Air-quality in the mid-21st century for the city of Paris under two climate scenarios; from regional to local scale.

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Abstract

Ozone and $PM_{2.5}$ concentrations over the city of Paris are modeled with the CHIMERE air-quality model at 4km x 4km horizontal resolution for two future emission scenarios. A high resolution (1x1 km) emission projection until 2020 for the greater Paris region is developed by local experts (AIRPARIF) and is further extended to year 2050 based on regional scale emission projections developed by the Global Energy Assessment. Model evaluation is performed based on a 10-year control simulation. Ozone is in very good agreement with measurements while $PM_{2.5}$ is underestimated by 20% over the urban area mainly due to a large wet bias in wintertime precipitation. A significant increase of maximum ozone relative to present time levels over Paris is modeled under the "business as usual" scenario (+7 ppb) while a more optimistic mitigation scenario leads to moderate ozone decrease (-3.5 ppb) in year 2050. These results are substantially different to previous regional scale projections where 2050 ozone is found to decrease under both future scenarios. A sensitivity analysis showed that this difference is due to the fact that ozone formation over Paris at the current, urban scale study, is driven by VOC-limited chemistry, whereas at the regional scale ozone formation occurs under NO_x-sensitive conditions. This

explains why the sharp NO_x reductions implemented in the future scenarios have a different effect on ozone projections at different scales. In rural areas projections at both scales yield similar results showing that the longer time-scale processes of emission transport and ozone formation are less sensitive to model resolution. $PM_{2.5}$ concentrations decrease by 78% and 89% under “business as usual” and “mitigation” scenarios respectively compared to present time period. The reduction is much more prominent over the urban part of the domain due to the effective reductions of road transport and residential emissions resulting in the smoothing of the large urban increment modelled in the control simulation.

1 Introduction

Climate change can affect air quality through a number of mechanisms related to meteorological variables such as temperature, humidity, precipitation, solar radiation, wind speed and the planetary boundary layer height. If the effects of atmospheric pollutants (greenhouse gasses, other gaseous species and aerosols) on climate change have been extensively investigated over the last decades (IPCC, 2001, 2007) the impact of these emissions and changed climate on air quality has only raised interest during the last few years and many issues remain open. Most of these studies use global chemistry-transport models (CTMs) to study the impact of changing climate on tropospheric ozone at either global (Brasseur et al., 1998; Liao et al., 2006; Prather et al., 2003; Szopa and Hauglustaine, 2007) or regional scale (Murazaki and Hess, 2006; Szopa et al., 2006). The resolution of these studies, typically a few hundreds of kilometers, is insufficient to capture the high spatial variability of air pollution due to the short lifetime of tropospheric ozone and particulate matter (in the range of some hours up to a few days) (Seinfeld and Pandis, 2006) as well as to the sharp horizontal gradients of anthropogenic emissions over urban areas. Moreover, the chemistry-climate models used in such large-scale studies suffer from a simplistic representation of regional scale chemistry.

Only few, recent, applications study the impact of changing climate on air quality using regional CTMs. These models include more sophisticated chemistry at typical resolutions of tenths of kilometers and therefore provide a better understanding of the underlying mechanisms at regional scale. The first documented modeling study on the impact of future conditions at regional scale is that of Hogrefe et al. (2004) while at the same time Knowlton et al. (2004) took a step forward and performed a health impact assessment study for the greater New York region.

Since then several researchers performed similar regional applications in order to derive future predictions of ozone and/or aerosols using offline climate-chemistry models (Colette et al., 2013; Kelly et al., 2012; Lam et al., 2011; Langner et al., 2005; Nolte et al., 2008; Steiner et al., 2006; Szopa and Hauglustaine, 2007; Tagaris et al., 2009), on-line climate-chemistry models (Forkel and Knoche, 2006, 2007) or multi-model studies (Colette et al., 2012; Langner et al., 2012).

An open research challenge today is the impact of climate change on air-quality at urban scale. Enhanced health effects due to increasing urbanization (UNFPA, 2007) should be addressed primarily at such scale especially given that the efforts to mitigate air-pollution are more intense in areas where the largest health benefits are observed (Riahi et al., 2011). The common approach to study the effects of climate change and emissions at city level is to use regional scale CTMs. However the resolution of current model studies remains too coarse to represent the high spatiotemporal variability of pollutants at urban scale. Regional scale emission inventories fail to represent the plethora of emission activities in terms of e.g. the underlying technologies used within a specific source sector or to take into account the habitual patterns of the population at large cities (Markakis et al., 2010, 2012). The substantial part of the published work uses emission projections based on scenarios developed to represent changes at global scale and are rarely well suited for local scale assessment e.g. they lack the detailed representation of all emission activities at a finer scale. For example, the more recent Representative Concentration Pathways (RCPs) (van Vuuren et al., 2011) developed to support modeling activities for the new assessment of the Intergovernmental Panel on Climate Change (IPCC) incorporate a diversity of radiative forcing scenarios that may be suitable for large scale chemistry-climate models but not appropriate to represent emission trends for specific cities and therefore regional scale air-quality assessment (Butler et al., 2012). Grid resolution is another key issue for urban scale modeling as shown by previous research (Arunachalam et al., 2006; Cohan et al., 2006; Valari and Menut, 2008). The assumption of instantaneous mixing of emissions within the volume of the regional scale CTMs' grid cells is unsuitable to represent local scale chemistry (such as ozone titration) when the grid-cell size is too large compared to the actual size of the emitting areas. It has been shown previously that use of coarse resolution leads to underprediction of daily ozone maxima and to overprediction of daily average ozone levels (Arunachalam et al., 2006; Tie et al., 2010; Vautard et al., 2007). Valin et al. (2011) show that grid resolution finer than ~10 km is often necessary to capture ozone chemistry near areas with large NO_x emissions. Due to the non-linearity of ozone chemistry the inability of large grid sizes to differentiate urban and rural areas can have a profound effect on the simulated ozone concentrations (Silman et al., 1990). Forkel and Knoche (2007) found that the increase of future maximum ozone concentrations in Europe occurred in areas where high mixing ratios of NO_x coincided with increased isoprene emissions, indicating that the failure of coarser resolutions to efficiently distinguish between urban and rural areas would result in overestimation of future ozone in cities near to high isoprene emitting sources. The current challenge is to develop realistic long-term emission projections at a city-scale and bridge the gap between local, regional and global scale modeling.

The aim of our study is to develop mid-21st century horizon air quality projections over the greater Paris area under two consistent emission and climate scenarios (10-year continuous simulation). To the authors knowledge this is the first time where a 10-year air-quality projection under climate and city level emission changes has been conducted at urban scale and resolution as fine as 4 km. Local emission projections used in this study are compiled by merging several pieces of information: i) a high resolution (i.e. 1 km) city specific emission inventory developed by AIRPARIF (present time emissions); ii) a city-scale short-term projection (until 2020) considering air-quality policies already in place and planned for the city for the next years iii) two mid-21st century regional scale emission scenarios including both climate and regional air quality policies developed by the Global Energy Assessment (GEA) (Riahi et al., 2012). The "reference" (REF) scenario is consistent with long-term climate outcomes of the RCP-8.5 adopting all current and planned air quality legislation until 2030 and assumes that no climate policies are implemented thereafter. The "mitigation" (MIT) scenario is consistent with long-term climate outcomes of the RCP-2.6 and additionally assumes stringent climate mitigation policies. The final fine scale, mid-21st horizon emission inventory for the greater Paris area, the Ile-de-France region (IdF), was obtained by scaling the 2020 high resolution emission inventory to the year 2050 using the regional scale GEA scenarios.

After describing the methodology in Section 2, an evaluation of the modelling system is described in Section 3 and scenario results are presented in Section 4. Our conclusions are discussed in Section 5.

2 Materials and methods

The Ile-de-France region (IdF) is located at 1.25–3.58° east and 47.89–49.45° north with a population of approximately 11.7 million, more than two million of which live in the city of Paris (Fig. 1). The area is situated away from the coast and is characterized by uniform and low topography, not exceeding 200 m above sea level.

An “off-line” modeling approach is used to assess air-quality in the study region as a response to climate and emission changes. For both climate and air-quality simulations we use a dynamical downscaling approach consisting of two one-way nesting steps: from global to regional scale over Europe and from regional to local scale over the IdF region. IPSL-CM5A-MR global circulation model (Dufresne et al., 2013) is used to derive future projections of the main climate drivers (temperature, solar radiation etc.) using a set of available global greenhouse gas emission scenarios (RCP-2.6 and RCP-8.5).

