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## Forty years of improvements in European air quality: regional policy-industry interactions with global impacts

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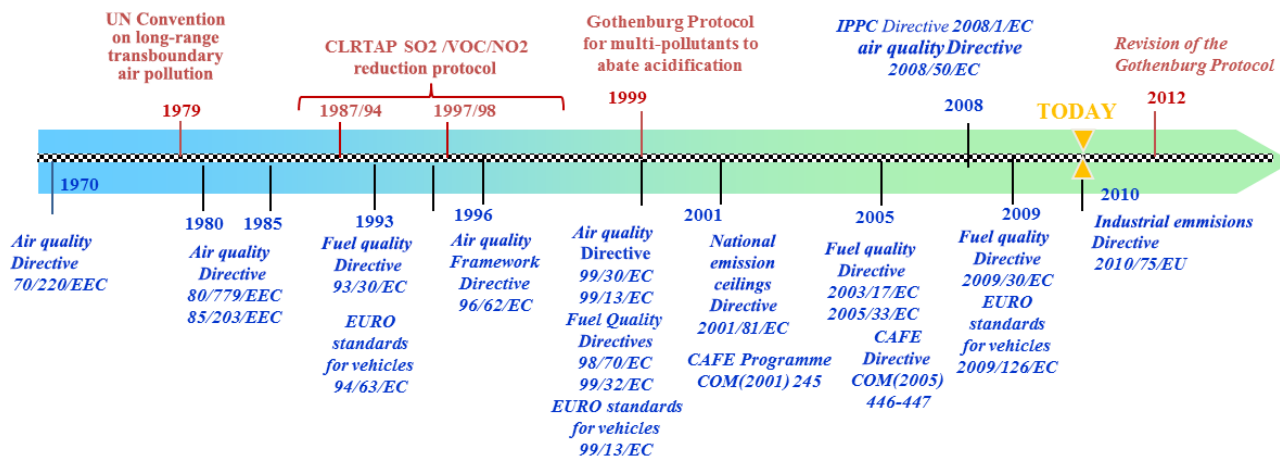
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**Abstract.** The EDGARv4.3.1 (Emissions Database for Global Atmospheric Research) global anthropogenic emissions inventory of gaseous ( $\text{SO}_2$ ,  $\text{NO}_x$ , CO, non-methane volatile organic compounds and  $\text{NH}_3$ ) and particulate ( $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , black and organic carbon) air pollutants for the period 1970–2010 is used to develop retrospective air pollution emissions scenarios to quantify the roles and contributions of changes in energy consumption and efficiency, technology progress and end-of-pipe emission reduction measures and their resulting impact on health and crop yields at European and global scale. The reference EDGARv4.3.1 emissions include observed and reported changes in activity data, fuel consumption and air pollution abatement technologies over the past 4 decades, combined with Tier 1 and region-specific Tier 2 emission factors. Two further retrospective scenarios assess the interplay of policy and industry. The highest emission STAG\_Tech scenario assesses the impact of the technology and end-of-pipe reduction measures in the European Union, by considering historical fuel consumption, along with a stagnation of technology with constant emission factors since 1970, and assuming no further abatement measures and improvement imposed by European emission standards. The lowest emission STAG\_Energy scenario evaluates the impact of increased fuel consumption by considering unchanged energy consumption since the year 1970, but assuming the technological development, end-

of-pipe reductions, fuel mix and energy efficiency of 2010. Our scenario analysis focuses on the three most important and most regulated sectors (power generation, manufacturing industry and road transport), which are subject to multi-pollutant European Union Air Quality regulations. Stagnation of technology and air pollution reduction measures at 1970 levels would have led to 129 % (or factor 2.3) higher  $\text{SO}_2$ , 71 % higher  $\text{NO}_x$  and 69 % higher  $\text{PM}_{2.5}$  emissions in Europe (EU27), demonstrating the large role that technology has played in reducing emissions in 2010. However, stagnation of energy consumption at 1970 levels, but with 2010 fuel mix and energy efficiency, and assuming current (year 2010) technology and emission control standards, would have lowered today's  $\text{NO}_x$  emissions by ca. 38 %,  $\text{SO}_2$  by 50 % and  $\text{PM}_{2.5}$  by 12 % in Europe. A reduced-form chemical transport model is applied to calculate regional and global levels of aerosol and ozone concentrations and to assess the associated impact of air quality improvements on human health and crop yield loss, showing substantial impacts of EU technologies and standards inside as well as outside Europe. We assess that the interplay of policy and technological advance in Europe had substantial benefits in Europe, but also led to an important improvement of particulate matter air quality in other parts of the world.



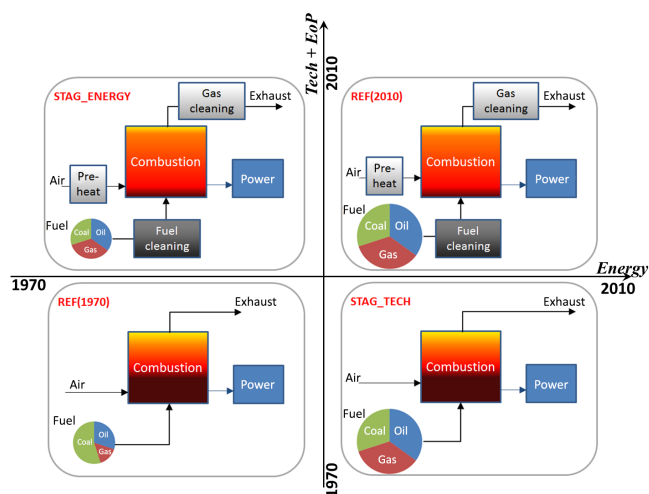
**Figure 1.** Overview of historical European Union (in blue) and international (in red) air quality regulations. UNECE/CLTRAP covers all European countries, USA, Canada, Belarus, Russia, Turkey, Israel, Ukraine, and central Asian states.

## 1 Introduction

In the last few decades, air quality issues have gained worldwide importance 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; Lelieveld et al., 2015), influences climate, visibility and ecosystems (Monks et al., 2009; IPCC, 2013), and therefore has significant effects on human life and the environment. It is crucial to understand the impacts of 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., EMEP (European Monitoring and Evaluation Programme, <http://www.emep.int/>), EPA (Environmental Protection Agency, <http://www.epa.gov/ttn/chief/eiinformation.html>) or HTAP (Hemispheric Transport of Air Pollution, <http://htap.org>). For some industrialized countries, legislation has been introduced since the mid-1980s for the power generation sector, and since mid-1990s for road transport. In many developing regions, e.g., in China, only recently regulations have been implemented for these two sectors (CSC, 2013). Therefore, we focus here on European air quality legislation, having a much longer history and affecting not only Europe, but several other countries elsewhere in the world (e.g., vehicle emission reduction standards in Japan and other Asian countries; refer to Crippa et al., 2016). Figure 1 and Table S5.1 in the Supplement summarize European regulations for air quality and air pollutant emissions since 1970, when the first air quality directive was introduced. In our work we make use of the EDGARv4.3.1 emission data (<http://edgar.jrc.ec.europa.eu/index.php>) to compare the recent (year 2010) situation with retrospective scenarios (years 1970–2010) that assess

the importance 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 crops. 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 role of the policy–industry interplay in avoiding emissions. Some publications 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 SO<sub>2</sub>, or Kurokawa et al., 2013 for Asia). Historical global emissions data sets for the past decades or century have been compiled by combining several emission inventories, e.g., Lamarque et al. (2010) for 1850–2000 and Granier et al. (2011) for 1980–2010. However, an analysis of the factors driving these emissions trends is difficult because of the heterogeneity and regional differences of the original data that might show inconsistencies over the full time period and in global coverage and cause artificial variability. Amann et al. (2013) report the evolution of anthropogenic emissions of key air pollutants between 1990 and 2010 for several world regions using the GAINS (Greenhouse Gas Air Pollution Interactions and Synergies, <http://gains.iiasa.ac.at/models/>) model. GAINS is also used to provide scenarios of future emissions (up to the year 2050) including specific assumptions of air quality and climate policies (e.g., Cofala et al., 2007). Few studies have been devoted to understand the drivers of historical emissions trends. Paruolo et al. (2015) performed a statistical causality analysis of income and SO<sub>2</sub> and CO<sub>2</sub> historic emission time series using EDGARv4.2, challenging the often assumed causal relationship between increasing GDP and decreasing emissions assumed in the environmental Kuznetz curve. Rafaj et al. (2014) aimed to identify the driving factors (historical energy balances, population and economic growth, fuel

mix, etc.) of air pollutants emissions in Europe from 1960 to 2010, using the RAINS (Regional Air Pollution and Simulation) and GAINS modeling frameworks. They decomposed the emissions into determinant factors (energy intensity, conversion efficiency, fuel mix and pollution control) to understand the evolution for SO<sub>2</sub>, NO<sub>x</sub> and CO<sub>2</sub> in Europe. They found that in Europe SO<sub>2</sub> emissions declined due to the combined effect of reduced energy intensity and shift to cleaner fuels, while abatement measures mainly reduced NO<sub>x</sub> emissions. In this work, we do not seek to analyze and decompose the emission determinant factors in view of assessing further potential of optimized reduction policies, but rather want to demonstrate the cumulative effect on emission levels in 2010 of two major factors influencing air pollution: increasing energy use and the combined technology-policy achievements to reduce emissions. To this end we develop two retrospective scenarios for 1970–2010 from a European industry–air policy perspective (Fig. 2), and we represent a range of emissions that would have been reached in 2010 under different scenario assumptions. The first and highest emission scenario, STAG\_TECH, assumes after 1970 no further improvements in technologies and abatement measures. The second retrospective and lowest emission scenario (STAG\_ENERGY) assumes stagnation of energy consumption since 1970, while the fuel mix, energy efficiency, emission factors and abatements are assumed as in the reference 2010 data. The change in global energy consumption over the last decades for the energy sector amounted to a factor of 3.6, and 2.6 for the transport sector; i.e., much more than the global population increase (1.8-fold). In addition, historical fuel consumption showed shifts in the energy mix. The latter is country-specific and depends, among others on policy choices, as well as on accessible natural reserves, the fuel price and stability, and the energy stored per unit of fuel volume. Compared to future scenarios analysis, retrospective scenarios have the advantage of using the well-known activity data time series. Obviously, no new policies can be proposed retrospectively, but compared to decomposition analysis (e.g., Rafaj et al., 2014), our results more explicitly show the impact of policy and technology choices, and energy developments arriving at 2010 emission levels. The data-driven analysis focuses both on European and global historical (1970–2010) emissions for a relatively complete set of gaseous air pollutants, SO<sub>2</sub>, NO<sub>x</sub>, CO, non-methane volatile organic compounds (NMVOCs) and NH<sub>3</sub> and particulate matter, i.e., PM<sub>10</sub>, PM<sub>2.5</sub>, black carbon (BC) and organic carbon (OC). Finally, deploying the TM5-FASST (Fast Screening Scenario Tool based on the global chemical Transport Model 5) source–receptor model, (Tavoni et al., 2014), we also demonstrate in this work the impacts of the two scenarios on health and crops, protection of which being primary objectives of environmental policies.



