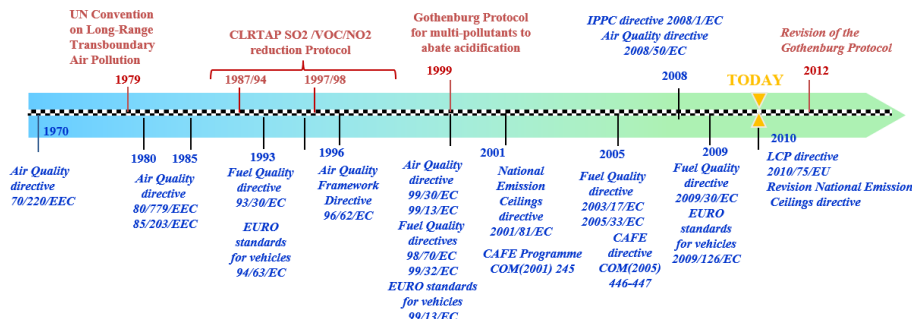
**Table 1.** Emission scenario assumptions.

Emission sectors	Reference 1970 and 2010	Scenario 1 in 2010: stagnation of fuel consumption (STAG_FUEL)	Scenario 2 in 2010: stagnation of technology and no end-of-pipe control (STAG_TECH)
Agricultural waste burning	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Energy industry	EDGAR v4.3	AD(1970) · EF(2010) · (1-RED(2010))	AD(2010) · EF(1970) · (1-RED(1970))
Solid waste disposal	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Combustion in manufacturing industry	EDGAR v4.3	AD(1970) · EF(2010) · (1-RED(2010))	AD(2010) · EF(1970) · (1-RED(1970))
Industrial processes and product use	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Oil production and refining	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Buildings (residential and others)	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Fossil fuel fires	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Road transportation	EDGAR v4.3	AD(1970) · EF(2010) · (1-RED(2010))	AD(2010) · EF(1970) · (1-RED(1970))
Aviation (international + domestic)	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Shipping (international + domestic)	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3
Non-road ground transport	EDGAR v4.3	EDGAR v4.3	EDGAR v4.3



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**Figure 1.** Overview of historical European Union (in blue) and international (in red) air quality regulations. UNECE/CLTRAP cover all European countries, USA, Canada, Belarus, Russia, Turkey, Israel, Ukraine and central Asian states.

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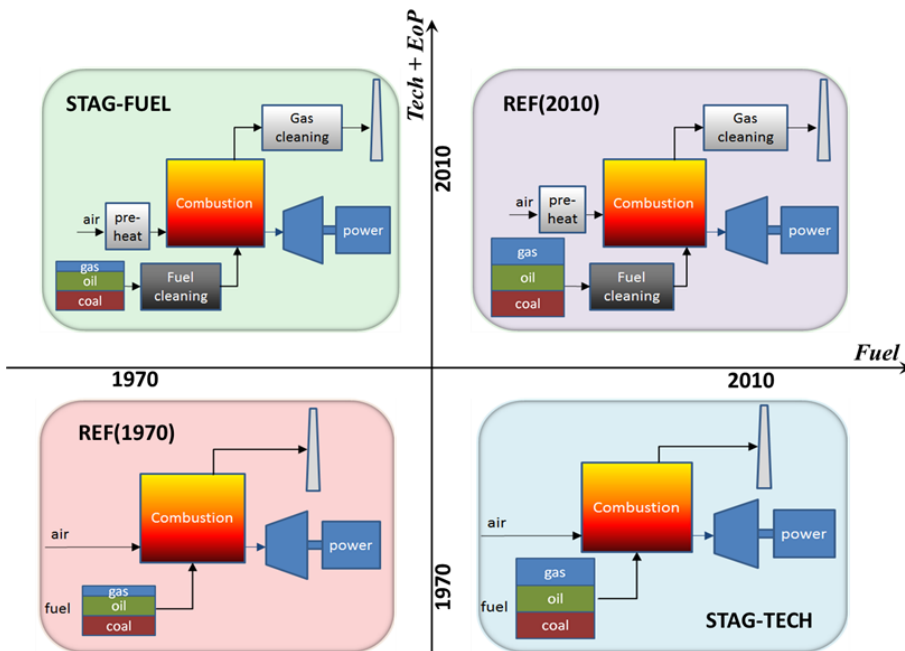
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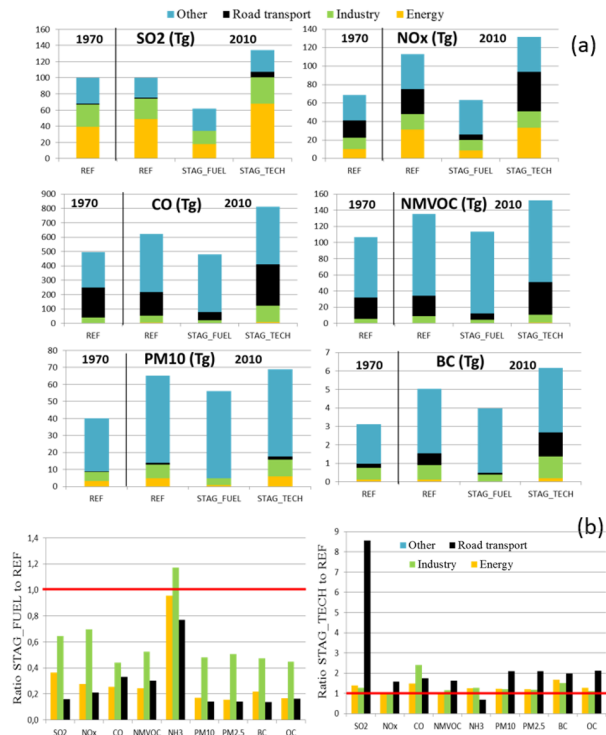
**Figure 2.** Schematic of the considered emission scenarios: REF\_1970, REF\_2010, STAG\_FUEL and STAG\_TECH.