Global climate output is downscaled with the Weather Research and Forecasting (WRF) mesoscale climate model (Skamarock and Klemp, 2008) at two steps: first over a 50km resolution grid over Europe and then on a 10 km resolution grid over France using one-way nesting. We drive the CTM over IdF using the 10km meteorology acknowledging both the computation overhead of a refined meteorology and the results of previous work over the same region using the CHIMERE CTM (Menut et al., 2005a; Valari and Menut., 2008). More specifically Menut et al. (2005a) showed that apart from the coastal areas where a refined meteorology improved air-quality modelling results, in the rest of France ozone peaks were better captured with lower resolution meteorological input. Valari and Menut (2008) showed that a refined meteorological input gives similar results for ozone and that model performance is much more sensitive to the resolution of emissions than to meteorology. These results suggest that in areas having the geographical characteristics of the greater Paris area (flat topography at great distance from any mountains or the ocean) increasing the resolution of the meteorological input does not necessarily improve the results of the chemistry-transport modelling. On the other hand Flagg and Taylor (2011) investigated the impact of the resolution of the surface layer input data on modelling results of high resolution simulations in an urban coastal environment and found that these results were quite dependent on that resolution. There are indications that one needs to go to very high resolution in order to capture the urban heat island circulation (LeRoyer et al. 2014) and the effects of the surface layer changes (Flagg and Taylor, 2008) in an urban meteorological simulation but this analysis is beyond the scope of the present work.

Air-quality data are downscaled with the CHIMERE model (Menut et al., 2013) a detailed description of which can be found at: http://www.lmd.polytechnique.fr/chimere. CHIMERE is an off-line chemistry-transport model (CTM), which models atmospheric chemistry and transport, forced by anthropogenic emissions, biogenic emissions, a meteorological simulation and boundary conditions. It is a cartesian-mesh grid model including gas-phase, solid-phase and aqueous chemistry, biogenic emissions modeling depending on meteorology with the MEGAN model (Guenther et al., 2006), dust emissions (Menut et al., 2005b) and resuspension (Vautard et al., 2005). Gas-phase chemistry is based on the MELCHIOR mechanism (Lattuati, 1997) which includes more than 300 reactions of 80 gaseous species. The aerosols model species are sulfates, nitrates, ammonium, organic and black carbon and sea-salt (Bessagnet et al., 2010) and the gas-particle partitioning of the ensemble Sulfate/Nitrate/Ammonium is treated by the ISORROPIA code (Nenes et al., 1998) implemented on-line in CHIMERE.

Initial and boundary conditions are taken from global scale concentrations modelled with the coupled LMDz-INCA global chemistry model (Hauglustaine et al., 2004; Szopa et al., 2012) and are then downscaled with CHIMERE using two-level one-way nesting first at a 0.5º resolution grid (~50 km) over Europe (Colette et al., 2013) and then at the 4km resolution grid over the IdF region (local scale runs). For more details on the global and regional modeling exercises the reader is referred to Hauglustaine et al. (2014) and Colette et al. (2013) respectively. We only note that the global scale simulations done with the LMDz-INCA model has a slightly different chemical speciation from that of CHIMERE. Finally Table 1 presents the basic configuration of the global, regional and local simulations used in the present study.

We performed three sets of 10-year runs at local scale: a continuous control run (CTL) from 1995 to 2004 representing present time air-quality and two continuous runs over the 2045-2054 decade representing air-quality projections to the mid-21st century horizon under two different pathways of climate and emissions. To minimize the effect of inter-annual variability and increase the statistical significance of model output longer term simulations should have been performed. However, given the high resolution of the modeling exercise the computational cost of longer simulations was considered too high for the present study.

2.1 Models setup

The modeling domain has a horizontal resolution of 4 km and consists of 39 grid cells in the west-east direction and 32 grid cells in the north-south direction, thus covering a total region of 156 x 128 km^2. Twelve σ-p hybrid vertical layers were used to represent the atmospheric column from the surface up to approximately 500hPa (~5.5 km) with the thickness of the first layer being 8 m. Simulations were performed with a time step of 5 minutes. Hourly meteorological fields necessary for CHIMERE were modeled with WRF model v3.4. WRF simulations were carried out on a 10 km resolution grid of 90x85 cells. Vertically the domain is divided into 31 σ-layers extending up to 55hPa (∼20 km). The time integration step was set to 1 minute. The physical options used for these simulations are the WRF Single Moment 6-class microphysics scheme (Hong and Lim, 2006), RRTM (rapid radiative transfer model) long-wave radiation scheme (Mlawer et al., 1997), Dudhia short-wave radiation scheme (Dudhia, 1989), NOAH land surface model (Chen and Dudhia, 2001), Asymmetric Convective Model version 2 Planetary Boundary Layer scheme (Pleim, 2007) and Kain-Fritsch cumulus parameterization scheme (Kain, 2004).

2.2 Data and metrics for model evaluation

CHIMERE model evaluation has been performed in numerous studies at both regional (see e.g. Solazzo et al., 2013a, 2013b and references therein) and urban scales (Hodzic et al., 2005; Vautard et al., 2007). However, due to the climate component of the study our modeling setup requires a different evaluation framework. We use 29 surface monitors of the local air-quality network AIRPARIF classified as urban (17 sites), suburban (4 sites) and rural (8 sites). Only monitoring sites with more than 70% of available data (hourly values) through the 10-year period were considered for the evaluation process. Observations are conducted at a height ranging from 3 to 8m from the surface, which makes the first model layer concentrations directly comparable with the observations.

Given that adverse health impact is mainly correlated to daytime ozone levels in the present framework we focus on the evaluation of daytime and maximum ozone only. A variety of metrics are used to evaluate model performance. For ozone the widely used mean normalized bias (MNB) and mean normalized gross error (MNGE) are estimated. US EPA suggests that MNB for modeled ozone concentrations should lie within the +-15% range and MNGE not exceeding 35%. Regarding $PM_{2.5}$ the Mean Fractional Bias (MFB) and the Mean Fractional Error (MFE) are used. A literature review of targeted range of values for fine particles can be found in

EPA (2007). The narrowest of the reported ranges suggests that for modeled $PM_{2.5}$ MFB should fall within the +-30% range and MFE not exceeding 50%.

We extract these metrics from the daytime concentrations values and not the decade average bearing in mind that this is not typical for runs forced by climate simulationsbut for operational forecast evaluation. Here we do not use re-analyses to force the regional CTM but global and regional climate simulations. Consequently one should expect lower scores than those yielded by an air-quality forecast simulation, especially in the presence of climate biases (Colette et al., 2013; Menut et al., 2013). Since this work is original in its concept we aim to evaluate whether the urban scale setup is sufficiently realistic but utilizing metrics that are timely averaged on a scale finer than a climatological one.

2.3 Climate and emission scenarios

Two long-term scenarios are used in the global-scale simulations of future climate conditions: RCP-8.5 and RCP-2.6. RCP-8.5 (Riahi et al., 2011) is a "reference" type scenario with no mitigation for greenhouse gases leading to a global radiative forcing of 8.5 W m^{-2} by the end of the century. RCP-2.6 (van Vuuren et al., 2011a) leads to a global radiative forcing of 2.6 W m^{-2} by the end of the century (2100), consistent with the goal of limiting the increase of global average temperature due to human activity to 2ºC. Both RCP scenarios include century-long estimates of air pollutant emissions, including aerosols, and were used as input in the LMDz-INCA global chemistry model.

Present time emission estimates for the IdF region are available in hourly intervals, with a spatial resolution of 1x1 km. The emission dataset was compiled by local experts using a variety of city-specific information integrating a number of anthropogenic activities in the IdF region (AIRPARIF, 2012). The spatial allocation of emissions is completed with proxies such as high-resolution population maps, road network and the location of industrial units. It includes emissions of CO, NO_x, Non-methane Volatile Organic Compounds (NMVOCs), SO_2, PM_{10} and $PM_{2.5}$ with a monthly, weekly and diurnal (source specific) temporal resolution. The distribution of emissions among different activity sectors reveals that in the IdF region the principal emitter of NO_x, on annual basis, is the road transport sector (50%), for NMVOCs the use of solvents (46%) and for fine particles the residential sector (40%). Annual emissions rates within the simulated decade were kept constant; only the vertical distribution of point source emissions across model layers varies in time since it depends on several meteorological variables such as temperature and wind (plume-rise algorithm) (Scire et al., 1990). Finally the 1 km resolution emissions were aggregated to the 4 km resolution grid.

Mid-21st century (year 2050) emission projections for IdF incorporate local scale information, including the implementation of planed policies. The short-term component consists of the local scale 2020 emission projection compiled by AIRPARIF in the framework of the “Plan de Protection de l’Atmosphere d’Ile de France” (PPA) and with the support of the “Direction Regionale et Interdepartementale de l’Environnement et de l’Energie d’Ile de France” (DRIEE-IF). Emissions for 2020 correspond to a “business as usual” scenario assuming the implementation of all regulatory measures planned by the PPA.