**Figure 2.** Schematic of the considered emission scenarios: REF(1970), REF(2010), STAG\_ENERGY and STAG\_TECH. The *x* axis represents the change in energy consumption from 1970 to 2010 (shown also by the increasing size of the fuel pie charts), while the *y* axis represents the change in abatement measures (EoP) and technologies from 1970 to 2010. The STAG\_TECH scenario has the same technologies and abatements of the REF(1970) scenario, but assumes increased energy consumption and different fuel mix (as shown in the pie chart composition) as in 2010. The STAG\_ENERGY scenario considers the energy consumption of 1970 but it includes all the technologies, abatements, fuel mix and energy efficiency of 2010.

## 2 Methodology

In Sect. 2.1 an overview of the data and assumptions used to develop the two emission scenarios is provided, addressing the EDGAR v4.3.1 bottom up emissions data set for the REF (reference), STAG\_TECH and STAG\_ENERGY emissions. This is followed by a short description 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 v4.3.1 emission data and scenarios

EDGARv4.3.1 (Emissions Database for Global Atmospheric Research version v4.3.1; <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, total non-methane volatile organic compounds (NMVOCs), NH<sub>3</sub>, as well as particulate matter compounds, namely PM<sub>10</sub>, PM<sub>2.5</sub>, BC, OC. EDGARv4.3.1 relies on international energy balances of IEA (2014) and agricultural statistics of FAO (2012) and regional or national information and assumptions on technology use and emission control standards. EDGARv4.3.1 is one of the few global emission inventories with consistent methodologies to calculate emission time

series covering 4 decades for air pollutants with high spatial resolution of  $0.1^\circ \times 0.1^\circ$  and consistent sector-specific breakdowns. Moreover, recent comparisons show the reliability of this emission inventory based on the good agreement between the EDGARv4.3.1 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). A more detailed comparison between EDGARv4.3.1 and the MACCity data at the country level is documented in Sect. S3 in the Supplement, showing regional differences from a few percent up to 50 %, which is within the range of uncertainties for specific components. The EDGAR data sets are calculated using 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 (EM) of a specific pollutant  $x$  at time  $t$  due to activity data (AD) of sector  $i$  with technologies  $j$  and end-of-pipe measures  $k$  and fuel type  $f$  in a country  $C$  as follows:

$$\begin{aligned} \text{EM}_{\text{REF},C,i}(t,x) = & \sum_{j,k,f} [\text{AD}_{C,i,f}(t) \times \text{TECH}_{C,i,j,f}(t) \\ & \times \text{EOP}_{C,i,j,k,f}(t) \times \text{EF}_{C,j,f,i}(t,x) \\ & \times (1 - \text{RED}_{C,i,j,f,k}(x))], \end{aligned} \quad (1)$$

with TECH representing the penetration fraction of a specific technology in a sector, EOP the installed fraction of end-of-pipe measures (EOP), 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 for the three scenarios on <http://edgar.jrc.ec.europa.eu/pegasos/>; in addition time series (1970–2010) of the reference data are reported as annual emissions by sector for each country on the same website. An overview of 2010 pollutant emissions is also reported in Sect. S2 in the Supplement for the three scenarios for 24 world regions. For the STAG\_TECH and STAG\_ENERGY scenarios we focus on the main sectors that were effectively targeted by air quality measures imposed by EU policies<sup>1</sup>. Firstly, the European power industry represents large national point sources, which continuously emit over a long period and have since the 1980s been equipped with additional end-of-pipe control measures, which is modeled at the Tier 2 level. The industrial combustion processes that are most suitable to be regulated are the power industry and non-power generating industry and the manufacturing industry (cement, steel and nonferrous metal industry, chemicals<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 1990s, 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. S4.

STAG\_TECH: we modeled the STAG\_TECH scenario by assuming for the three sectors of interest constant 1970 emission factors to all technologies present in the emission database globally (specified mainly regionally, but few also globally). We further assumed in the STAG\_TECH scenario no implementation of end-of-pipe (EOP) control measures in Europe. Other regional standards regulating end-of-pipe control, e.g., US power plant standards were not changed, because they fell outside the European scope of this study. Thus in STAG\_TECH, European energy production is generated by the power plants of 1970 without additional end-of-pipe measures as shown in Eq. 2 (in other words the emission reduction RED equals the lower limit<sup>3</sup>).

$$\begin{aligned} \text{EM}_{\text{STAG\_TECH},C,i}(2010,x) = & \sum_{j,k,f} [\text{AD}_{C,i,f}(2010) \\ & \times \text{TECH}_{C,i,j,f}(2010) \times \text{EOP}_{C,i,j,k,f}(1970) \\ & \times \text{EF}_{C,j,f,i}(1970,x) \times (1 - \text{RED}_{C,i,j,f,k}(x))] \end{aligned} \quad (2)$$

In the 1970s and 1980s there was a relatively large turnover of power plants (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, which is weighing strongly in the STAG\_TECH scenario results. However, in other world regions, while 1970 EFs were kept constant, the emission reduction factors (RED) were changing over time. Here STAG\_TECH results lead to even larger emissions. 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 technology stagnation was mainly reflected by not considering the emission reductions from particle filters and catalysts of all the vehicles under EURO standards, mainly present in the fleet inside Europe but also outside<sup>4</sup> (under this scenario EURO standards 1 to 5 equal the pre-EURO standards). The EURO standard penetration is shown in Table S5.2 for diesel and petrol passenger cars, light- and heavy-duty vehicles, busses and motorcycles. The change over time in technology and the implementation of the air pollutant abatement measures is assumed

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

<sup>4</sup>Aside of the American continent (North, Central, South), which implements mainly US Tier standards, other countries show a considerable share of EURO standards even outside Europe, in particular but not only Japan, Korea, etc.

<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.

**Table 1.** Emission scenario assumptions. AD (activity data), EF (emission factors), RED (reduction factor), EFF (efficiency factor), TECH (technology), EOP (end-of-pipe) and substance  $x$ .

Emission sectors	Reference 1970 and 2010	Scenario 1 in 2010: Stagnation of technology & no End-of-Pipe control (STAG_TECH)	Scenario 2 in 2010: Stagnation of energy consumption (STAG_ENERGY)
Agricultural waste burning	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Energy industry	EDGARv4.3.1	$AD(2010) \times TECH(2010) \times EOP(1970) \times EF(1970,x) \times (1-RED(x))$	$AD(1970) \times TECH(1970)/(EFF(1970)/EFF(2010)) \times EOP(2010) \times EF(2010,x) \times (1-RED(x))$
Solid waste disposal	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Combustion in manufacturing industry	EDGARv4.3.1	$AD(2010) \times TECH(2010) \times EOP(1970) \times EF(1970,x) \times (1-RED(x))$	$AD(1970) \times TECH(1970)/(EFF(1970)/EFF(2010)) \times EOP(2010) \times EF(2010,x) \times (1-RED(x))$
Industrial processes & product use	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Oil production & refining	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Buildings (residential & others)	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Fossil fuel fires	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Road Transportation	EDGARv4.3.1	$AD(2010) \times TECH(2010) \times EOP(1970) \times EF(1970,x) \times (1-RED(x))$	$AD(1970) \times TECH(1970)/(EFF(1970)/EFF(2010)) \times EOP(2010) \times EF(2010,x) \times (1-RED(x))$
Aviation (international+domestic)	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Shipping (international+domestic)	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1
Non-road ground transport transport	EDGARv4.3.1	EDGARv4.3.1	EDGARv4.3.1

in EDGARv4.3.1 to start in the year the directive came into force, but the actual timing of the implementation is subject to large uncertainty as it could be pre-empted by, e.g., striving towards newer technologies in the case of the manufacturing industry. It could also be 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 by 2010, by comparing the reference with STAG\_TECH emissions in 2010.

**STAG\_ENERGY:** the STAG\_ENERGY scenario was modeled by assuming that the three sectors of interest consumed the same amount of energy (TJ) as in 1970, but with the 2010 fuel mix, energy efficiency (EFF), technologies, and end-of-pipe abatements, as shown in Eq. (3). 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 with the 2010 share but at the 1970 energy level (in TJ). In addition to the calibration per sector of the energy consumption level (in TJ),

we evaluated the change in energy efficiency by fuel type, sector and region. For the power generation sector we scaled for each country the “main activity producer electricity plants (TJ)” with the 1970 over 2010 ratio of the “electricity output of main activity producer electricity plants (GWh)” from IEA (2014). For the road transport sector we scaled the “fuel consumption for road transport” with a factor composed of the 1970 over 2010 road transport fuel consumption ratio multiplied with the 1970 over 2010 fuel efficiency ratio. The latter was calculated with the macro-regional averaged values of petrol and diesel economies ( $L\ 100\ km^{-1}$ ) in 2010 and 1975 (because of missing 1970 data) for different type of vehicles (passenger cars, light-duty vehicles, heavy-duty vehicles, busses, mopeds and motorcycles) distinguishing the fuel consumption for petrol and diesel based on the EPA Trends report (EPA, 2013).



$$\begin{aligned}
 & EM_{\text{STAG\_ENERGY},C,i}(2010,x) \\
 &= \frac{\left[ \sum_f AD_{C,i,f}(1970) \right] / \text{EFF}_{C,i,f}(1970)}{\left[ \sum_f AD_{C,i,f}(2010) \right] / \text{EFF}_{C,i,f}(2010)} \\
 &\times EM_{\text{REF},C,i}(2010,x) \quad (3)
 \end{aligned}$$

The comparison of the STAG\_ENERGY emissions scenario with the reference emissions REF in 2010 highlights the emission reductions that were not realized because of increased energy consumption (e.g., to generate extra kWh electricity or to drive more distance per vehicle) with the 2010 technology and 2010 end-of-pipe abatement. Compared to the 1970 reference emissions, STAG\_ENERGY demonstrates the benefit of all industrial developments towards less energy-intensive and less polluting technologies. It includes not only the technological progress with end-of-pipe measures but also the shifts towards less carbon-intensive fuels (e.g., natural gas instead of coal) and increase of fuel economy and energy efficiency. On the other hand, compared to the REF(2010) data, STAG\_ENERGY assesses to what extent emission increases by consumption growth. It should be noted that pre-combustion treatment (cleaning) of fuels, such as coal washing or desulfurization of diesel and heavy residual fuel oil is not part of the STAG\_ENERGY scenario but is addressed as a technology effect in the STAG\_TECH scenario. Therefore, the fuel quality directives show their emissions savings (mainly on sulfur) in the STAG\_TECH scenario while the fuel taxation policies (e.g., preferring diesel over petrol) are present in the STAG\_ENERGY scenario.