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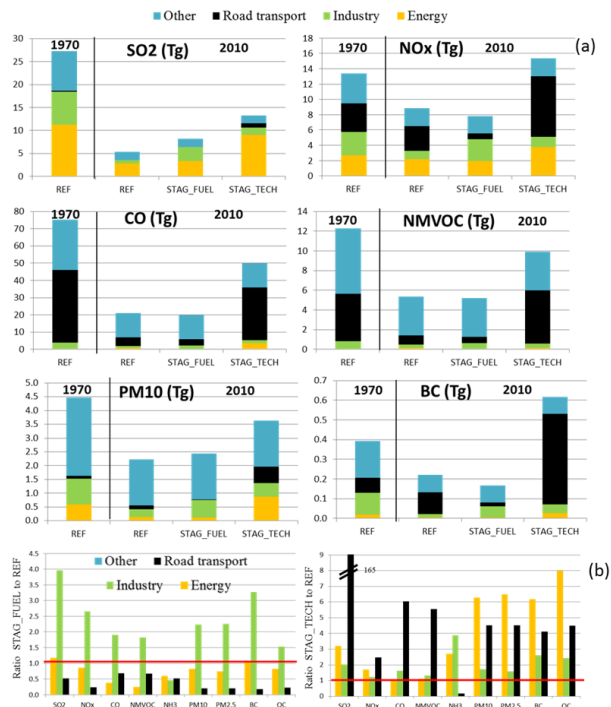
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**Figure 3.** Overview of 2010 emissions for REF (2010), STAG\_TECH (2010) and STAG\_FUEL (2010), as well as of the real emissions in 1970 (REF, 1970) (a) at the global scale. The main anthropogenic emission sectors are “energy” stands for power industry, “industry” stands for manufacturing industry, “road transport”, “other” stands for all other anthropogenic emission sectors such as residential, agriculture, fuel transformation sector, refineries and waste disposal. In (b), the ratios STAG\_FUEL to REF and STAG\_TECH to REF are presented. A ratio of 1 (red line in the graph) means no change between the reference emissions and the scenarios, while values lower than 1 indicate a decrease in today’s emissions would scenario have happened, whereas values higher than 1 indicate the opposite situation).