The long-term component of the projection is established by linking two primary sources of information: (i) the aforementioned 2020 AIRPARIF inventory and (ii) a set of scenarios from the recently published Global Energy Assessment (GEA) (Riahi et al., 2012) to extend projections until 2050. The GEA scenarios, while consistent with similar long-term climate outcomes as the RCPs, include a more detailed representation of short-term air quality legislations from the GAINS model (Amann et al., 2011). Outputs are based on the MESSAGE model (Riahi et al., 2007) and include estimates of energy and greenhouse gas emissions for 11 global regions while air pollutants are further available at 0.5^o x 0.5^o resolution estimated based on inventory data described in Granier et al. (2011) and Lamarque et al. (2010) and an exposure-driven algorithm for the downscaling of the regional air-pollutant emission projections. The GEA scenarios have been used to estimate global health impacts of outdoor air pollution (Rao et al., 2012, 2013) as well as for regional impacts analysis (Colette et al., 2012, 2013). Two GEA scenarios are used in our analysis: i) a “reference” (REF) scenario (consistent with long-term climate outcomes of the RCP-8.5) which adopts all current and planned air quality legislation until 2030 and assumes that no climate policies are implemented thereafter ii) a “mitigation” (MIT) scenario (consistent with long-term climate outcomes of the RCP-2.6) which additionally assumes stringent climate mitigation policies consistent with a target of 2^oC global warming by the end of the century (2100). Thus the MIT scenario can be used to assess the co-benefits of climate induced strategies. We derived national-wide annual totals (for France) in 2020 and 2050 for a number of pollutants from the GEA scenarios. 2050 over 2020 scaling ratios were calculated for each pollutant for each source sector accounted for in the GEA dataset. The resulting ratios are used as coefficients to scale the local 2020 AIRPARIF inventory without modifying the spatial and temporal patterns.

We decided to derive scaling ratios based on national totals and not from the GEA 0.5º resolution grid to avoid adopting the relatively simplistic approaches of the GEA downscaling algorithm for the distribution of emissions at the finer scale. Thus, our approach benefits from the highly resolved emission variability over Paris from the AIRPARIF inventory but it must be noted that at the same time we inherently assume the energy transition over the 2020 to 2050 period according to the REF and MIT scenarios. This is particularly relevant as the GEA scenarios implement significant improvements in energy efficiency and a complete implementation and continuation of air-quality policies in Europe. Thus we observe significant reductions in emissions for the IdF region in 2050 (Table 2). While using different scenarios would lead to alternative estimates of 2050 emissions, it is important to note that significant technological change in the energy systems would likely mean that air pollutant emissions in Europe would decline over the long-term in any case, although the exact distribution of such reductions is uncertain.

3 Model evaluation based on the control simulation (present time)

3.1 Meteorology

It is beyond the purpose of this study to provide an in-depth meteorology evaluation, however to build confidence in the meteorological fields used as input for our air-quality simulations we present a short model evaluation based on the CTL run.

WRF meteorology for the CTL simulation ran over a 10 km resolution grid. Model results are compared to surface observations from 7 meteorological stations found inside the IdF region. Only one of these monitors (MONT) is actually inside the city of Paris. The mean wintertime (DJF) and summertime (JJA) modeled and observed daily average values were calculated for 4 different variables relevant to air quality: 2m temperature, wind speed at 10m, relative humidity and total precipitation (Table 3). A warm bias during summer reaches +0.8ºC in the city center (MONT station). This is bound to lead to enhanced ozone formation due to the thermal decomposition of PAN releasing NO_x (Sillman and Samson, 1995). Modeled wind speeds are higher than the observed ones both during winter and summer, a bias consistent with previous studies (see e.g. Jimenez et al. (2012) for WRF or Vautard et al. (2012) for other models). This is expected to enhance pollutants' dispersion and lead to less frequent stagnation episodes. The model underestimates summertime relative humidity (bias=-13.3%). A systematic wet bias in wintertime precipitation is observed in the city reaching +26.5%. This is expected to lead to underestimation of PM levels through rain scavenging (Fiore et al., 2012).

3.2 Ozone concentrations

Scatter plots in Fig. 2 compare modeled ozone with surface measurements. Mean daytime surface ozone and O_x (NO_{2}+O_3) (averaged over the April-August period) as well as the daily maximum of 8-hour running means (MD8hr) from the urban stations are shown. Modeled daytime ozone is in very good agreement with measurements over the urban cluster with a bias of only +0.25 ppb (0.8% overestimation). Note that the spread of ozone bias among individual stations is also small. O_x is often used as an indicator of ozone photochemical build-up because it rules out titration process. Therefore, the low O_x bias indicates that local scale modeling reproduces sufficiently both urban titration (daytime) and photochemical formation. NO_2 bias (not shown) is also low (-0.32 ppb). Model results for ozone are considered satisfactory given the nature of these runs (forced by climate simulations and not forecast meteorology as discussed in Sect. 2.2). The MNB (+16.5%) and MNGE (+40.6%) are outside but near the US EPA targeted ranges (+-15% and +35% respectively). For downtown Paris sites (not shown) MNB is +10.9% and MNGE is equal to +39.8%. However, the model underestimates MD8hr by -7.8% (bias =-3.2 ppb). The 95th percentile (not shown) of observed and modeled ozone daily maxima differ by 13.8 ppb (-20.1%) indicating that the model fails to reproduce ozone levels under extreme photochemical episodes which in any case are produced in timely short periods of very specific meteorological conditions characterized by stagnated air masses and low vertical mixing favoring ozone build-up. The meteorological input used in our local simulation represents poorly the observed wind speeds which are overestimated significantly (Sect. 3.1). This affects stagnation (Jacob and Winner, 2009; Vautard et al., 2007) but also vertical diffusivity through an increased boundary layer height. At the same time, the climate model suffers from a cold and overcast bias (Colette et al., 2013) that inhibits the emission of biogenic precursors and ozone buildup.

Overestimation of mean daytime ozone is observed in all suburban and rural stations (+10.9% and +11.3% respectively) even though O_x is in relatively good agreement with measurements.

This points to an underestimation of NO_2 and titration is represented less faithfully. Ozone overestimation is due to some extent to the boundary conditions. This is supported by the fact that both upwind and downwind rural stations show the same level of overestimation portraying a spatially symmetric influence of boundary conditions (not shown). In the urban areas the strong titration probably depletes the excess of ozone from the boundaries pointing to a possible exaggeration of urban titration in the model. MNB and MNGE scores for suburban and rural sites (Fig. 2) in general lie close but outside the US EPA targeted ranges.

3.3 Fine particulate matter

Modeled $PM_{2.5}$ surface concentrations are compared to all available measurements (i.e four urban sites only one of which located in Paris). Results for wintertime (DJF), summertime (JJA) and on annual basis are shown in Fig. 3. The model underestimates $PM_{2.5}$ by 20% in wintertime (-14% at the Paris site). This underestimation could be attributed to the significant positive bias in modeled precipitation and wind speeds (Sect. 3.1). Another explanation could be that the monthly temporal profiles used to distribute the annual emissions of residential heating are based on mean temperature variations. This could lead to strong underestimation of emissions in wintertime. The analysis of the annual model results yields even higher underestimation for both Paris site and domain-wide values (-18.9% and -26.6% respectively). The poorer model performance during summertime suggests a possible underestimation of summertime emissions in the local inventory. Vehicle-induced resuspension of particles might be a significant missing source, especially during the drier summertime months (Schaap et al., 2009 and references therein). $PM_{2.5}$ underestimation, to some extent, is due to a poor representation of secondary organic aerosol (SOA) in the model (modeled SOA concentrations are <0.5% in wintertime and ~1.5% in summertime). This is a well-known shortcoming of regional CTMs (Simpson et al., 2007; Solazzo et al., 2012; Stern et al., 2007). In particular Hodzic et al. (2009; 2010) found significant underprediction of observed SOA concentrations modeled with CHIMERE in the framework of the MILAGRO campaign. Modeled meteorology (wind and precipitation overestimation) also contributes to the observed PM underestimation. Nevertheless, wintertime and annual metrics are within the US EPA targeted range (MFB within +-30% and MFE <50%).

3.4 Conclusions on model evaluation

The model is able to represent the main features of ozone photochemical cycle (ozone formation and titration) over the Ile-de-France region but fails to reproduce high ozone episodes. Urban features of ozone chemistry, such as ozone titration are resolved showing that the emission inventory is realistic. Therefore, this local scale modeling makes it possible to discern between urban, suburban and rural areas and provides a good representation of both ozone build-up and titration processes. The underestimation of $PM_{2.5}$ is attributed to errors in modeled meteorology (high precipitation and winds) and possibly to missing emission sources and chemical processes (secondary organic aerosol).

4 Future scenario analysis

4.1 Climate projections for the mid-21st century in Paris

Table 4 summarizes the projections of key meteorological variables. It is beyond the scope of this study to disentangle the effects of climate and emission changes on air-quality and therefore only a brief qualitative discussion is provided in this section.

Under the REF scenario, which assumes that greenhouse gases increase monotonically until the end of the century, the annual mean surface temperatures in the mid-21st horizon exhibits an overall domain-wide increase of +1.1 ºC (+1.0ºC over the city) compare to the present time period. Consequently, enhanced ozone formation is expected especially in the rural part of the domain due to increase of biogenic organic compounds (BVOCs). Monoterpenes are especially sensitive to temperature while isoprene to both temperature and sunlight. We do not observe any significant changes to short-wave radiation, RH or precipitation under the REF scenario.