In the ACPD version of this paper (Crippa et al., 2015), another scenario called STAG\_FUEL discussed the combined impact of stagnation of fuel-mix and fuel amount. However, in revising the manuscript we decided to focus on the consumption of energy (TJ) instead of fuel because we consider the fuel mix and efficiency choices as exogenous variables just as the technology progress and end-of-pipe measures. When considering a stagnation of fuel with constant fuel mix and energy since 1970, due to the remaining contributions of relatively dirty fuel, this scenario results in higher emissions than STAG\_ENERGY. This scenario is here not further discussed, since the interpretation of results is not adding much to the STAG\_ENERGY scenario. The interested reader is referred to the corresponding ACPD paper.

## 2.2 Reduced-form air quality model TM5-FASST

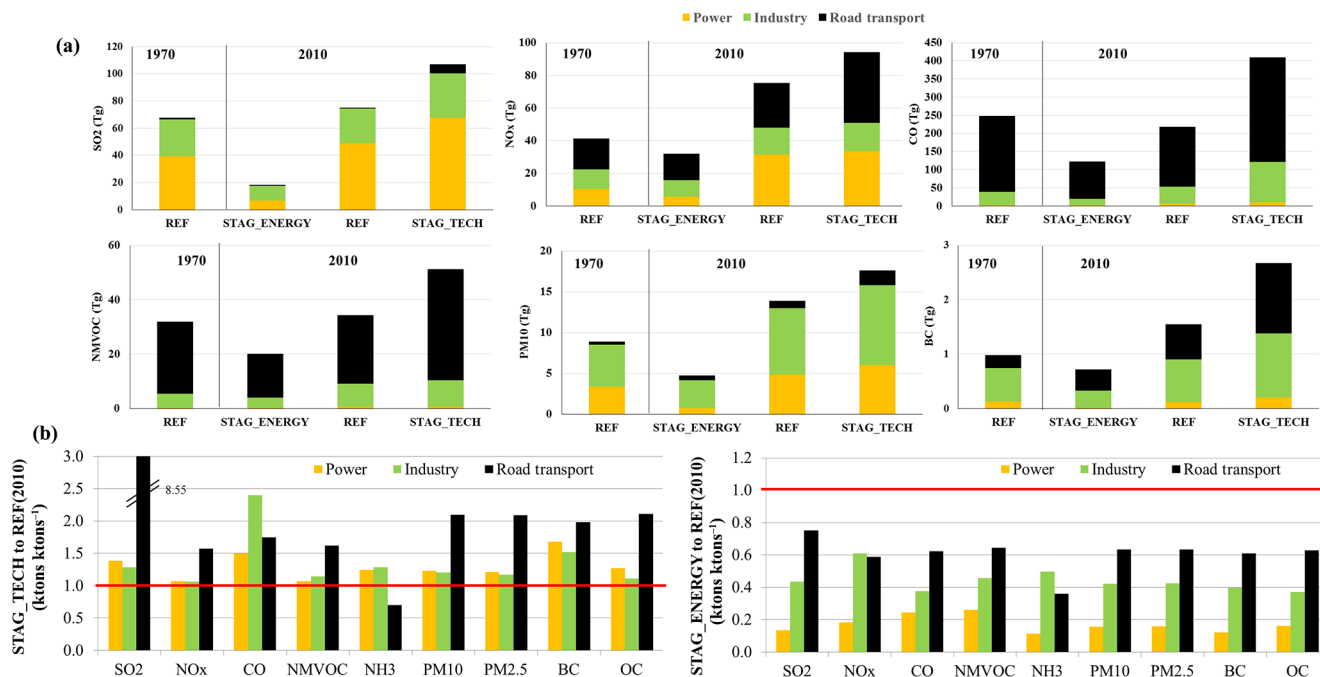
The TM5-FASST model (Fast Scenario Screening Tool, version v4.2.0\_2014) is a linearized source–receptor model derived at a receptor resolution of  $1^\circ \times 1^\circ$  from the global chemical transport model TM5-CTM (Tracer model 5, chem-

istry transport model, Krol et al., 2005) for gaseous and particulate matter atmospheric pollutants. It considers 56 world regions (both as source and receptor regions), with a higher detail over Europe which is represented by 16 regions. 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 base concentration (BASE) and the contribution of the linearized matrix function for each precursor (Eq. 4):

$$\begin{aligned}
 & \begin{bmatrix} \text{CONC}_{R1} \\ \dots \\ \text{CONC}_{R56} \end{bmatrix} (t,x) = \begin{bmatrix} \text{BASE}_{R1} \\ \dots \\ \text{BASE}_{R56} \end{bmatrix} (t,x) \\
 & + \sum_l \begin{bmatrix} \alpha_{R1S1}(t,x,l) & \dots & \alpha_{R1S56}(t,x,l) \\ \dots & \dots & \dots \\ \alpha_{R56S1}(t,x,l) & \dots & \alpha_{R56S56}(t,x,l) \end{bmatrix} \\
 & \times \begin{bmatrix} \Delta \text{EM}_{S1}(t,l) \\ \dots \\ \Delta \text{EM}_{S56}(t,l) \end{bmatrix}, \quad (4)
 \end{aligned}$$

where  $\alpha_{R1S1}, \dots, \alpha_{R56S56}$  are the source receptor coefficients for precursors  $l$  of substance  $x$  and  $\Delta \text{EM}$  the difference between the reference emission and the actual data. They have been derived from precursor emission perturbation model runs with TM5 using a reference emission data set for the year 2000 and meteorology fields for the year 2001.

For each source region, the TM5-FASST model requires as input the annual emissions of primary  $\text{PM}_{2.5}$ , to be specified as BC, primary organic matter (assumed to be  $1.3 \times \text{OC}$ ), and of the precursors ( $\text{SO}_2$ ,  $\text{NO}_x$ , 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 any source region to pollutant concentrations at any receptor region, emulating underlying 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 anthropogenic SOA (secondary organic aerosols) is performed. We note that natural emissions, e.g., secondary organic aerosol from biogenic sources, lightning  $\text{NO}_x$ , and biogenic sources of VOCs (etc.), are included in the reference simulation following the AEROCOM recommendations in Dentener et al. (2006), however without accompanying source–receptor relationship calculations. Ozone formation is simulated through the reactions involving VOCs and  $\text{NO}_x$ . FASST evaluates the impacts of  $\text{O}_3$  and  $\text{PM}_{2.5}$  concentrations on health and crops and vegetation. Tropospheric ozone and particulate matter negatively affect human health, increasing respiratory and cardiovascular diseases, lung can-



**Figure 3.** (a) Overview of 1970 (only for REF) and 2010 emissions for REF, STAG\_TECH and STAG\_ENERGY at the global scale for the power generation, industry and road transport sectors. (b) The ratios in 2010 of STAG\_ENERGY to REF(2010) and STAG\_TECH to REF(2010) are presented. The red line indicates no change relative to the reference emissions in 2010.

cer, etc. (WHO, 2013). Through parameterizations relating pollutant concentrations and exposed population (Anenberg et al., 2009; Jerrett et al., 2009; Burnett et al., 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.

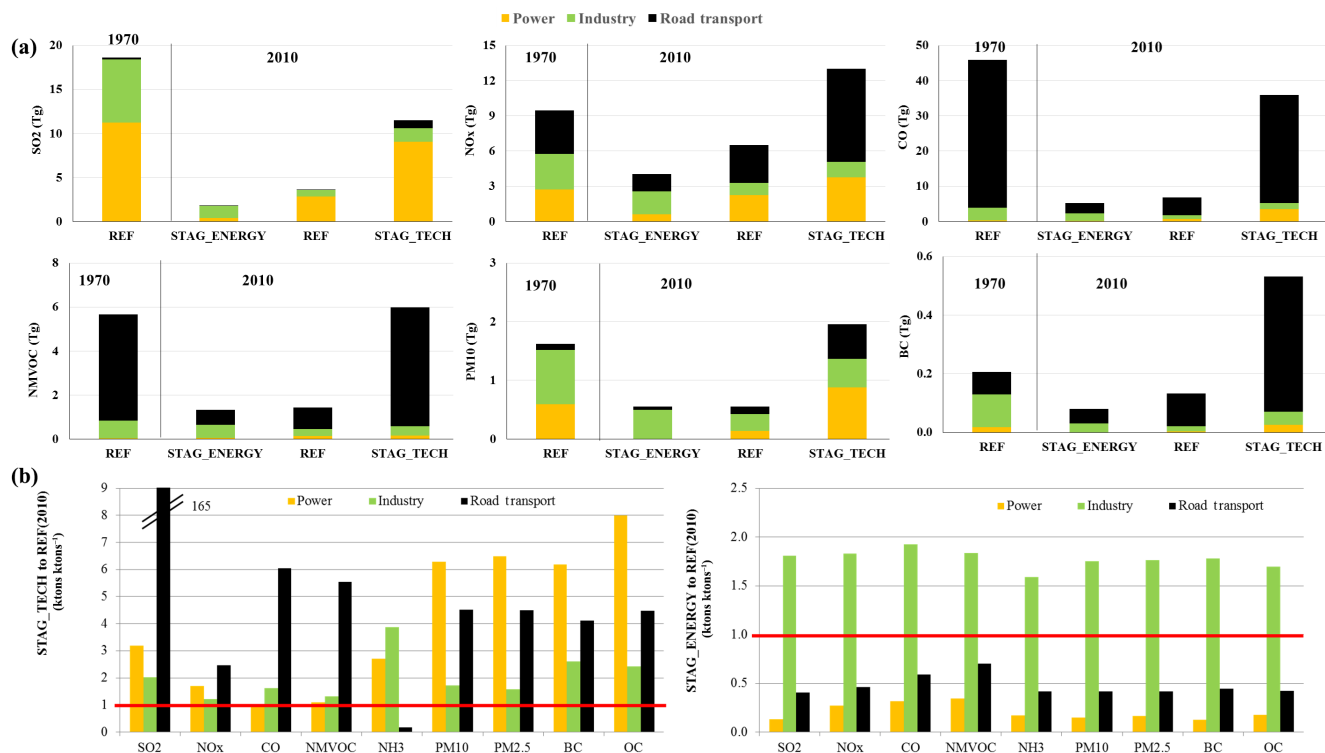
### 3 Emission scenarios results

In Fig. 3, we first compare the global reference emission levels in 1970 (REF(1970)) and 2010 (REF(2010)), and then the two retrospective scenarios (STAG\_TECH and STAG\_ENERGY) to the REF(2010). With the scenarios we focus on the three selected sectors: power industry (labeled “power”), non-power industrial combustion of the manufacturing industry (“industry”) and road transport (“road”). We then evaluate the changes at the EU level in Fig. 4. Note that in our evaluations we consider the year 2010 as the reference year.