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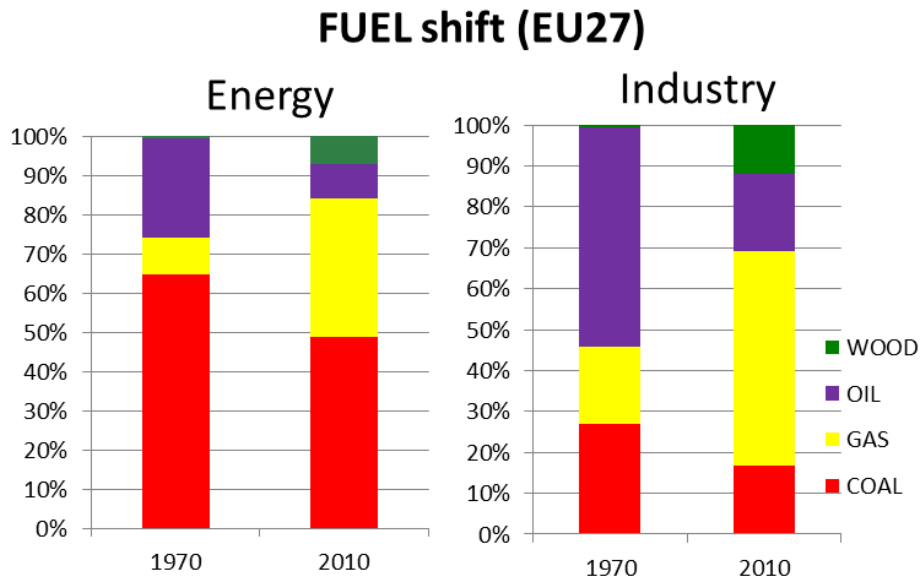
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**Figure 4.** Overview of 2010 emissions for REF (2010), STAG\_TECH (2010) and STAG\_FUEL (2010), as well as of the real emissions in 1970 (REF, 1970) (a) at the European (EU27) scale. The main anthropogenic emission sectors are (“energy” stands for power industry, “industry” stands for manufacturing industry, “road transport”, “other” stands for all other anthropogenic emission sectors such as residential, agriculture, fuel transformation sector, refineries and waste disposal. In (b), the ratios STAG\_FUEL to REF and STAG\_TECH to REF are presented. A ratio of 1 (red line in the graph) means no change between the reference emissions and the scenarios, while values lower than 1 indicate a decrease in today’s emissions would scenario have happened, whereas values higher than 1 indicate the opposite situation).

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**Figure 5.** Caloric fuel composition of (a) the energy and (b) industrial sectors (terajoule per terajoule) in the EU27 from 1970 to 2010. A large shifting of fuel types occurred in the power generation and manufacturing industry sectors. Percentages are expressed as terajoule per terajoule.

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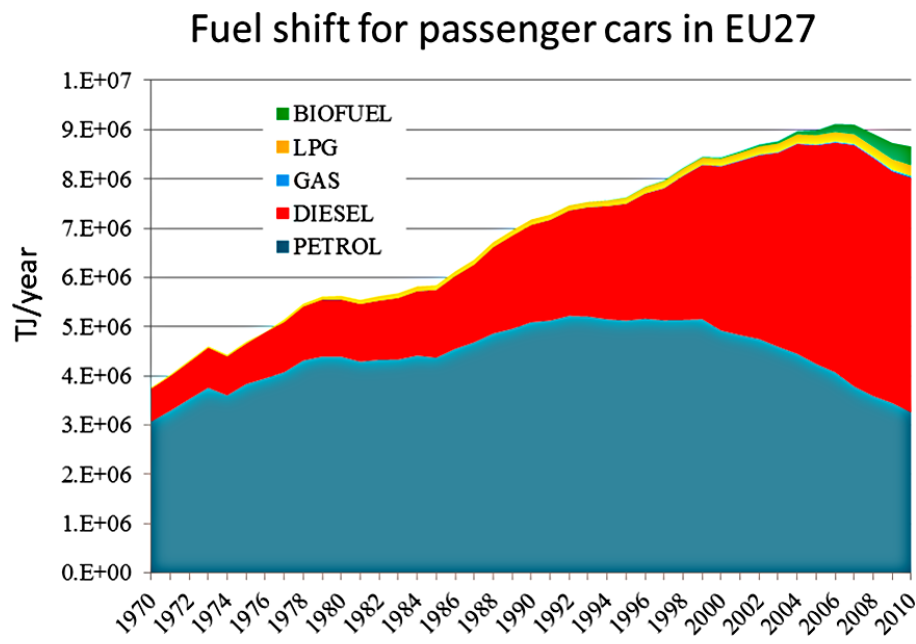
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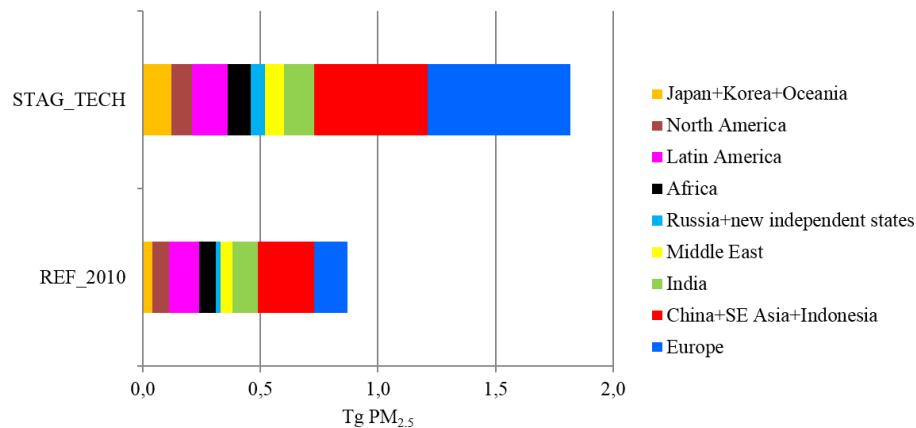


**Figure 6.** Fuel composition ( $\text{TJyr}^{-1}$ ) change between 1970 and 2010 in the EU27 passenger cars (note the large shift from petrol to diesel).