A -0.5ºC decrease in the annual surface temperature compared to present time period is modeled under the MIT trajectory, with two particularly cold years (2051 and 2052) lowering the decade average. The inter-annual variability (standard deviation) is 2.54ºC around a decade average of 11.3ºC while under the REF scenario the variability is 0.8ºC around a decade average of 13ºC. The internal natural variability of temperature can be very large at regional scale (Deser et al., 2012), which stresses the need for longer simulation periods over which the inter-annual variability would be smoothed. Lower temperatures are expected to inhibit ozone formation while the drop of shortwave downward radiation by 16.6% relative to present will lead to less BVOCs in the rural areas. We observe a significant increase in total precipitation compared to the control simulation by a factor of 3.2 under the MIT pathway where, similarly to temperature, two particularly wet years (2050 and 2051) are simulated. Modeled precipitation is high only on a limited amount of days within those two years (standard deviation=0.73 around a decade average of 0.32). In the regional simulation from which boundaries were utilized to force the local runs, climate is also shown significantly wetter under MIT by a factor of 3.6.

4.2 Ozone projections

Mid-21st century ozone projections averaged over the April through August period are presented in Fig. 4 for the REF and the MIT scenarios. Panel a shows present time MD8hr levels (CTL simulation) whereas panels b and c provide differences between each of the REF and MIT scenarios and the CTL simulation. The CTL simulation is able to dissociate the fast ozone titration process from the longer scale photochemical build-up: low ozone levels are modeled over the city of Paris (36-38 ppb) due to titration by NO (road transport mainly) and higher levels (47-50 ppb) are modeled at the surrounding rural area due to photochemical formation.

Projections show an increase in maximum ozone (+7 ppb) over the city of Paris under the REF scenario relative to CTL and only a small decrease under the MIT scenario (-3.5 ppb). The average daytime ozone (not shown) also increases substantially in downtown Paris by approximately +10 ppb under REF. The results are consistent with the near future projection of Roustan et al. (2011) who also employed local scale emissions and modelled maximum ozone increase in Paris for the year 2020. Reductions in rural ozone are modelled under both scenarios: REF: -3.2 ppb and MIT: -13.5 ppb.

Previous studies have shown that anthropogenic emissions over urban areas are the driver of ozone concentrations compared to climate, boundary conditions and other influencing factors (Colette et al., 2013). Therefore, to gain further insight in the ozone response one should study the differences between present and future time ozone precursors' emissions. However emission changes under different chemical regimes may also lead to different ozone responses. For example, air-quality projections conducted at a 0.5ºx0.5º resolution grid over Europe forced with GEA emissions and the same meteorology as in our study found significant decrease compared to present time values in ozone concentrations over the Paris region at the 2050 horizon for both REF (-5 ppb) and MIT (-16 ppb) scenarios (Colette et al., 2013), in contrast to our findings showing increase in ozone under the REF scenario and only small decrease under the MIT scenario. We are interested to see whether this difference in the sign between the local and regional ozone projections could be explained by the photochemical regime under which ozone is produced in each simulation.

4.2.1 Analysis of the chemical regimes

It is known that areas under NO_x-sensitivity or VOC-sensitivity are more effectively mitigated when NO_x or NMVOCs emission reductions are implemented respectively (Beekman and Vautard, 2010). Previous modeling exercises using the CHIMERE model have shown that the ozone photochemistry in Paris is VOC-sensitive (Beekman and Derognat, 2003; Beekman and Vautard, 2010; Deguillaume et al., 2008).

In this experiment we perform a model sensitivity analysis in which anthropogenic NO_x and NMVOCs emissions let vary from a factor of one fourth up to two times their actual value with a step of 0.25 (all combinations of 8 different scenarios for both NO_x and NMVOCs). In total 64 simulations were performed for a typical summertime ozone episode. Emission changes were applied over the whole 3-dimensional domain. We conducted the same experiment twice: once using as initial local-scale emission inventory developed by AIRPARIF (LOC case) and once using the regional scale inventory (REG case). In order to isolate the effect of emissions only and exclude the potential impact of other resolution-related effects the sensitivity analysis for the REG realization was not performed in the native REG domain; instead we first downscaled the regional emission inventory on the 4km grid and then ran the regional scale simulation. Obviously, emission totals between the two simulations do not match and no direct comparison should be made based on these results. However, it is interesting to see how different emission starting points may lead to ozone production under different photochemical regimes and therefore to different future ozone projections. We note that present time NO_x/NMVOCs emission ratios within the ozone period over Paris are much higher in the LOC inventory than in the REG dataset (0.5 vs. 0.2). This ratio is a key factor for the modeling of ozone production. We chose to run the sensitivity analysis over a one-day, present time, summer ozone episode that emission fluxes and meteorological conditions favoring the photochemical ozone built-up are similar in most ozone events. We also note that while boundary conditions may impact local scale model predictions, we focus here on the impact of local emissions through the use of a common set of boundary conditions, to ensure that the differences arise from the sensitivity to local emissions.

Iso-contours of day ozone maxima concentrations as a function of NO_x and NMVOCs emission rates are given for the two simulations (isopleth plots of Fig. 5). Two cases are discussed separately: downtown chemistry (panels a and b) and in-plume chemistry (panels c and d). “In-plume” corresponds to the chemical reactions taking place when high concentrations of nitrous oxides coming from the city and NMVOCs from the rural areas help to accumulate ozone on the downwind site of the city. To extract “in-plume” results we identify at each hour of the simulated episode the grid cell with the maximum concentrations inside the domain. Downtown chemistry in the LOC simulation is mainly sensitive to NO_x changes with sharp ozone increases related to NO_x reductions. This is a typical ozone response near high NO_x emission sources where titration and removal of radicals by nitrogen dioxide are the main drivers of ozone concentrations (VOC-sensitive regime). Under the REF scenario NO_x reductions in 2050 are much more drastic than NMVOC ones (Table 2) because the GEA-derived scaling coefficients used to project local scale emissions from 2020 to 2050 mainly implement NO_x reductions through domestic heating and road transport regulation. Under VOC-sensitive conditions NO_x reductions without additional regulation of NMVOCs lead to inhibited ozone titration and increase in ozone concentrations. Ozone mitigation would most likely be more effective if the domestic/industrial use of solvents (the main source of NMVOCs) were targeted as well. Under the MIT scenario however, where both NO_x and NMVOCs are mitigated more effectively than in REF, ozone concentrations decrease in 2050 compared to present time levels. This feature stands-out in the NO_x-limited rural areas in which ozone follows the fate of its precursors showing a much more drastic decrease of ozone under MIT (Fig. 4c) compared to REF (Fig. 4b). On the other hand chemistry in the REG simulation is driven by a NO_x-sensitive regime. The titration process is much less pronounced compared to the LOC run. As a result ozone decreases in response to the significant NO_x reductions (see also discussion in Sect. 2.3) under both REF and MIT scenarios.

In the regional setup both urban and rural chemistry is characterized by NO_x-sensitive conditions (panel b vs. d). On the contrary, if urban chemistry is clearly VOC-sensitive at the local scale run (panel a), in-plume ozone built-up is found on the ridge line separating the two regimes (panel c). This is consistent with previous studies (Menut et al., 2000; Sillman et al., 2003). Urban ozone levels modeled at the local scale are much lower than those modeled at the regional scale by almost 50 ppb (~54 ppb vs. ~100 ppb). This is due to the higher NO_x emission ratios prescribed by the local scale inventory (LOC) compared to the regional inventory (REG). As far as the in-plume chemistry is concerned the difference between the two simulations is less pronounced (~20 ppb). This is consistent with the fact that the reductions in rural ozone modelled at regional scale (REF: -3.2 ppb and MIT: -13.5 ppb) and in our study for the two future projections relative to present time levels are similar. Photochemical ozone built-up occurs at longer time and space scale compared to titration and therefore the increase in the resolution of the emissions brought about at the local scale does not provide much new information to the modeling of rural ozone.

This sensitivity analysis shows that the integration of high-resolution emission projections in the modeling of ozone is critical. Clearly the regional and local simulation will respond differently to emission changes in the future but unfortunately it is not possible to assess quantitatively the impact of this change since besides the spatial gradients in emissions, projections also differ between the LOC and REG simulations. The conclusion that can be drawn though from this sensitivity analysis is that the simulation forced by the regional scale emissions fails to distinguish the chemical regime between urban and rural chemistry (both shown as NO_x-limited) and that ozone titration over Paris is most likely underestimated in the regional setup which may explain the ozone decrease modelled for 2050 over Paris under the REF scenario. Increase in ozone due to sharp NO_x emission reductions under a VOC-limited environment as modelled with the local setup may be more realistic.

4.2.2 Future chemical regimes indicators

Based on the aforementioned previous studies as well as on the sensitivity exercise carried out here, ozone formation in the city of Paris occurs under VOC-sensitive conditions. However, it is plausible that the implementation of emission reductions in the long run causes a shift of the chemical regime towards a more NO_x-sensitive chemistry (Beekman and Vautard, 2010; Colette et al., 2012; Tarasson et al., 2003).