### 3.1 Global emission trends

Global SO<sub>2</sub> emissions from power, industry and road transport sectors (Fig. 3) do not show a significant trend from 1970 (at 68 Tg) to 2010 (75 Tg), because the emission reductions by pre-combustion preparation of fuel (e.g., coal sulfur wash) and the post combustion exhaust treatment of SO<sub>2</sub> (e.g., by flue gas desulfurization (FGD) units) were counterbalanced by the increased use of fuel (in particular of coal) worldwide. In both 1970 and 2010 the power sector contributed strongest to total sulfur emissions, and for the STAG\_TECH scenario, when no technological progress for sulfur emission reduction would have taken place in the EU, global power SO<sub>2</sub> emissions would have been 1.3 times higher than REF. However, for STAG\_ENERGY, assuming constant energy consumption as in 1970 for the power, industry and road transport sectors, but with current technology and end-of-pipe measures and 2010 fuel mix and efficiency, global SO<sub>2</sub> emissions would be only 24 % of the 2010 emissions. For the other pollutants, like NO<sub>x</sub>, CO and NMVOCs, we see a significant increase in REF from 1970 to 2010, which reflects mainly the increase in energy consumption in the power sector (e.g., NO<sub>x</sub> emissions from power generation tripled globally, from 10.5 to 31.4 Tg). Depending on the pollutant, the STAG\_ENERGY emissions are lower by 40 to 75 % in 2010 (Fig. 3). The largest impact of STAG\_ENERGY in 2010 is seen in the power generation sector emissions (more than 80 % as global average for





**Figure 4.** (a) Overview of 1970 (only for REF) and 2010 emissions for REF, STAG\_TECH and STAG\_ENERGY at the European scale for the power generation, industry and road transport sectors. (b) The ratios in 2010 of STAG\_ENERGY to REF(2010) and STAG\_TECH to REF(2010) are presented. The red-line indicates no change relative to the reference emissions in 2010.

all pollutants), ranging from ca. 20 % reduction in developing and emerging regions (e.g., Asia, Latin America, etc.) to more than 95 % reduction in industrialized regions (USA, EU, Japan, etc.).

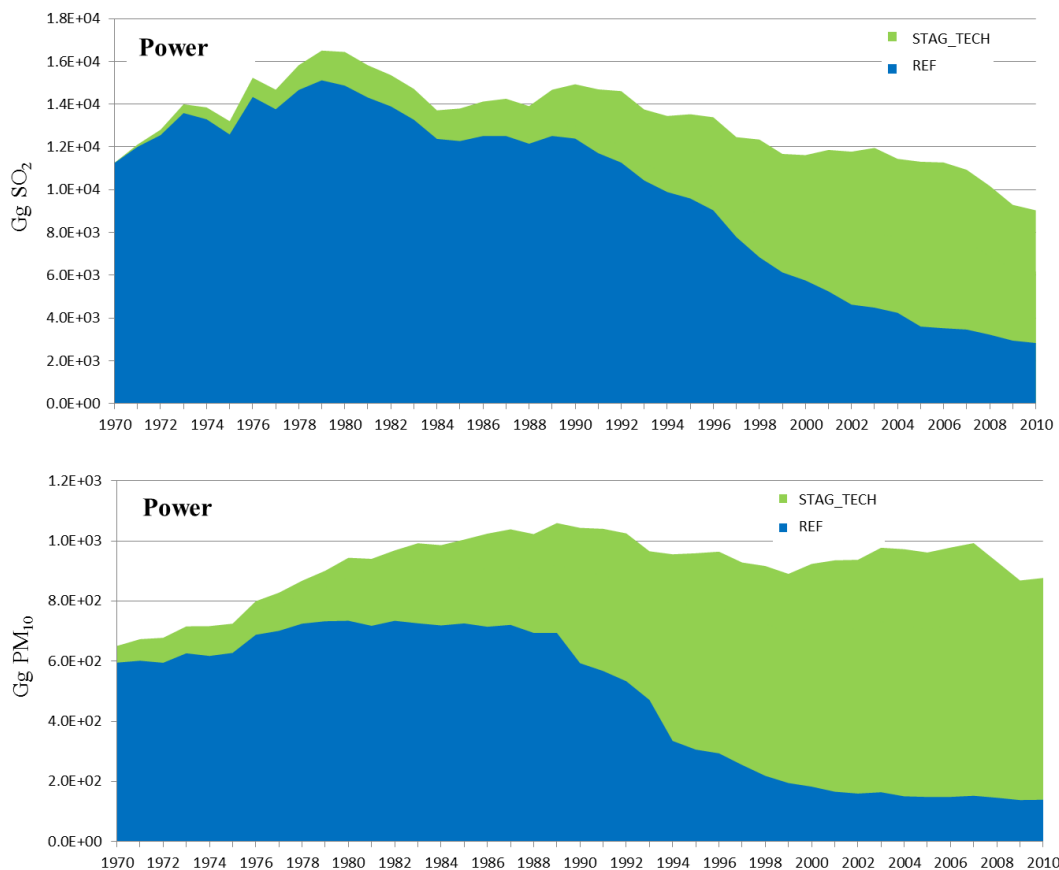
When comparing the STAG\_TECH to the REF(2010) case, we find a global reduction of road emissions by 8.5 times for SO<sub>2</sub>, ca. 1.5 times for NO<sub>x</sub>, CO and NMVOCs and 2 times for PM, which clearly illustrates the phasing-in of less emitting vehicles due to stricter EURO standards. A shift from diesel to petrol could have caused a further reduction of PM emissions but the opposite took place: diesel vehicles represented only ca. 20 % of the global fleet in 1970 compared to ca. 75 % in 2010. Therefore the PM emission reductions shown in the STAG\_ENERGY scenario compared to the REF(2010) is less strong for transport emissions and mainly driven by the power sector. Other sectors, such as residential combustion, also contribute significantly to the total emissions, but are not so easily controlled and moreover there was no comprehensive legislation in place. For the aerosols (PM<sub>10</sub>, PM<sub>2.5</sub> and BC) the increase in REF emission levels from 1970 to 2010 is stronger than for other pollutants (ca. 40 %), although we remark here that the major contribution comes from residential combustion, which was not evaluated in our scenarios. More information on the ratios between each retrospective scenario and the reference case are given

in Tables S1.1 and S1.2. In the sections below, we focus on the three sectors separately for the historical trend in Europe with REF(1970) and REF(2010), and then the STAG\_TECH and STAG\_ENERGY scenario cases.

## 3.2 European emission trends

### 3.2.1 EU power industry (“power”)

Figure 5 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 and by the change in emission factors and technology (green area, STAG\_TECH). The concurrent effects due to the change in fuel quality (Directive 98/70/EC, 1998, as well as international conventions like CLRTAP and Gothenburg Protocol) leading to the introduction of abatement measures (non-regenerative dry and/or semidry and wet flue gas desulfurization), following the directive regulating emissions from large combustion plants (2001/80/EC, 2001), determined the actual REF(2010) emission levels. Concerning particulate matter (here represented by PM<sub>10</sub>, but similar results are obtained for PM<sub>2.5</sub> and its carbonaceous components), in 1970 power plants were already equipped with some abatement measures (e.g., cyclones), which installed regularly through-

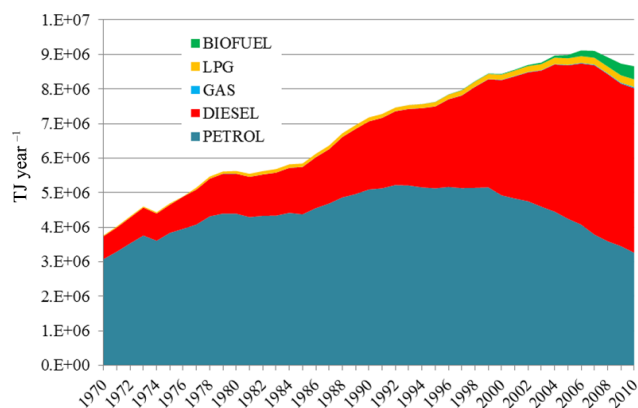


**Figure 5.** Effect of air quality abatements (STAG\_TECH scenario) on annual power generation emissions in Europe (EU27), SO<sub>2</sub> (upper panel) and PM<sub>10</sub> (lower panel).

out in the nineties (using finer filters) not only due to legislative restrictions. In particular pre-combustion treatment of fuel and an engine design optimizing the combustion process helped not only to reduce the emissions but also to increase the fuel efficiency (decreasing the variable cost of fuel input) and robustness (protecting delicate components from corrosive air pollutant gases and to shorten outages). Therefore, the major reduction in REF PM<sub>10</sub> emissions between 1970 and 2010 is due to the application of abatement measures. The impact of European legislation on emissions from the power generation sector, evaluated by STAG\_TECH, can strictly be interpreted only in the European context. As we explained earlier, STAG\_TECH took into account a stagnation of technologies at the global level and in addition for Europe alone, regulations on air pollution reduction equipment. The effect of additional policies outside of Europe has not been taken into account here as discussed in the introduction and methodology sections. Figure 4 shows that in Europe the REF emissions decreased from 1970 to 2010 for all air pollutants and aerosols. This reduction was primarily obtained through the introduction of end-of-pipe measures.

