Reviewer 1:

Specific Comments:

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Reply: The authors believe that the fact that the two test cases implement different model chains is overemphasized in the manuscript therefore we find it appropriate to revise the phrasing "a different chain of models was used for the two case studies" as well as a possible misleading phrase "therefore particularly interesting to compare" in page 27044, line 28. A careful discussion of the implications due to that particular choice would be relevant in a cross-city comparison. This is not the case here as results are presented separately for the two cities and at the final stage (section 5.5) the cities are linked but still not compared; they are used as illustrative examples (Stockholm is dominated by regional pollution while IdF exhibits less regional influence) to discuss possible policy misclassification issues. It is commonplace that biases are indeed model-specific but without an intercomparison experiment or ensemble modelling within each case study more conclusions cannot be drawn. These types of experiments are not within the scope of this study as in any other single case, single model study. We add "We should note that the range of uncertainty in the results presented here is probably underestimated due to the choice of a single model chain for each case study."

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Reply: Indeed, the simulations on both domains were performed based on the standard practice and similarly to the reviewer we expect small differences. We have performed a linearity check for the Stockholm simulations and confirmed the linear relationship. We added in parenthesis "the linearity of this relationship was confirmed for the Stockholm simulations and assumed for the IdF simulations".

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Reply: For the regime analysis we extract MD8hr values for all chemical compounds involved. This is because regimes can be more easily distinguished at the time of the local maximum when the phenomena of production/loss are stronger. A common practice is to use the 1h maximum (if it can be easily identifiable) or take the average of several hours around the local maximum. Since there is no convention on this we arbitrarily use MD8hr in our analysis. In fact, the analysis was also performed implementing the daily averages but results remained the same. We have revised the caption of Fig. 6 to make clear that all values correspond to MD8hr.

Reviewer 2:

General Comments

1. Why are different chains of models used for Paris and Stockholm for the climate and air quality simulations (Table 1)? More importantly, by utilizing different models for the two regions, how are cross-city comparisons affected. Have the authors performed any sensitivity runs with respect to different model configurations? Some discussion of why different chains of models were employed in this analysis is needed somewhere in Section 2.1.

Reply: The sensitivity of modelled results to the use of different models is not the scope of this study (such intercomparison experiment is labour intensive and requires the redesign of many inputs in order to be implemented). In contrast, the current simulations were conducted in support of local scale health impact assessment in those regions (this information was added in the beginning of section 2.1) under the framework of the ACCEPTED project (Assessment of changing conditions, environmental policies, time-activities, exposure and disease) and the different models simply reflect the corresponding expertise of the two research teams in Paris and Stockholm. Of course it is well known that biases can be model-specific. Indeed, such discussion would be relevant in a cross-city comparison study which is not the case here. Results are presented separately for the two cities and at the final stage (section 5.5) the cities are used as illustrative examples (Stockholm is dominated by regional pollution while IdF exhibits less regional influence) to discuss possible policy misclassification issues. In that respect we find it appropriate not to overemphasize the use of different chain of models and the phrasing "a different chain of models was used for the two case studies" has been changed. Another possible misleading phrase ("therefore particularly interesting to compare") in page 27044, line 28 was also removed. We add "We should note that the range of uncertainty in the results presented here is probably underestimated due to the choice of a single model chain for each case study."

2. Why is the urban-scale modeling performed at 1 km resolution for Stockholm, but at 4 km resolution for Paris? If the emissions are available for Paris at 1 km resolution (Page 27049, Line 6), it seems odd not to model Paris at the same urban-scale resolution as for Stockholm. I would expect the higher resolution to be more important for Paris, since Stockholm has a stronger regional influence on

air quality (Page 27058, Line 6). Some rationale is needed for why the horizontal resolutions are different between the two cities, and how this might affect modeling results.

Reply: Paris region was modelled at 4km resolution based on findings of Markakis et al. (2105) (already referenced in the manuscript). There we show that modelled results in climate based simulations over IdF were not very sensitive to model resolution changes. Previous work (Valari and Menut, 2008) conducted in IdF showed that higher resolution also induces potential for error and model results were not improved by the increased model resolution. We have revised the first paragraph of section 2.2 accordingly: "In Markakis et al. (2015) we have conducted a sensitivity analysis on a decade simulation over IdF to test the response of modeled ozone and PM2.5 concentrations to the refinement of information related to model setup and inputs. On the basis of those findings, in the present study we implement a mesh-grid of 4km horizontal resolution (consisting of 39 grid cells in the west-east direction and 32 grid cells in the north-south direction), vertically resolved with 8 σ-p hybrid layers from the surface (999hPa) up to 5.5km (500hPa). The lowest layer is 25m thick."