To investigate this shift, we use here a number of chemical regime indicators, which explain emission accumulation and radical production/loss processes: O_3/NO_y, O_3/NO_z and H_2O_2/NO_z. Each indicator accounts for different aspects of ozone chemistry, for example the H_2O_2/NO_z ratio takes into account both the impact of emissions and radical production (Sillman, 1995) by comparing the HO_x + NO_x radical sink (yielding NO_z) to the HO_2 + HO radical sink yielding H_2O_2. The latter sink corresponds to enhanced radical production, which favors NO_x-sensitive conditions (Kleinman, 1997). These indicators are used in order to avoid the large number of sensitivity runs such as the ones described in Sect. 4.2.1 that would be necessary to perform if one used a more direct approach such as above.

Chemical regime indicators are estimated from domain-wide daytime O_3, NO_y, NO_z and H_2O_2 concentrations for the CTL run and the two REF and MIT projections (Fig. 6). Higher O_3/NO_y, O_3/NO_z and H_2O_2/NO_z ratios point to more NO_x-sensitive conditions while lower ratio values point to VOC-sensitive chemistry. A visual inspection of the figure reveals that all three chemical indicator ratios increase under both REF and MIT scenarios compared to CTL due to decreases in NO_y and NO_z concentrations induced by the implemented NO_x emission reductions. For example the decade mean O_3/NO_y molar ratio shifts from the preset-time value of 0.64 to 2.75 and 10.6 in the REF and MIT scenarios respectively. Based on these indicators we deduce that a shift towards more NO_x-sensitive chemistry should be expected in 2050 under both REF and MIT scenarios.

4.2.3 Ozone health indicators

Here, we study how different ozone health indicators change under the two future scenarios (Table 5). We focus on three indexes: i) the sum of the differences between maximum daily 8-hour running means and the 35 ppb threshold value (SOMO35); ii) the number of days where maximum daily 8-hour running mean ozone concentration exceeds the 60 ppb threshold (Nd120) and iii) the mean of the ten highest daily max concentrations during the ozone period (MTDM). These metrics are typically used in health impact assessment studies and account for the non-linearity in the ozone dose-response function. For example SOMO35 was developed in the Joint WHO/Convention Task Force in 2004 and represents the cumulative annual exposure to ozone. The threshold value was established on the fact that a statistically significant increase in mortality risk estimates was observed at ozone concentrations above 25–35ppb. .

Under the REF scenario SOMO35 modelled at local scale in the city is almost doubled compared to present-time (Table 5) while the reductions of the Nd120 index from 29 to 18 days points to less frequent ozone episodes in the future. The MTDM index remains constant suggesting that ozone episodes will not decrease in intensity.

All three health related proxies improve under the MIT scenario. SOMO35 decreases by 79% and MTDM by 26.5% relative to CTL corresponding to future conditions with no significant ozone episodes (Nd120 falls to zero). The modelled reductions in the health indicators represent a much more efficient mitigation compared to maximum ozone concentrations (see discussion in Sect. 4.2) highlighting the high sensitivity of these indicators to their respective cut-off thresholds. It becomes clear, that from a human health perspective the MIT scenario is very effective and the co-benefits of climate and air quality strategies are significant. In the downwind rural areas south of Paris all health indicators under both scenarios are improved, as seen in Table 5, following the subsiding of ozone levels in the future (Fig. 4).

4.3 $PM_{2.5}$

Local scale $PM_{2.5}$ projections (wintertime) are presented in Fig. 7 as well as differences between each of the REF and MIT scenarios and the CTL simulation. Major decrease in $PM_{2.5}$ is modelled under both REF and MIT projections compared to present time levels. This is mainly due to significant reductions in primary particle emissions (Table 2). In downtown Paris, concentrations are reduced from 14.2 $\mu g/m^3$ in CTL to 3.4 $\mu g/m^3$ and 1.6 $\mu g/m^3$ under the REF and MIT scenarios respectively. The reductions are much stronger in the city than at rural areas due to the effective mitigation of road transport emissions. Consequently the large wintertime urban increment (difference between urban and rural concentrations) of approximately 6.8 $\mu g/m^3$ modelled with CTL is significantly reduced in both future projections: ~1 $\mu g/m^3$ in REF and less than 0.2 $\mu g/m^3$ in MIT.

5 Conclusions

Mid-21st century ozone and $PM_{2.5}$ projections over the city of Paris have been modelled with the CHIMERE air-quality model at a 4 km horizontal resolution under two consistent climate and emission scenarios: a reference and a mitigation scenario. To our knowledge, this is the first time that a study of a 10-year air-quality projection under climate and city level emission changes is conducted over a large European agglomeration at such fine scale. A key innovation of this work is that we use local-scale emissions and their projections until 2020 developed by local experts (AIRPARIF) and extend those until 2050 based on coefficients extracted from large-scale emission projections. A 10-year control simulation served model evaluation purposes. Furthermore, we investigate how ozone projections under the two future scenarios depend on the photochemical regime.

Model evaluation showed a very good agreement between model and measurements for ozone and an underestimation of wintertime $PM_{2.5}$ by 20% over the urban area, which is mainly attributed to a large wet bias in wintertime precipitation (+26.5%). The comparison between modeled and measured O_x showed that the model at 4 km resolution accurately resolves O_3 titration by NO over the highly urbanized city of Paris. The decade average bias within the ozone period for O_x (O_3+NO_2) and ozone is found to be less that 0.3 ppb.

Under the reference scenario ozone increases over the city of Paris by +7 ppb relative to present time values and only a small decrease is modelled under the mitigation scenario (-3.5 ppb). Through a sensitivity analysis, we showed that ozone formation in Paris occurs under VOC-sensitive chemistry. Under such conditions and due to the stronger mitigation of NO_x compared to NMVOCs in the REF scenario, titration is inhibited and ozone increases in the 2050 horizon. Following the MIT trajectory both NO_x and NMVOCs are more effectively regulated leading to ozone reduction in 2050. The same sensitivity analysis applied on the regional emission projections showed that ozone formation in Paris occurred under NO_x-sensitive conditions. The discrepancy in the chemical regime is attributed to differences in ozone precursor emissions prescribed by the two inventories: NO_x/NMVOCs emission ratios are much lower in the regional (0.2) compared to the local inventory (0.5). Modelling at regional scale most likely underestimates ozone titration prescribing NO_x-limited chemistry for Paris. Under this chemical regime ozone precursor emission cutbacks prescribed even for the less optimistic REF scenario benefits ozone air quality to such an extent that reductions are observed while the local setup yields ozone increases instead. If the regional scale model is inadequate to resolve urban features of ozone chemistry, mainly titration close to high NO_x emission sources, it yields similar rural ozone levels as the local scale run showing that the longer time-scale processes of emission transport and ozone formation are less sensitive to model resolution in the high ozone concentration plume. Our findings suggest that the estimates based on European scale applications are likely to overestimate the downward ozone trend under VOC-sensitive conditions while differences in rural areas are limited.

Finally in downtown Paris, $PM_{2.5}$ concentrations are reduced by 78% and 89% under the REF and MIT scenarios respectively. The reduction is much more prominent over the urban part of the domain due to significant reductions in primary emissions as a result of the effective mitigation of the road transport and residential sectors. Therefore the large urban increment modelled at the control run is significantly reduced in both future projections.

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Table 1. Configuration description of the global, regional and local simulations of the present study.

	Global[a]	Regional[b]	Local
GCM/CTM	LMDz-INCA	CHIMERE	CHIMERE
Horizontal	1.9° x 3.75°	0.5° x 0.5°	4km x 4km
Anthropogenic emissions (present)	RCP	GEA	AIRPARIF
Anthropogenic emissions scenarios[c]	RCP-2.6 / RCP-4.5	MIT / REF[d]	MIT/REF consistent[e]
Climate model	IPSL-CM5A-MR	WRF	WRF
Resolution	2.5° x 1.26°	0.5°	10km
Climate scenarios	RCP-2.6 / RCP-4.5	RCP-2.6 / RCP-4.5	RCP-2.6 / RCP-4.5

a For details see Hauglustaine et al. (2014).

b For details see Colette et al. (2013).

c Description given in Sect. 2.3.

d The mitigation (MIT) and the reference (REF) anthropogenic emission scenarios were developed to be consistent with RCP-2.6 and RCP-4.5 respectively.

e The future emissions of the local scale simulations are developed based on a hybrid methodology utilizing local and regional information (details in Sect. 2.3).

Table 2. Current time and future emission estimates (kt) over a 50km^2 area around Paris extracted from the 1st model layer.

	NO_x[a]	NMVOCs[a]	$PM_{2.5}$[b]
CTL	11.2	22.6	1.5
2050 REF	3.1	10.6	0.34
2050 MIT	1.0	8.1	0.18

a ozone period (April-August)

b winter period

Table 3. Observed and modelled meteorological variables over the Ile-de-France region. Modelled data stem from the WRF simulation at 10km resolution. Absolute model bias is given in parenthesis.