For the STAG\_ENERGY scenario, we observe the lowest emissions both at global and European scales. With this

scenario, in 2010 pollutant emissions would vary between 10 and 30 % of the REF(2010) emission values both at the European and global scales (refer to Figs. 3b and 4b as well as to Table S1.2). Compared to REF(1970), the STAG\_ENERGY scenario shows around 95 % lower SO<sub>2</sub> and PM emissions, 76 % lower NO<sub>x</sub> and ca. 30 % lower CO and NMVOC due to all technological progress, enhanced energy efficiency and an important shift to less carbon-intensive fuel use. In 2010, coal-related fuels represented ca. 49 % of EU fuel consumption, a decrease of 16 compared to the 65 % in 1970, while the EU phased out the use of heavy residual fuel oil in the power sector, leading to a contribution of oil decreasing from 25.5 to 9 %. The EU power sector increased its fuel consumption by adding natural gas, in particular in the 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. The balance is made by a higher share of clean, low-sulphur fuels, such as natural gas and wood, (35 vs. 9 % and 7 vs. 0.5 %, in 2010 and 1970, respectively). Therefore, 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 con-



**Figure 6.** Change between 1970 and 2010 of energy content of major fuels ( $\text{TJ yr}^{-1}$ ) used by EU27 passenger cars. Note the large shift from petrol to diesel.

tent) and flue gas (to lower  $\text{SO}_2$  emission) following EU policies, confirming the analysis of Rafaj et al. (2014). At the European level, in 2010 the STAG\_TECH scenario produced 1.7 times higher  $\text{NO}_x$  emissions than REF, when assuming technology stagnation and less optimized combustion processes (lower efficiency, lower air–fuel mix; see Table S1.1 and Fig. 4b). Interestingly, also  $\text{NH}_3$  emissions are higher in STAG\_TECH compared to REF due to the fuel shift from oil to gas which emits much less  $\text{NH}_3$  than oil. European legislation has also been successful in effectively abating  $\text{PM}_{10}$  emissions from power plants, reducing them by a factor of 6.3 compared to the STAG\_TECH scenario. Similar results are found for  $\text{PM}_{2.5}$  emissions and its components (BC and OC).

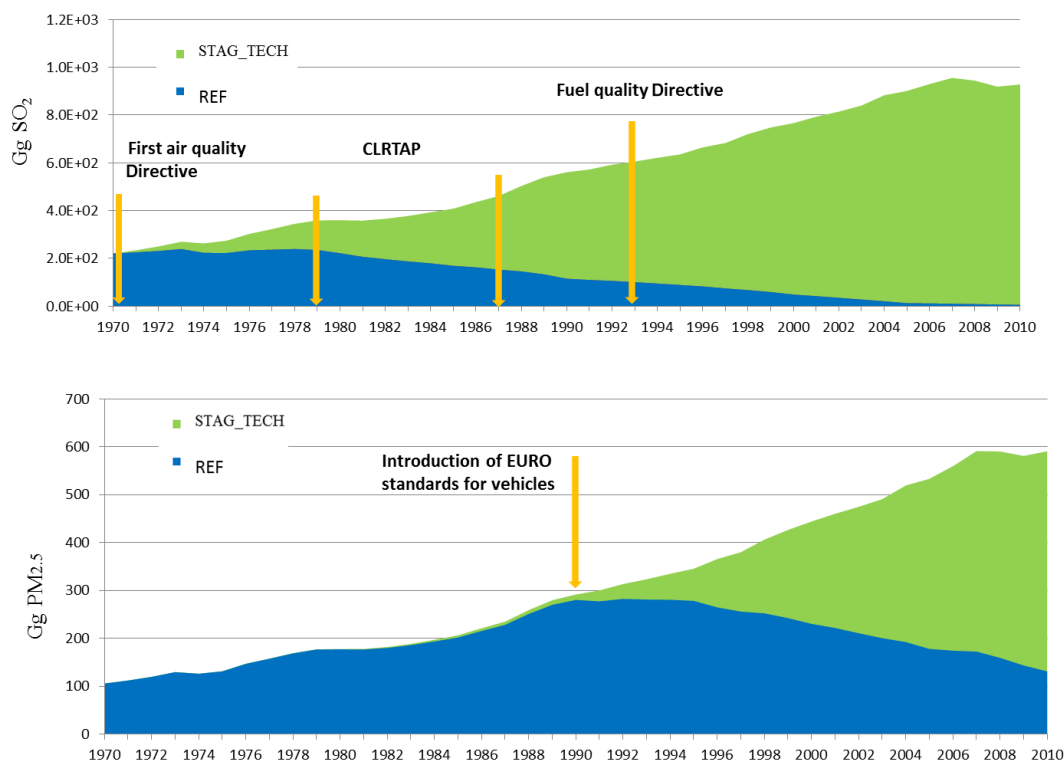
### 3.2.2 EU manufacturing industry (“industry”)

The primary emissions of industrial activities, including all manufacturing activities, are  $\text{SO}_2$ ,  $\text{NO}_x$ , CO and PM, while NMVOC emissions are to a large extent due to the use of solvents and specific chemical processes. As shown in Table S1.2, the ratio of STAG\_ENERGY to REF(2010) for the industrial sector is larger than 1 for EU27 due to the presence of heavy industry in European countries in the seventies (the ratio of STAG\_ENERGY to REF(2010) for Central Europe is 2.1, while for OECD Europe is 1.6). Ratios higher than 1 are also observed for other world regions such as USA, Japan, Central Asia (Asia-Stan), Russia, Turkey and Ukraine due to the shift of the heavy industry to emerging countries like China and India having a STAG\_ENERGY to REF(2010) ratios equal to 0.2–0.3. We note that the ratio STAG\_ENERGY to REF(2010) does not vary for the different substances since the change in energy efficiency and its fuel dependence were not considered for the industrial sector. The emissions from the manufacturing industry were affected by the shift to cleaner fuels from 1970 to 2010 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. 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), we find that only the emissions from  $\text{SO}_2$ , CO,  $\text{NH}_3$  and PM components are moderately affected in Europe with ratios ranging from 1.2 to 2.6, except for  $\text{NH}_3$  where, as mentioned before, the ratio is 3.9 due to higher emissions from oil combustion than from gas. Therefore, even in Europe, REF emissions from the manufacturing industry sector are generally higher in absolute terms than 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. The recent European directive 2010/75/EU (2010) will further regulate emissions from industrial activities, with emission reductions expected to materialize after 2010.

### 3.2.3 EU road transport (“road”)

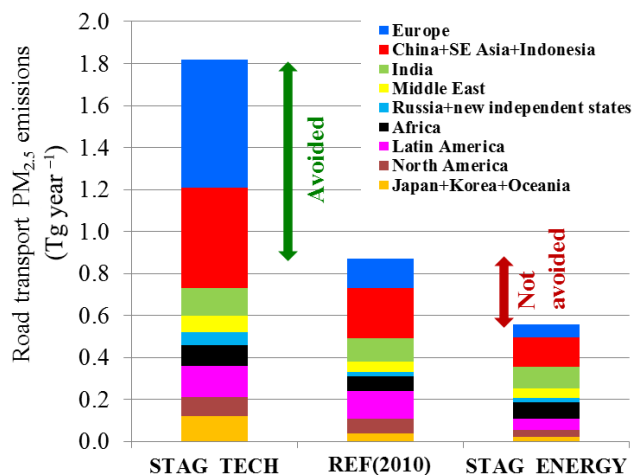
The largest effects of technology changes and end-of-pipe control measures are observed in the road sector in the EU. The fuel quality directive reduced  $\text{SO}_2$  emissions by 2 orders of magnitude (a factor 160, see Fig. 4b). Lower  $\text{SO}_2$  emissions in 2010 are mainly associated with the implementation of EU fuel quality directives (in accordance with international conventions), the shift to cleaner fuels, and less by the presence of EOP measures. With the optimization of combustion technology (motor inside flow and combustion, common rail fuel injection, preheating) CO and NMVOC emissions have also been reduced. With the adoption of EURO standards, particulate filters have increasingly been introduced in the car fleets, and exhaust PM levels were reduced by more than a factor of 4. However, real-world emissions become more determined by a relatively small fraction of vehicles with lower emissions standards or defect equipment, such as the so-called super-emitting cars. Moreover, particulate filters do not work properly when the S content in the fuel is larger than 50 ppm, an issue which arises when cars are exported to developing countries (such as Africa) with less clean fuel on the market. Furthermore, EURO standards reduced  $\text{NO}_x$  emissions by 2.5 times, at the expense of a 5.5 times increase in  $\text{NH}_3$  emissions because of the catalysts ( $\text{NH}_3$  is the only substance that is decreased in emissions under the STAG\_TECH scenario, refer to Fig. 4b and Table S1.1). REF(2010) CO emissions are reduced by 6 times in 2010 compared to STAG\_TECH. Figure 4 highlights the impact of the increased energy consumption in the road transport sector from 1970 to 2010, resulting in the lowest emissions for all pollutants as observed for the STAG\_ENERGY scenario. At the European scale pollutants are reduced from 1.4 to 2.5 times of the actual REF(2010) values if the energy



**Figure 7.** Effect of air quality abatements (STAG\_TECH scenario) on annual road transport emissions in Europe (EU27), SO<sub>2</sub> (upper panel) and PM<sub>10</sub> (lower panel).

consumption remained at 1970 levels (STAG\_ENERGY). Similar values are observed for USA and other industrialized countries, while lower impact is seen for developing regions (from a few percent up to 20 % reduction). European NO<sub>x</sub> and BC emissions in the EU increased in 2010 by a factor of ca. 2.3, compared to the STAG\_ENERGY scenario (see Fig. 4a), 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 (Fig. 6). Figure 7 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 agreements made in the international CLRTAP and Gothenburg Protocol, thus reducing SO<sub>2</sub> road emissions. In 1999 the European Union directive 1999/32/EC (1999) 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. 7). Only with the gradual introduction of particle filters in the 1990s, requested by EURO standards for vehicles from EURO1 in 1992 to EURO5 in 2009 (Table S5.2), PM road transport emissions reduced by a factor 4–5. This exemplifies the policy response to different types of pollutants and sources through the implementation of new policies. Figure 8 reports road transport PM<sub>2.5</sub> emissions for the year 2010 and for the two scenarios

(STAG\_TECH and STAG\_ENERGY) for world regions (details about region classifications can be found in Sect. S4.4). The comparison of STAG\_TECH and REF(2010) PM<sub>2.5</sub> data represents the emissions reductions due to the implementation of 2010 technologies and EURO standard abatements, while the difference between the STAG\_ENERGY scenario and the REF(2010) data represents the enhanced emissions (60 %) due to the increased energy consumption from 1970 to 2010. 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 (i.e., the North America UT1, UT2, UT3, PH1 and PH2 standards). Further analysis about the spillover of the EURO standards outside Europe is presented in Crippa et al. (2016).