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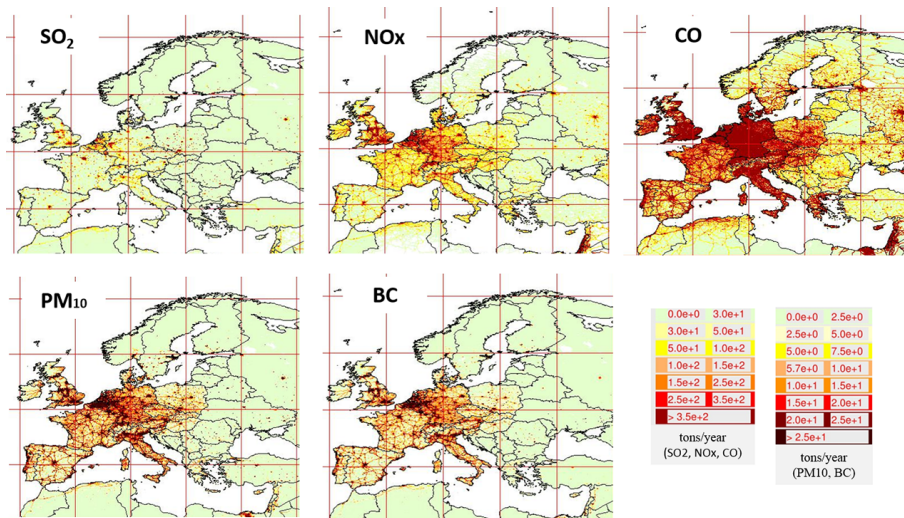


**Figure 7.** Impact of European legislation and technological stagnation (STAG\_TECH) on PM<sub>2.5</sub> road transport emissions inside and outside the European (EU27) region. The effect of this scenario can be observed by the difference of the STAG\_TECH to the REF bars.

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**Figure 8.** Hotspots of avoided emissions due to progressive implementation of in Europe: the difference of STAG\_TECH and REF emissions in 2010 ( $\text{t year}^{-1}$  ( $0.1^\circ \times 0.1^\circ$  gridcell) $^{-1}$ ).

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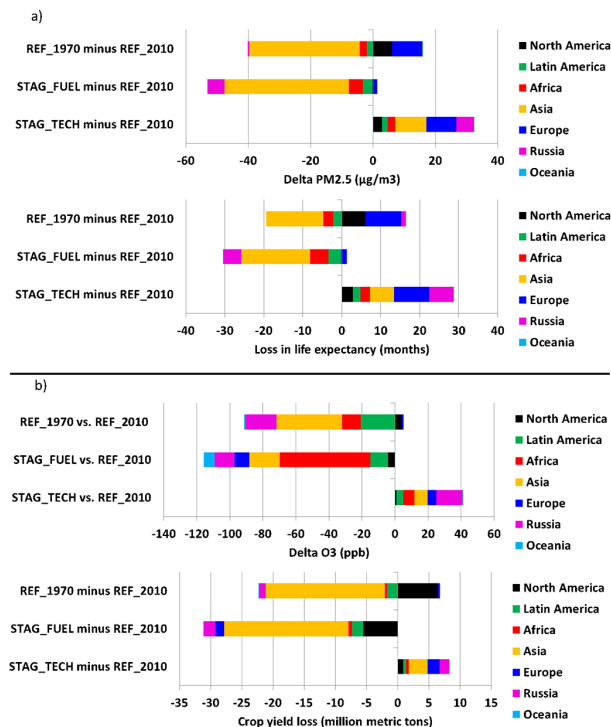
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**Figure 9.** Increase in PM<sub>2.5</sub> and in O<sub>3</sub> concentrations (delta PM<sub>2.5</sub> and delta O<sub>3</sub>) compared to reference 2010 values for each of the three situations: the reference case of 1970, the stagnation in fuels and the stagnation in technology (REF\_1970 minus REF\_2010, STAG\_FUEL minus REF\_2010, STAG\_TECH minus REF\_2010) and the corresponding life expectancy and crop yield losses for major world regions. **(a)** represents delta PM<sub>2.5</sub> concentrations for the three scenarios (top part of the graph) while the corresponding health impacts are reported at the bottom of **(a)**. **(b)** represents delta O<sub>3</sub> concentrations for the three scenarios (top part of the graph) while the corresponding crop yield loss is reported at the bottom of **(b)**.

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