Variable	Summer (JJA)		Winter (DJF)	
	Obs	Model	Obs	Model
7 stations average				
T2 (°C)	19.2	19.8 (+0.6)	4.3	4.2 (-0.1)
WS10 (m/s)	2.9	3.7 (+0.8)	3.6	5.5 (+1.9)
RH (%)	69.1	55.8 (-13.3)	85.0	83.9 (-1.1)
PRECIP (mm/day)	0.076	0.073 (-0.003)	0.07	0.094 (+0.024)
MONT (Paris)				
T2 (°C)	20.1	20.9 (+0.8)	5.4	4.9 (-0.5)
WS10 (m/s)	2.8	3.3 (+0.5)	3.3	4.7 (+1.4)
RH (%)	64.9	50.8 (-14.1)	79.8	79.5 (-0.3)
PRECIP (mm/day)	0.073	0.085 (+0.012)	0.064	0.081 (+0.017)

Table 4. Future changes in key meteorological variables[a] under the two simulated climate scenarios.

Variable	CTL	Δ(REF-CTL)	Δ(MIT-CTL)
Temperature (°C)	11.6	+1.1	-0.5
RH (%)	72.3	+1.1	+1.7
Precipitation (mm/day)	0.1	+0.007	+0.22
Radiation (W/m^2)	154.3	-5.4	-25.4

a Annual domain-wide values extracted from 10-yr mean of daily averages.

Table 5. Ozone exposure indicators for the control simulation and the relative differences between the latter and the two future projections for the city of Paris.

Variable	CTL	REF	MIT
City			
SOMO35 (ppb days)	13763	27470	2904
Nd120 (days)	29	18	-
MTDM (ppb)	53.9	54.7	39.6
Rural			
SOMO35 (ppb days)	39581	29101	2115
Nd120 (days)	134	4	-
MTDM (ppb)	63.8	52.9	38.9

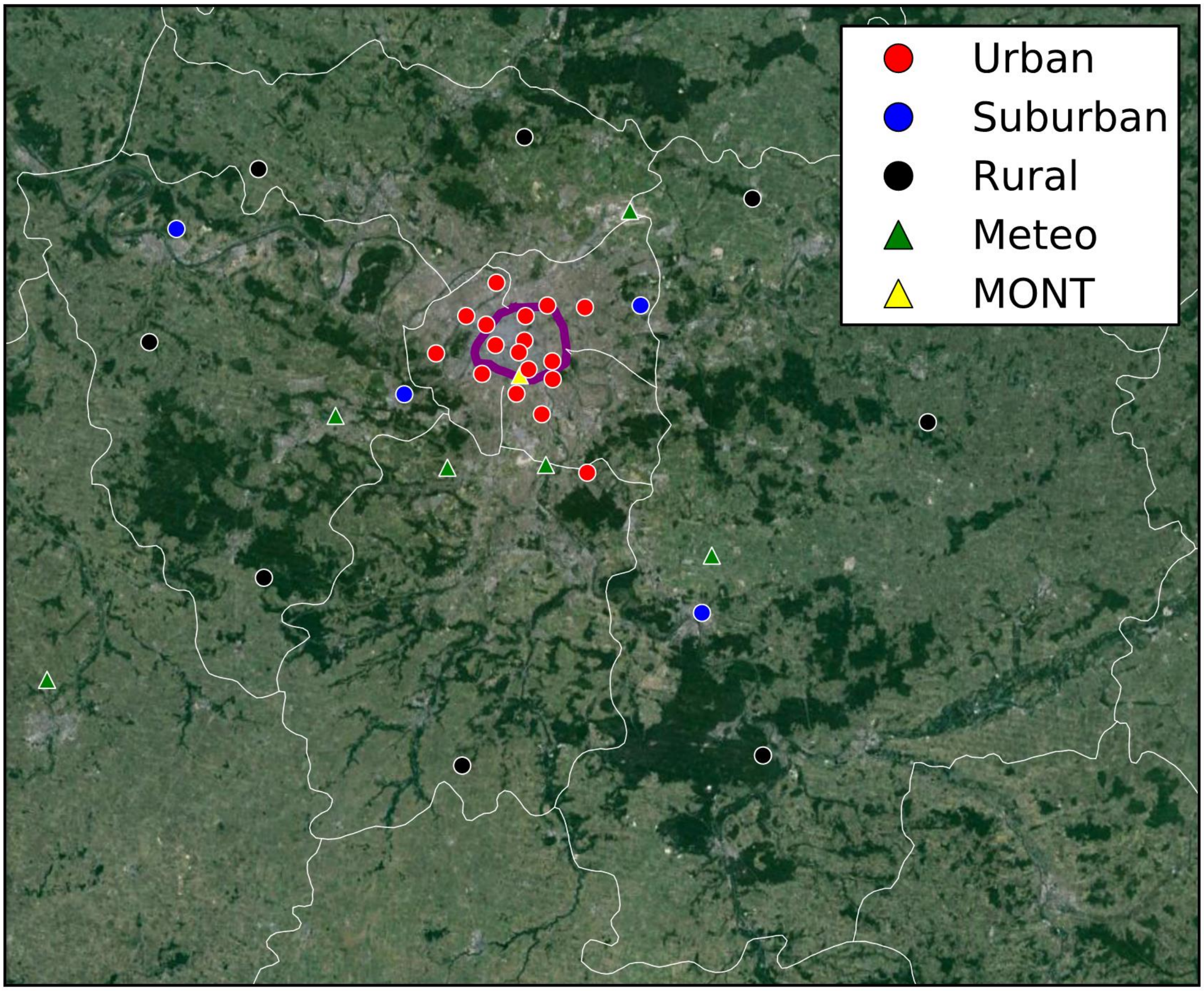

Figure 1. Local scale (IdF) simulation domain, with the city of Paris in the center (area enclosed in the purple line). Circles correspond to sites of the local air-quality monitoring network (AIRPARIF) with red for urban, blue for suburban and black for rural. Triangles correspond to the meteorological stations (the yellow triangle correspond to the Montsouris meteorological located in the city center).

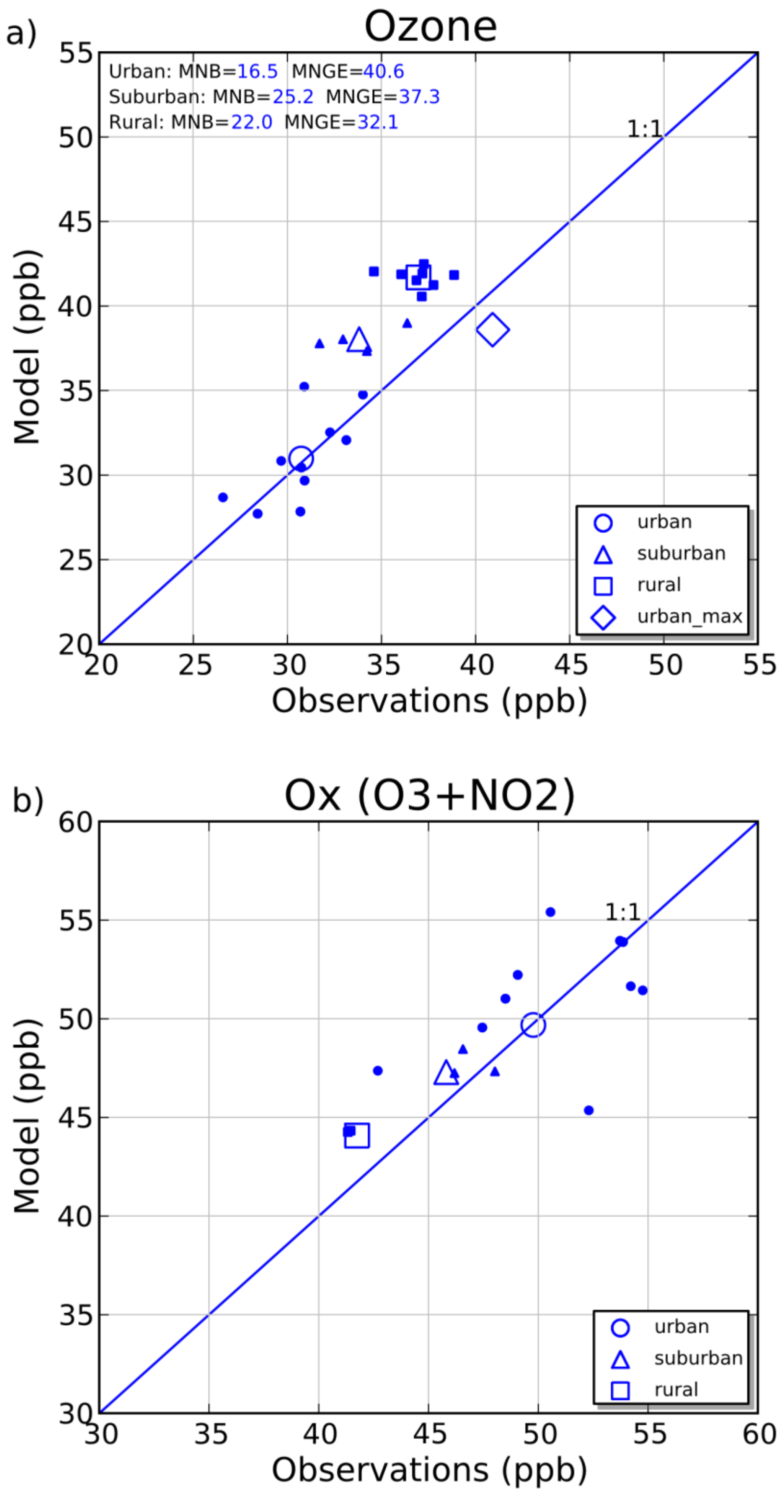

Figure 2. Scatter plots of mean daytime ozone (a) and O_x (b) during the April to August period from the model run against observations. Full small markers represent concentrations at individual stations while large hollow markers stand for the aggregated value. “Urban_max” stands for the daily maximum of an 8-hr running average.