**Figure 8.** Regional contributions to  $\text{PM}_{2.5}$  road transport emissions in 2010 for the STAG\_TECH, REF and STAG\_ENERGY scenarios. The green arrow (STAG\_TECH minus REF) indicates the amount of emissions avoided by the combination of legislation and technological advancements, and the red arrow (REF minus STAG\_ENERGY) the additional emissions associated with increased energy use between 1970 and 2010.

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

Figure 9 shows the spatial ( $0.1^\circ \times 0.1^\circ$  long-lat) distribution in Europe of the difference in emissions of the STAG\_TECH and REF(2010) scenarios for selected pollutants ( $\text{SO}_2$ ,  $\text{NO}_x$ , CO,  $\text{PM}_{10}$  and BC). These maps can be interpreted as the emissions reduction in Europe due to the implementation of European air quality legislation on the power generation and road transport, together with the change in emission factors and technologies, which also affected the industrial sector. The avoided  $\text{SO}_2$  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 major emitting activities, while many point sources (power plants and industries) are spread over Europe, leading to the reduction in  $\text{SO}_2$  emissions due to the switch to cleaner fuels (shifting of fuel types and lower sulfur content). A different spatial distribution of emission reductions is found for  $\text{NO}_x$  and CO, where in addition to urban areas, a large reduction is observed for road transport (road tracks are visible for both pollutants). Interestingly, Italy, Germany, the United Kingdom and the Benelux region display strong and more uniform CO emission reductions compared to other European regions (e.g., France and Spain) because of the contributions of the manufacturing industry.  $\text{PM}_{10}$  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 highly effective in very in-

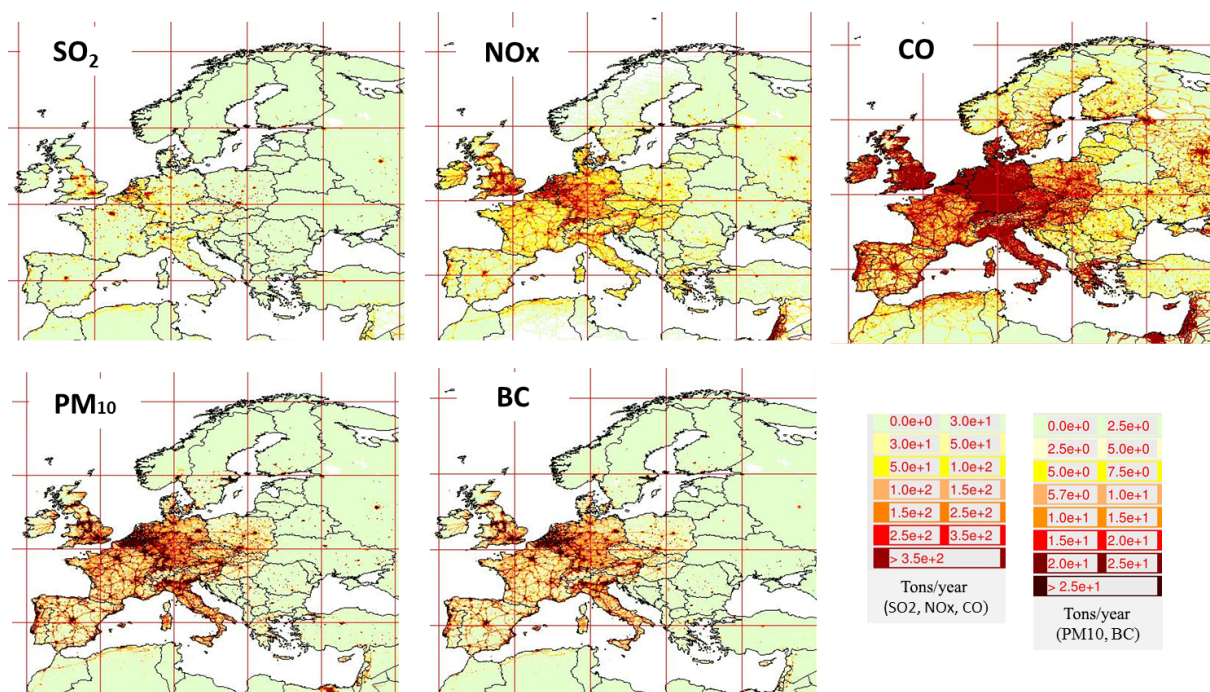
dustrialized areas (e.g., Benelux) and other major conurbations.

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

### 4.1 Concentration and composition changes

Power, industry and road emissions data from the considered scenarios have been used in the TM5-FASST model to derive the corresponding population weighted-average  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentrations for the main world regions. Figure 10 shows the global impact of the historical change in emissions of 1970 compared to 2010 (REF(1970) vs. REF(2010)) on  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentrations, and those of STAG\_ENERGY and STAG\_TECH relative to the REF(2010) data. Note that delta emissions are calculated as the difference between each scenario and the REF(2010) data.  $\text{PM}_{2.5}$  REF concentrations decreased from 1970 to 2010 by 4.7 and  $5 \mu\text{g m}^{-3}$  in the USA and Europe, respectively, while increased concentrations in 2010 are especially observed for Asian countries (15 and  $12.5 \mu\text{g m}^{-3}$  for China and India respectively), Africa ( $0.7 \mu\text{g m}^{-3}$ ) and Latin America ( $0.5 \mu\text{g m}^{-3}$ ). A similar pattern is also observed for  $\text{O}_3$  where industrialized countries had higher computed concentrations in 1970 compared to 2010 (delta  $\text{O}_3$  equal to 3.1 ppb for USA and to 0.4 ppb for Europe), while developing countries increased their concentrations in 2010 by 12.8, 5, 2.7 and 0.9 ppb for Asia, Latin America, Africa and Russia, respectively. Similarly to the change observed from 1970 to 2010, comparing the STAG\_ENERGY scenario to the REF(2010) case,  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentrations show the expected opposite patterns for industrialized and emerging countries; however, the largest impact is observed for Asia where the stagnation of energy consumption at 1970 levels would have produced much lower concentrations (annual population-weighted average delta  $\text{PM}_{2.5} = 11.9 \mu\text{g m}^{-3}$  and delta  $\text{O}_3 = 13.7$  ppb) compared to the reference 2010 levels. A markedly different impact pattern is observed for the STAG\_TECH scenario versus the REF(2010), 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  $\text{PM}_{2.5} = 4.9 \mu\text{g m}^{-3}$  and delta  $\text{O}_3 = 2.5$  ppb), Asia (delta  $\text{PM}_{2.5} = 1.7 \mu\text{g m}^{-3}$  and delta  $\text{O}_3 = 1.8$  ppb) and Russia (delta  $\text{PM}_{2.5} = 0.8 \mu\text{g m}^{-3}$  and delta  $\text{O}_3 =$  ppb). As shown in more detail in Fig. S6.1 in the Supplement, 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  $4\text{--}5 \mu\text{g m}^{-3}$  lower concentrations in Europe as  $\text{PM}_{2.5}$  (including both the primary and secondary particulate components simulated by TM5-FASST). Power plant  $\text{PM}_{2.5}$  concentrations decreased by 1.9 and  $2.9 \mu\text{g m}^{-3}$  in western and central Europe (with a





**Figure 9.** Hotspots of avoided emissions due to progressive implementation of air pollution policy and better technology in Europe: difference of STAG\_TECH and REF emissions in 2010 ( $\text{tyr}^{-1}$  ( $0.1^\circ \times 0.1^\circ$  gridcell) $^{-1}$ ).

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  $\text{PM}_{2.5}$  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 market impacts of this specific sector are studied in more detail separately in Crippa et al. (2016). For the USA, the large reduction in  $\text{PM}_{2.5}$  concentration is due to the power generation sector. Although the end-of-pipe measure implementation in US power plants is ascribed to US air quality legislation (and thus 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  $\text{PM}_{2.5}$  changes for the power, road transport and industrial sectors (Fig. S6.2), 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  $\text{PM}_{2.5}$  chemical composition is mainly formed by secondary inorganic components (nitrates, sulfates and ammonium) as well as other  $\text{PM}_{2.5}$ . 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  $\text{PM}_{2.5}$  chemical composition response is found for road transport, which consists of primary organic mat-

ter, BC, as well as ammonium nitrate particles formed by the chemical reaction of  $\text{NO}_x$  and  $\text{NH}_3$  emitted by this sector. The delta particulate  $\text{SO}_4$  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.  $0.5 \mu\text{g m}^{-3}$  less for the year 2010 on average ( $1.5$  for USA,  $1.3$  for China,  $1.1$  for central Europe and  $0.9 \mu\text{g m}^{-3}$  for western Europe). While the impact on  $\text{PM}_{2.5}$  concentrations due to technology and emission reductions (STAG\_TECH scenario) is mostly found within the source region with emissions change (Europe, Japan, China), longer-range effects are found for ozone. This is an important result because it represents the need of having intercontinental policies for some pollutants.  $\text{O}_3$  formation is driven by the reaction of the precursors  $\text{NO}_x$ , CO and NMVOCs, derived mostly from the road sector and strongly abated over the past 2 decades with the EURO standards. The avoided annual and regional average  $\text{O}_3$  concentrations range between 0.5 and 6 ppb, which significantly affects the current  $\text{O}_3$  levels 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 surface delta  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentrations are used in TM5-FASST to estimate the impact of the specific scenario assumptions on human health (using the life expectancy standard of GBD, 2010) and crop yield (expressed