Valari, M. and Menut, L.: Does an Increase in Air Quality Models' Resolution Bring Surface Ozone Concentrations Closer to Reality? J. Atmos. Oceanic Technol., 25, 1955–1968.

3. Section 2.1 (Page 27048, Line 7). Does the CHIMERE model include SOA chemistry like the MATCH model (Line 13)? If SOA chemistry is not included for Paris, how would this affect modeling PM under present and future climate scenarios?

Reply: Yes, CHIMERE includes SOA chemistry. A reference is added in section 2.1.

Specific Comments

4. Section 2.3 (Page 27051). A figure of sectoral emissions for Stockholm, similar to Paris (Figure 2), would be useful. To an unfamiliar reader, it is not clear how different the two cities are in their emission sources, and how this might affect future mitigation scenarios. For example, is it realistic for Stockholm to adopt the more aggressive local emissions mitigation strategy of Paris (Page 27051, Lines 16-19), beyond reducing traffic-related emissions only?

Reply: Unfortunately, we cannot reach the high level of detail found on the IdF inventory as regards the Stockholm emissions due the difficulties related to the corresponding emission processing system. We have added some more information in the text.

5. Section 3 (Page 27052, Line 4). How is PM measured from the urban stations? I'm guessing by filter samples. If so, could there be sampling artifacts in the measurements that affect the model evaluation for PM (Page 27052, Lines 18-26)? For example, sampling artifacts for OC can be large and dependent on measurement techniques.

Chow, J.C., et al., Quantification of PM2.5 organic carbon sampling artifacts in US networks. Atmospheric Chemistry and Physics, 2010. 10: p. 5223-5239. DOI: 10.5194/acp-10-5223-2010.

Turpin, B.J., P. Saxena, and E. Andrews, Measuring and simulating particulate organics in the atmosphere: problems and prospects. Atmospheric Environment, 2000. 34: p. 2983-3013. DOI: 10.1016/S1352-2310(99)00501-4.

Reply: To our best of knowledge the measurements are conducted using TEOM instruments (which to our knowledge is not the method reported by the reviewer) that have been associated with artefacts due to unaccounted semi-volatile compounds like ammonium nitrate. To some degree the measurements underestimate the true atmospheric load but we do not have information on the magnitude. We have included a brief reference to this in the evaluation section: "We note that the

measurements of particulate matter for the period in question was conducted using the Tapered-Element Oscillating Microbalance (TEOM) method that has been associated with negative sampling artefacts depending on the season, location and particle size (Allen et al., 1997)"

Allen, G., Sioutas, C., Koutrakis, P., Reiss, R., Lurmann, F.W. and Roberts, P.T.: Evaluation of the TEOM method for measurement of ambient particulate mass in urban areas, J. Air Waste Manag. Assoc., 47 (6), 682-689, 1997.

6. Section 3 (Page 27052, Line 29). Can the Norr Malma (rural) site be identified in Figure 1? Also, is the prevailing wind pattern such that this site is upwind or downwind of Stockholm?

Reply: Norr Malma is outside the model domain, and it is upwind from Stockholm. Thus it is slightly affected by the urban area. The description of the Stockholm measurement sites has been improved in the manuscript.

7. Section 5.6 (Page 27063, Lines 4-6). Not sure what is meant by this sentence. If Paris does not undergo a regime shift by 2050, shouldn't stronger NMVOC controls be emphasized?

Reply: Yes, this is correct but this is not what the local emission projection portrays. We revised this part: "Under VOC-sensitivity ozone benefit may be attained by either pushing NMVOCs mitigation over NOx or by enforcing enough mitigation on NOx emissions that will allow a shift of the photochemical regime towards NOx-limited conditions prior to 2050. In contrast the local emission projection enforces NOx over NMVOCs reductions while according to the long-term evolution of chemical regimes, studied with the use of chemical regime indicators, NOx mitigation is not strong enough for the aforementioned shift to take place by 2050."

8. Figure 3. To improve robustness of model evaluation, these plots would benefit from the addition of error bars that show the variability of the mean for the model and observations. Also, correlation coefficients of the model against observations should be reported somewhere.

Reply: Due to the nature of our simulations the evaluation process is based on decade average concentrations (for model and observations) rather than hourly or daily averages, typical in more short-term evaluations. This means that temporal correlation cannot be provided. Also, the evaluation of the variability of concentrations based on some intra-period temporal average (e.g., annual) is beyond the scope of this paper. Here we only interested in the ability of the model to simulate period average concentrations.

9. Figures 4 and 5. Can a consistent color scale be applied between Figure 4 (Paris) and Figure 5 (Stockholm)? It is hard to interpret the color scales easily between the two cities otherwise.