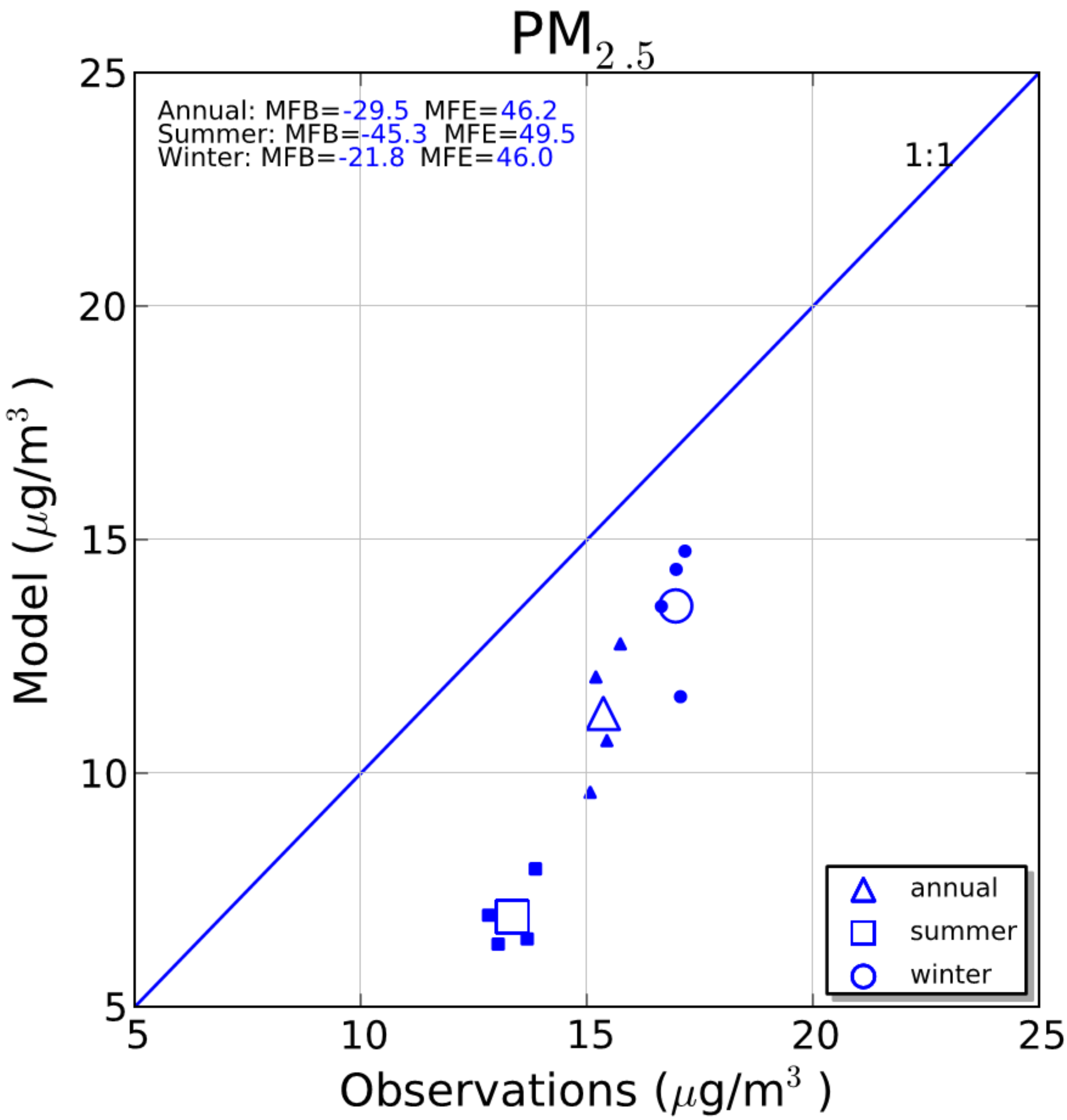

Figure 3. Scatter plot of mean daily $PM_{2.5}$ from the model run during summer (JJA), winter (DJF) and on annual basis against observations. Full small markers represent concentrations at individual stations while large hollow markers stand for the aggregated value.

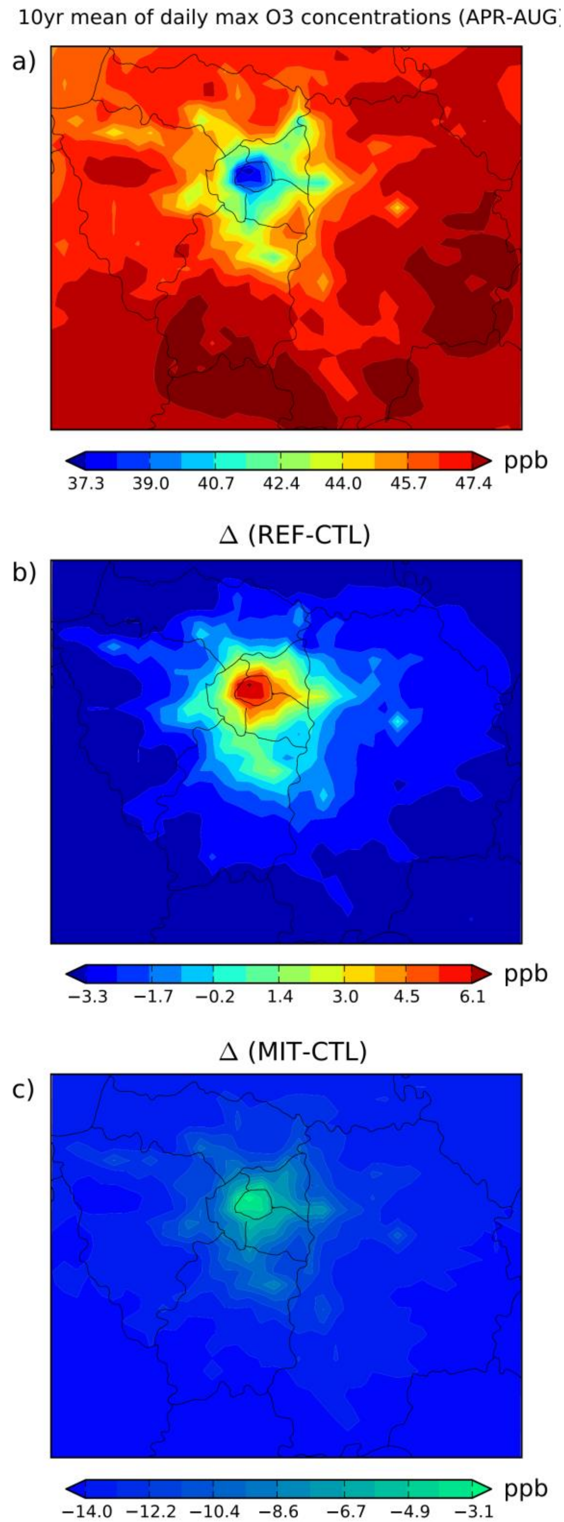

Figure 4. Present time ozone daily maximum (ppb) of 8-hr running means averaged over the April-August period (a) and differences between the CTL run and the REF (b) and MIT projections (c).