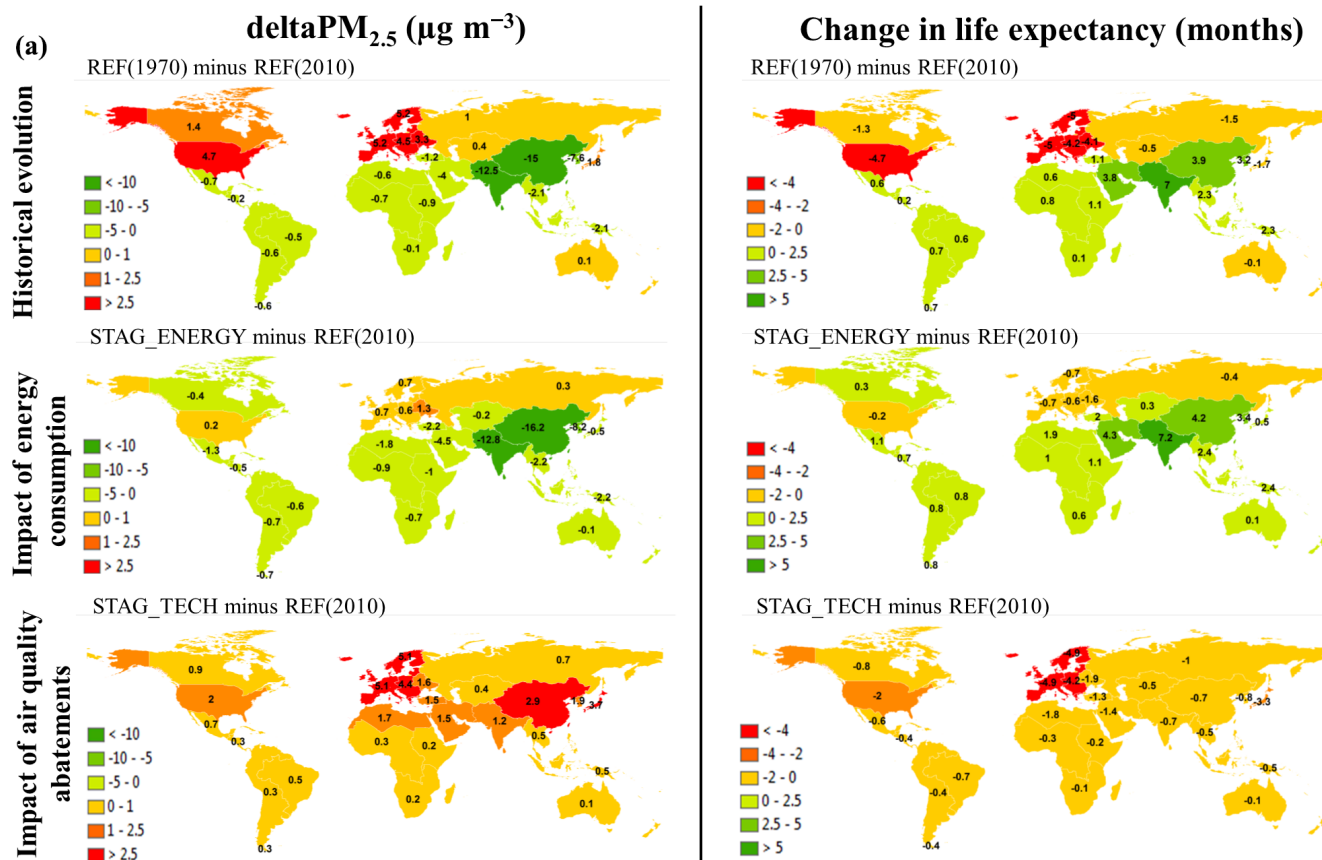


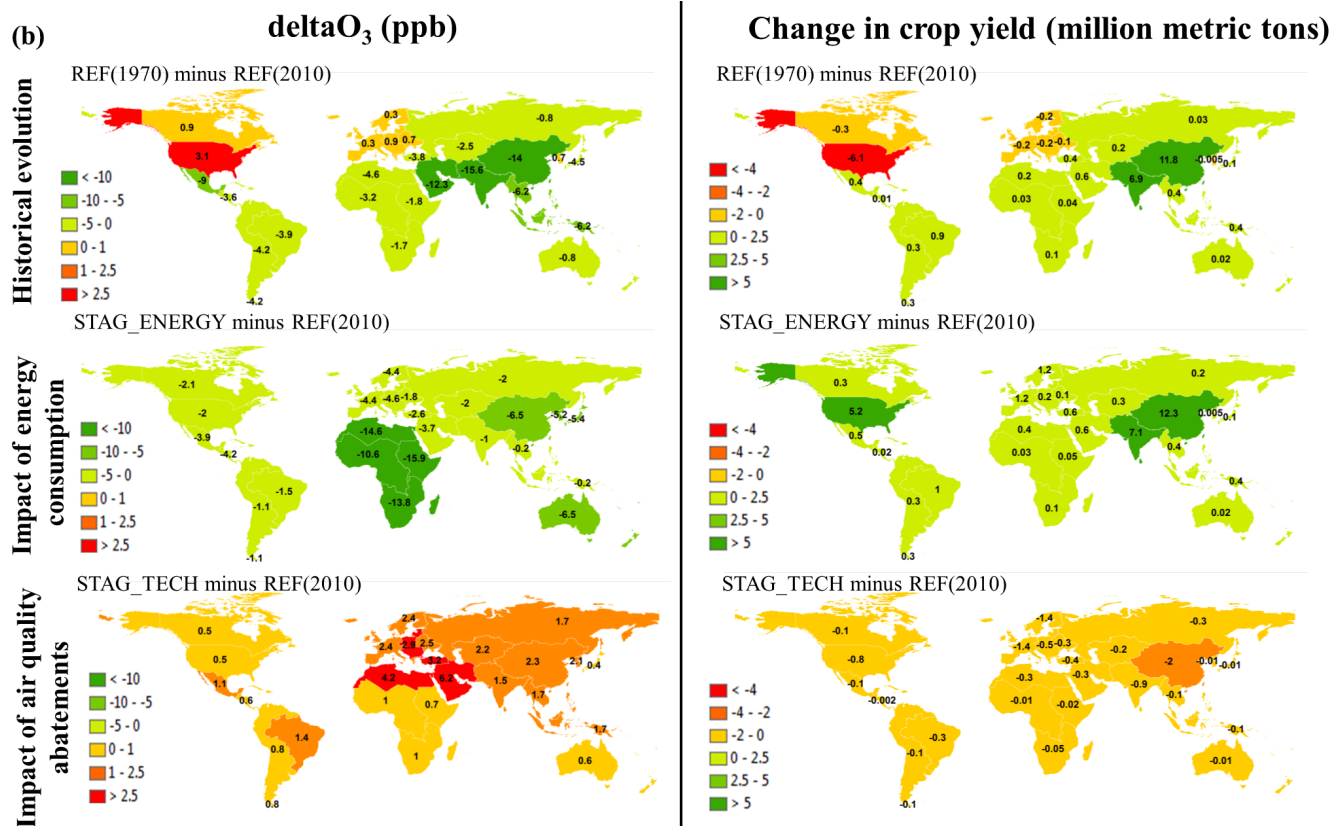
Figure 10.

in tons of crops produced extra or not lost; see Fig. 10). An increase of  $\text{PM}_{2.5}$  is reflected by a reduction of the average life expectancy, since enhanced PM concentrations are the main harmful components impacting human health. So, comparing the reference 1970 situation with the 2010 one, globally a loss in average life expectancy is observed mainly for North America (4.7 months for USA and 1.4 months for Canada) and Europe (4.2 and 5 months for central and western Europe, respectively), while increased life expectancy is found for developing countries (4.8 months for Asia; see Fig. 10a). Considering the STAG\_ENERGY case, life expectancy increases in most of the world's regions (4.2 and 7.2 months for China and India, 1–2 months for African countries and 0.8 months for Latin America), while a loss in life expectancy is observed for the STAG\_TECH scenario versus the REF(2010). As expected, in STAG\_TECH a significant negative impact on life expectancy is found for western and central European countries (4–5 months, see also Fig. S6.3), where the impact of emission reduction measures is largest. Coherently, life expectancy decreases also for other industrialized countries, like Japan and USA (3.3 and 2 months) and to a lesser extent in developing and emerging countries. Major health benefits from emission reduction measures are observed in highly populated areas where PM

and ozone changes are large. It is obvious that the road transport sector, important in densely populated (urban) areas is contributing significantly to the health impact.  $\text{O}_3$  concentrations negatively influence crop growth, so the reduction in  $\text{O}_3$  concentrations observed for the REF(1970) vs. REF(2010) and STAG\_ENERGY vs. REF(2010) is reflected in a net positive crop yield (global gain equal to 15.5 and 28 million Mt for the two scenarios, respectively). Conversely, as shown in Fig. 10b and more in detail in Fig. S6.3, also here, the emission control measures on vehicles are 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 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



**Figure 10.** Relative change between the scenarios (STAG\_ENERGY, STAG\_TECH and REF(1970)) and the reference case (REF(2010)): regional change in (a) PM<sub>2.5</sub> ( $\mu\text{g m}^{-3}$ ) and associated life expectancy (months) and (b) in ozone mixing ratios (ppb) and associated impacts on crops. Note that the same color scales are used for deltaPM<sub>2.5</sub> and deltaO<sub>3</sub> (positive delta are associated with red colors representing bad impacts, while negative delta are in green colors representing improvements). Opposite color scales are applied for change in life expectancy and crop yield compared to the delta one (positive values are reported in green and negative values in red representing good and bad impacts, respectively).

Europe and the world over the last 40 years has been investigated. Our analysis looks back to 1970, when the first European air quality directive was introduced and compares with 2010, the last year with reliable statistical data availability. In addition to our reference EDGARv4.3.1 reference emissions, we introduced two retrospective scenarios in order to analyze separately the impact of concurrent factors on 2010 emission levels. Specifically, the STAG\_ENERGY scenario evaluates the change in energy consumption, energy efficiency and fuel shift, and the STAG\_TECH scenario evaluates the change in technologies with implementation of abatement measures. The two scenarios present a range of emissions, lower and higher than the reference case for the year 2010, which assess the specific role and impact of EU legislation on air quality, of technology development and of fuel use. The story told by these scenarios can be informative for designing multi-pollutant abatement policies in emerging economies. Here we focus on the emissions of the most relevant pollutants (SO<sub>2</sub>, NO<sub>x</sub>, CO, NMVOCs and NH<sub>3</sub>) and of particulate matter (PM<sub>10</sub>, PM<sub>2.5</sub>, BC, OC) af-

fecting air quality at global and European levels. Global REF 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 those implemented in industrialized countries. For example, European SO<sub>2</sub> 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 assess the progress leading to the 2010 emission levels for three key sectors. For the power and manufacturing industry sectors, the increased fuel consumption 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 volumes, the overall transport sector 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 partly due to the shift from petrol to diesel passenger cars (emitting less CO, but more particles and NO<sub>x</sub>). Therefore, our study indicates that a variety of EU air quality policies since 1970 have avoided a dramatic deterioration of air quality in Europe and beyond. For example, fuel quality directives (sulfur), were among the most influential policies impacting air quality globally, e.g., 88 % reduction of SO<sub>2</sub>, while the EURO norms for vehicles led to a 50 % reduction of PM<sub>2.5</sub> from global road transport exhaust emissions. In contrast, the global increased energy consumption in 2010 compared to 1970 off-set between 40 and 75 % of the emissions reductions by the technological progress with end-of-pipe abatement and less carbon-intensive fuel use. 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 air quality policies and change in technologies 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 3–12 ppb in several world regions (reducing in the order of 10 % of the regional average annual O<sub>3</sub> levels). We estimate that EU policies increased life expectancy 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, contributing to up to 8.3 million Mt increased crop yield, which corresponds to 0.3 % of the present world production of maize, wheat, rice and soybeans.