Reply: This was done because the purpose of these figures is to visualize the responses within each city separately. Consistent colorbars across cities smooths even further some of the spatial gradients found within cities for example in the present-time and the ozone responses.

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2. Why is the urban-scale modeling performed at 1 km resolution for Stockholm, but at 4 km resolution for Paris? If the emissions are available for Paris at 1 km resolution (Page 27049, Line 6), it seems odd not to model Paris at the same urban-scale resolution as for Stockholm. I would expect the higher resolution to be more important for Paris, since Stockholm has a stronger regional influence on air quality (Page 27058, Line 6). Some rationale is needed for why the horizontal resolutions are different between the two cities, and how this might affect modeling results.

Reply: Paris region was modelled at 4km resolution based on findings of Markakis et al. (2105) (already referenced in the manuscript). There we show that modelled results in climate based simulations over IdF were not very sensitive to model resolution changes. Previous work (Valari

and Menut, 2008) conducted in IdF showed that higher resolution also induces potential for error and model results were not improved by the increased model resolution. We have revised the first paragraph of section 2.2 accordingly: "In Markakis et al. (2015) we have conducted a sensitivity analysis on a decade simulation over IdF to test the response of modeled ozone and PM2.5 concentrations to the refinement of information related to model setup and inputs. On the basis of those findings, in the present study we implement a mesh-grid of 4km horizontal resolution (consisting of 39 grid cells in the west-east direction and 32 grid cells in the north-south direction), vertically resolved with 8 σ-p hybrid layers from the surface (999hPa) up to 5.5km (500hPa). The lowest layer is 25m thick."

Valari, M. and Menut, L.: Does an Increase in Air Quality Models' Resolution Bring Surface Ozone Concentrations Closer to Reality? J. Atmos. Oceanic Technol., 25, 1955–1968.

3. **Section 2.1 (Page 27048, Line 7).** Does the CHIMERE model include SOA chemistry like the MATCH model (Line 13)? If SOA chemistry is not included for Paris, how would this affect modeling PM under present and future climate scenarios?

Reply: Yes, CHIMERE includes SOA chemistry. A reference is added in section 2.1.

Specific Comments

4. Section 2.3 (Page 27051). A figure of sectoral emissions for Stockholm, similar to Paris (Figure 2), would be useful. To an unfamiliar reader, it is not clear how different the two cities are in their emission sources, and how this might affect future mitigation scenarios. For example, is it realistic for Stockholm to adopt the more aggressive local emissions mitigation strategy of Paris (Page 27051, Lines 16-19), beyond reducing traffic-related emissions only?

Reply: Unfortunately, we cannot reach the high level of detail found on the IdF inventory as regards the Stockholm emissions due the difficulties related to the corresponding emission processing system. We have added some more information in the text.

5. Section 3 (Page 27052, Line 4). How is PM measured from the urban stations? I'm guessing by filter samples. If so, could there be sampling artifacts in the measurements that affect the model evaluation for PM (Page 27052, Lines 18-26)? For example, sampling artifacts for OC can be large and dependent on measurement techniques.

Chow, J.C., et al., Quantification of PM2.5 organic carbon sampling artifacts in US networks. Atmospheric Chemistry and Physics, 2010. 10: p. 5223-5239. DOI: 10.5194/acp-10-5223-2010.

Turpin, B.J., P. Saxena, and E. Andrews, Measuring and simulating particulate organics in the atmosphere: problems and prospects. Atmospheric Environment, 2000. 34: p. 2983-3013. DOI: 10.1016/S1352-2310(99)00501-4.

Reply: To our best of knowledge the measurements are conducted using TEOM instruments (which to our knowledge is not the method reported by the reviewer) that have been associated

with artefacts due to unaccounted semi-volatile compounds like ammonium nitrate. To some degree the measurements underestimate the true atmospheric load but we do not have information on the magnitude. We have included a brief reference to this in the evaluation section: "We note that the measurements of particulate matter for the period in question was conducted using the Tapered-Element Oscillating Microbalance (TEOM) method that has been associated with negative sampling artefacts depending on the season, location and particle size (Allen et al., 1997)"

Allen, G., Sioutas, C., Koutrakis, P., Reiss, R., Lurmann, F.W. and Roberts, P.T.: Evaluation of the TEOM method for measurement of ambient particulate mass in urban areas, J. Air Waste Manag. Assoc., 47 (6), 682-689, 1997.

6. Section 3 (Page 27052, Line 29). Can the Norr Malma (rural) site be identified in Figure 1? Also, is the prevailing wind pattern such that this site is upwind or downwind of Stockholm?