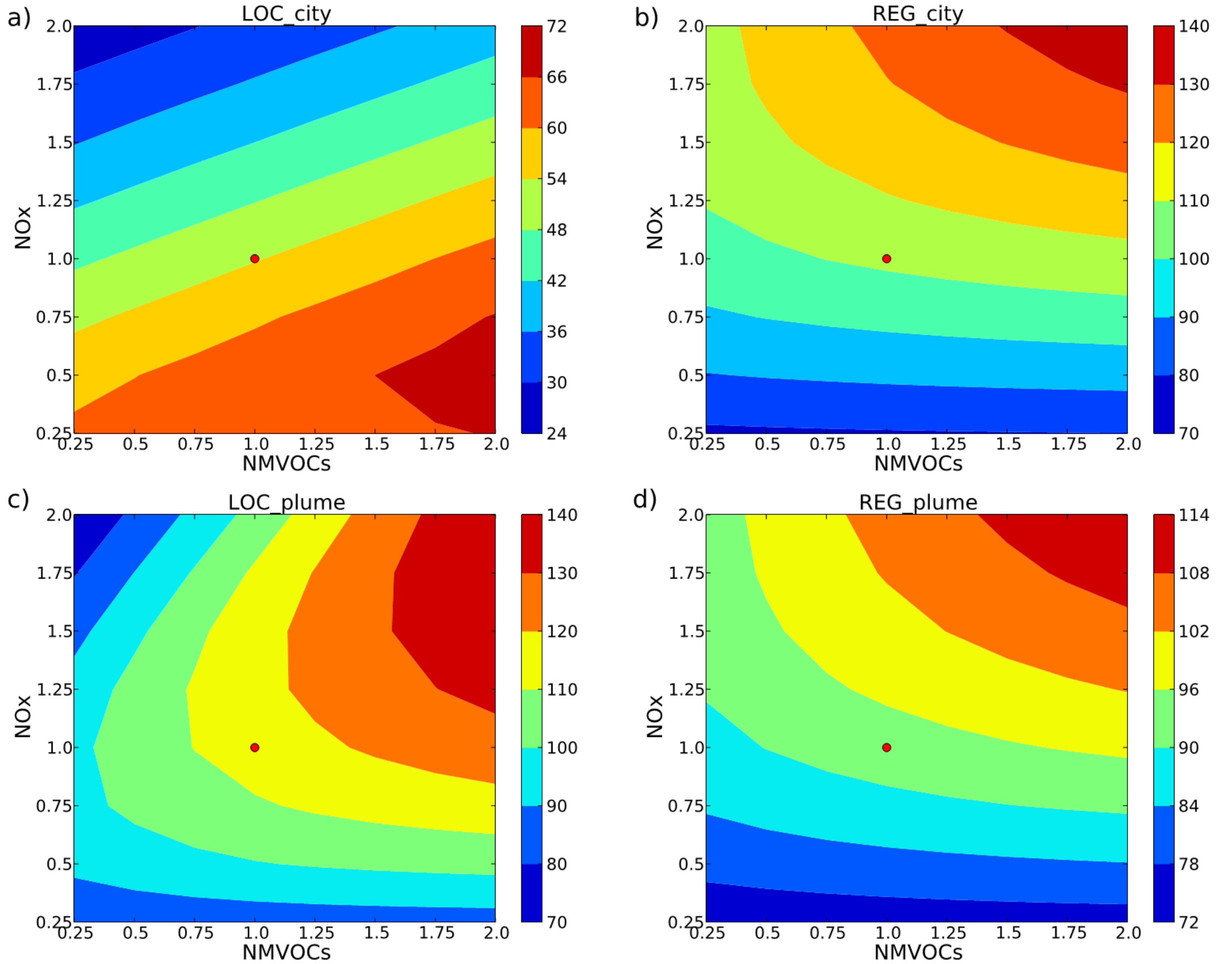

Figure 5. Isocontours of maximum ozone concentrations over downtown Paris (top panels) and over the entire modelling domain (IdF) (bottom panels) as a function of ozone precursor emissions. Axis units are the coefficients of emission changes (in molecules/cm^2/s) with respect to their baseline values (point 1.0, 1.0). Isopleths on the left are modelled with the LOC simulation and on the right with the REG simulation.

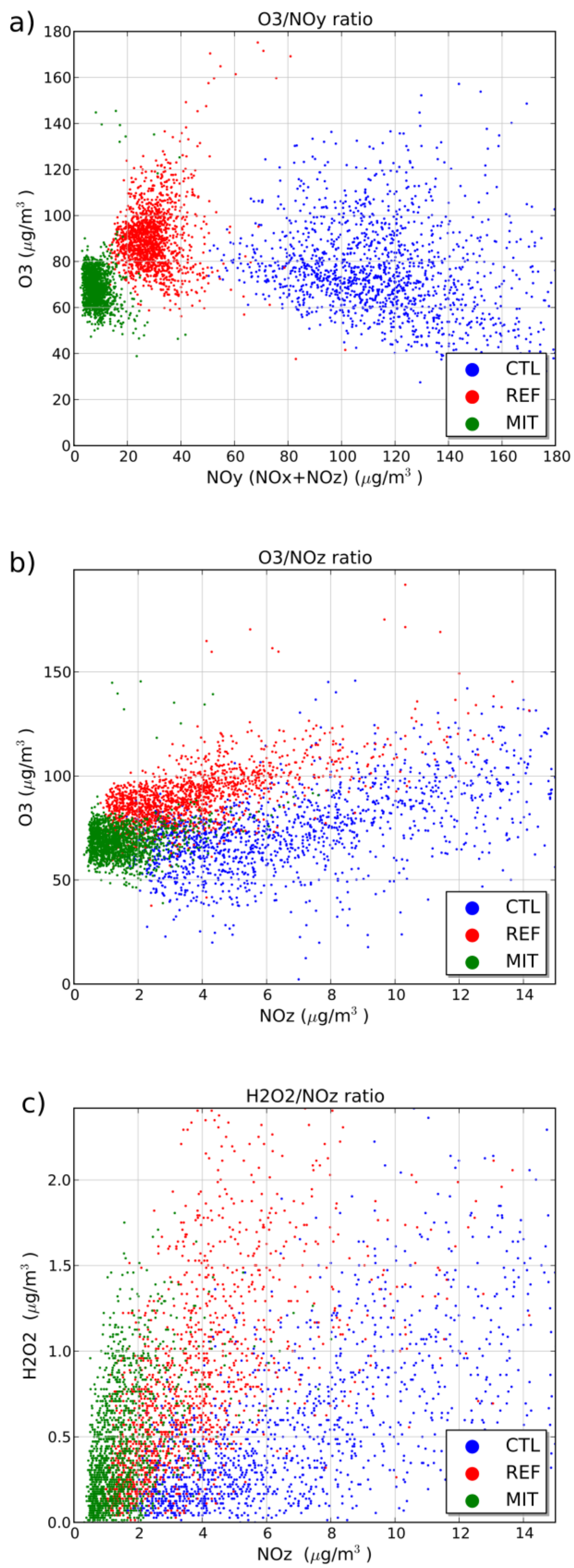

Figure 6. Ozone chemical regime indicators for the control run and the two future projections. Dots represent daily means averaged during the ozone period over the grid-cells of the downtown Paris area.

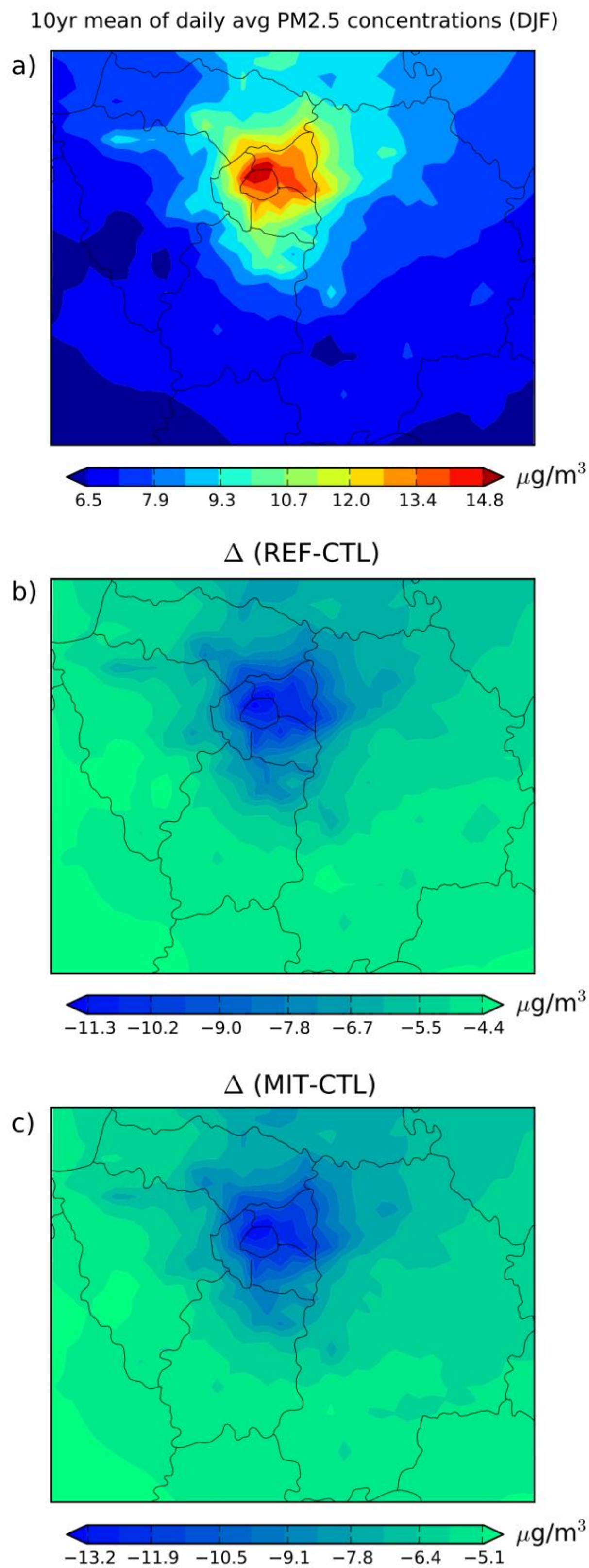

Figure 7. Wintertime (DJF) $PM_{2.5}$ daily average fields ($\mu g/m^3$) for the control simulation (a) and the differences between the latter and the REF (b) and MIT (c) future projections.