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## References

- 93/12/EEC: Council Directive 93/12/EEC of 23 March 1993 relating to the sulfur content of certain liquid fuels, 1993.  
 98/70/EC: Directive 98/70/EC of the European Parliament and of the Council of 13 October 1998 relating to the quality of petrol and diesel fuels and amending Council Directive 93/12/EEC, 1998.  
 1999/32/EC: Directive 1999/32/EC on reduction of sulfur content of certain liquid fuels, 1999.

- 2001/80/EC: Directive 2001/80/EC on the limitation of emissions of certain pollutants into the air from Large Combustion Plants, 2001.  
 2008/50/EC: Directive 2008/50/EC on ambient air quality and cleaner air for Europe, 2008.  
 2010/75/EU: Directive 2010/75/EU of the European Parliament and of the Council of 24 November 2010 on industrial emissions (integrated pollution prevention and control), 2010.  
 Amann, M., Klimont, Z., and Wagner, F.: Regional and global emissions of air pollutants: recent trends and future scenarios, *Annu. Rev. Env. Resour.*, 38, 31–55, doi:10.1146/annurev-environ-052912-173303, 2013.  
 Anderson, J., Thundiyil, J., and Stolbach, A.: Clearing the air: a review of the effects of particulate matter air pollution on human health, *J. Med. Toxicol.*, 8, 166–175, doi:10.1007/s13181-011-0203-1, 2012.  
 Anenberg, S. C., West, J. J., Fiore, A. M., Jaffe, D. A., Prather, M. J., Bergmann, D., Cuvelier, K., Dentener, F. J., Duncan, B. N., Gauss, M., Hess, P., Jonson, J. E., Lupu, A., MacKenzie, I. A., Marmer, E., Park, R. J., Sanderson, M. G., Schultz, M., Shindell, D. T., Szopa, S., Vivanco, M. G., Wild, O., and Zeng, G.: Intercontinental impacts of ozone pollution on human mortality, *Environ. Sci. Technol.*, 43, 6482–6487, doi:10.1021/es900518z, 2009.  
 Burnett, R. T., Pope, C. A., Ezzati, M., Olives, C., Lim, S. S., Mehta, S., Shin, H. H., Hwashin, H., Singh, G., Hubbell, B., Brauer, M., Anderson, H. R., Smith, K. R., Balmes, J. R., Bruce, N. G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M. C., Gapstur, S. M., Diver, W. R., and Cohen, A.: An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure, *Environ. Health Persp.*, 122, 397–403, doi:10.1289/ehp.1307049, 2014.  
 Cofala, J., Amann, M., Klimont, Z., Kupiainen, K., and Höglund-Isaksson, L.: Scenarios of global anthropogenic emissions of air pollutants and methane until 2030, *Atmos. Environ.*, 41, 8486–8499, doi:10.1016/j.atmosenv.2007.07.010, 2007.  
 Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., Van Dingenen, R., and Granier, C.: Forty years of improvements in European air quality: the role of EU policy-industry interplay, *Atmos. Chem. Phys. Discuss.*, accepted, 15, 20245–20285, doi:10.5194/acpd-15-20245-2015, 2015.  
 Crippa, M., Janssens-Maenhout, G., Guizzardi, D., and Galmarini, S.: EU effect: exporting emission regulations through global market economy, *J. Environ. Manage.*, submitted, 2016.  
 CSC, Chinese State Council: Atmospheric Pollution Prevention and Control Action Plan, available at: [http://www.gov.cn/zwggk/2013-09/12/content\\_2486773.htm](http://www.gov.cn/zwggk/2013-09/12/content_2486773.htm) (last access: 20 July 2015), 2013 (in Chinese).  
 Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, *Atmos. Chem. Phys.*, 6, 4321–4344, doi:10.5194/acp-6-4321-2006, 2006.  
 EPA: Light-Duty Automotive Technology, Carbon Dioxide Emissions, and Fuel Economy Trends: 1975 Through 2013, EPA-420-R-13-011, 2013.

- FAO: Food and Agricultural Organization of the United Nations: Statistics Division, Livestock, Crop and Fertilizer data, available at: <http://faostat.fao.org>, and Geonetwork Digital Soil Map of the World, available at: <http://www.fao.org/geonetwork/srv/en/metadata.show?id=14116&currTab=simple> (last access: 20 July 2015), 2012.
- Fenger, J.: Air pollution in the last 50 years – from local to global, *Atmos. Environ.*, 43, 13–22, doi:10.1016/j.atmosenv.2008.09.061, 2009.
- Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., Denier van der Gon, H., Frost, G., Heil, A., Kaiser, J., Kinne, S., Klimont, Z., Kloster, S., Lamarque, J.-F., Liousse, C., Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J.-C., Riahi, K., Schultz, M., Smith, S., Thompson, A., Aardenne, J., Werf, G., and Vuuren, D.: Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period, *Climatic Change*, 109, 163–190, doi:10.1007/s10584-011-0154-1, 2011.
- GBD: Global Burden of Disease, available at: <http://www.healthdata.org/gbd>, (last access: 18 January 2016), 2010.
- IEA: Energy Statistics of OECD and Non-OECD Countries (IEA/EXD/ECD3), available at: <http://data.iea.org> (last access: 20 July 2015), 2014.
- IPCC, Intergovernmental Panel on Climate Change: Fifth Assessment Report, available at: <http://www.ipcc.ch/> (last access: 20 July 2015), 2013.
- Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP\_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, *Atmos. Chem. Phys.*, 15, 11411–11432, doi:10.5194/acp-15-11411-2015, 2015.
- Jerrett, M., Burnett, R. T., Pope, C. A., Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E., and Thun, M.: Long-term ozone exposure and mortality, *New Engl. J. Med.*, 360, 1085–1095, doi:10.1056/NEJMoa0803894, 2009.
- Klimont, Z., Smith, S. J., and Cofala, J.: The last decade of global anthropogenic sulphur dioxide: 2000–2011 emissions, *Environ. Res. Lett.*, 8, 014003, doi:10.1088/1748-9326/8/1/014003, 2013.
- 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, doi:10.5194/acp-5-417-2005, 2005.
- Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2, *Atmos. Chem. Phys.*, 13, 11019–11058, doi:10.5194/acp-13-11019-2013, 2013.
- Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, *Atmos. Chem. Phys.*, 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.
- Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, *Nature*, 525, 367–371, doi:10.1038/nature15371, 2015.
- Monks, P. S., Granier, C., Fuzzi, S., Stohl, A., Williams, M. L., Akimoto, H., Amann, M., Baklanov, A., Baltensperger, U., Bey, I., Blake, N., Blake, R. S., Carslaw, K., Cooper, O. R., Dentener, F., Fowler, D., Fragkou, E., Frost, G. J., Generoso, S., Ginoux, P., Grewe, V., Guenther, A., Hansson, H. C., Henne, S., Hjorth, J., Hofzumahaus, A., Huntrieser, H., Isaksen, I. S. A., Jenkin, M. E., Kaiser, J., Kanakidou, M., Klimont, Z., Kulmala, M., Laj, P., Lawrence, M. G., Lee, J. D., Liousse, C., Maione, M., McFiggans, G., Metzger, A., Mieville, A., Moussiopoulos, N., Orlando, J. J., O'Dowd, C. D., Palmer, P. I., Parrish, D. D., Petzold, A., Platt, U., Pöschl, U., Prévôt, A. S. H., Reeves, C. E., Reimann, S., Rudich, Y., Sellegri, K., Steinbrecher, R., Simpson, D., ten Brink, H., Theloke, J., van der Werf, G. R., Vautard, R., Vestreng, V., Vlachokostas, C., and von Glasow, R.: Atmospheric composition change – global and regional air quality, *Atmos. Environ.*, 43, 5268–5350, doi:10.1016/j.atmosenv.2009.08.021, 2009.
- Paruolo, P., Murphy, B., and Janssens-Maenhout, G.: Do emissions and income have a common trend? A country-specific, time-series, global analysis, 1970–2008, *Stoch. Env. Res. Risk A.*, 29, 93–107, doi:10.1007/s00477-014-0929-9, 2015.
- Pope, C. A. and Dockery, D. W.: Health effects of fine particulate air pollution: lines that connect, *JAPCA J. Air Waste Ma.*, 56, 709–742, doi:10.1080/10473289.2006.10464485, 2006.
- Rafaj, P., Amann, M., Siri, J., and Wuester, H.: Changes in European greenhouse gas and air pollutant emissions 1960–2010: decomposition of determining factors, *Climatic Change*, 124, 477–504, doi:10.1007/s10584-013-0826-0, 2014.
- Tavoni, M., Kriegler, E., Riahi, K., van Vuuren, D., Petermann, N., Jewell, J., Herreras Martinez, S., Rao, S., van Sluisveld, M., Bowen, A., Cherp, A., Calvin, K., Marangoni, G., McCollum, D., and van der Zwaan, B.: LIMITS: Limiting Global Warming to 2 °C, Policy findings from Durban Platform scenario analyses, FP7-LIMITS publication, European Union, 2014.
- UNEP/WMO: Integrated Assessment of Black Carbon and Tropospheric Ozone, ISO 14 001 : 2004, UNON/Publishing Services Section, Nairobi, 2011.
- Van Dingenen, R., Dentener, F. J., Raes, F., Krol, M. C., Emberson, L., and Cofala, J.: The global impact of ozone on agricultural crop yields under current and future air quality legislation, *Atmos. Environ.*, 43, 604–618, doi:10.1016/j.atmosenv.2008.10.033, 2009.
- Van Dingenen, R., Dentener, F., and Leitao, J.: TM5-FASST: a global atmospheric source receptor model for rapid impact analysis of emission changes on air quality and short-lived climate pollutants, in preparation, 2015.
- WHO: Review of Evidence on Health Aspects of Air Pollution – REVIHAAP, Technical report, The WHO European Centre for Environment and Health, WHO Regional Office for Europe, Bonn, 2013.