Reply: Norr Malma is outside the model domain, and it is upwind from Stockholm. Thus it is slightly affected by the urban area. The description of the Stockholm measurement sites has been improved in the manuscript.

7. Section 5.6 (Page 27063, Lines 4-6). Not sure what is meant by this sentence. If Paris does not undergo a regime shift by 2050, shouldn't stronger NMVOC controls be emphasized?

Reply: Yes, this is correct but this is not what the local emission projection portrays. We revised this part: "Under VOC-sensitivity ozone benefit may be attained by either pushing NMVOCs mitigation over NOx or by enforcing enough mitigation on NOx emissions that will allow a shift of the photochemical regime towards NOx-limited conditions prior to 2050. In contrast the local emission projection enforces NOx over NMVOCs reductions while according to the long-term evolution of chemical regimes, studied with the use of chemical regime indicators, NOx mitigation is not strong enough for the aforementioned shift to take place by 2050."

8. Figure 3. To improve robustness of model evaluation, these plots would benefit from the addition of error bars that show the variability of the mean for the model and observations. Also, correlation coefficients of the model against observations should be reported somewhere.

Reply: Due to the nature of our simulations the evaluation process is based on decade average concentrations (for model and observations) rather than hourly or daily averages, typical in more short-term evaluations. This means that temporal correlation cannot be provided. Also, the evaluation of the variability of concentrations based on some intra-period temporal average (e.g., annual) is beyond the scope of this paper. Here we only interested in the ability of the model to simulate period average concentrations.

9. Figures 4 and 5. Can a consistent color scale be applied between Figure 4 (Paris) and Figure 5 (Stockholm)? It is hard to interpret the color scales easily between the two cities otherwise.

Reply: This was done because the purpose of these figures is to visualize the responses within each city separately. Consistent colorbars across cities smooths even further some of the spatial gradients found within cities for example in the present-time and the ozone responses.

Mid-21st century air quality at the urban scale under the influence of changed climate and emissions. Case studies for Paris and Stockholm.

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Abstract

Ozone, PM_{10} and $PM_{2.5}$ concentrations over Paris, France and Stockholm, Sweden were modeled at 4km and 1km horizontal resolutions respectively for the present and 2050 periods employing decade-long simulations. We account for large-scale global climate change (RCP-4.5) and fine resolution bottom-up emission projections developed by local experts and quantify their impact on future pollutant concentrations. Moreover, we identify biases related to the implementation of regional scale emission projections by comparing modeled pollutant concentrations between the fine and coarse scale simulations over the study areas. We show that over urban areas with major regional contribution (e.g., the city of Stockholm) the bias related to coarse scale projections may be significant and lead to policy misclassification. Our results stress the need to better understand the mechanism of bias propagation across the modeling scales in order to design more successful local-scale strategies. We find that the impact of climate change is spatially homogeneous in both regions, implying strong regional influence. The climate benefit for ozone (daily mean and maximum) is up to -5% for Paris and -2% for Stockholm city. The climate benefit on $PM_{2.5}$ and PM_{10} in Paris is between -5 and -10%, while for Stockholm we estimate mixed trends of up to 3% depending on season and size class. In Stockholm, emission mitigation leads to concentration

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reductions up to 15% for daily mean and maximum ozone and 20% for PM_x. Through a sensitivity analysis we show that this response is entirely due to changes in emissions at the regional scale. On the contrary, over the city of Paris (VOC-limited photochemical regime), local mitigation of NO_x emissions increases future ozone concentrations due to ozone titration inhibition. This competing trend between the respective roles of emission and climate change, results in an increase in 2050 daily mean ozone by 2.5% in Paris. Climate and not emission change appears to be the most influential factor for maximum ozone concentration over the city of Paris, which may be particularly interesting in a health impact perspective.

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1 Introduction

There is a growing body of literature on the projected effects of climate and emission reduction scenarios on future air quality. The published research encompass an envelope of models and methodologies; up to now global scale models have been extensively used to study the impact of climate on tropospheric ozone at global or regional scales (Liao et al., 2006; Prather et al., 2003; Szopa and Hauglustaine, 2007), while chemistry transport models (CTMs), having more advanced parameterization of physical and chemical processes, are applied to study selected regions with refined horizontal resolution (Andersson and Engardt, 2010; Colette et al., 2012, 2013; Katragkou et al., 2011; Langner et al., 2012a; Nolte et al., 2008; Zanis et al., 2011).

Numerical models are used to study future evolution of air quality as they allow the evaluation of the effectiveness of planned strategies to mitigate pollutants concentrations. This is particularly important since it is now well established that elevated concentrations deteriorate human health (Jerrett et al., 2009; Lepeule et al., 2012), while new scientific evidence indicate that pollution is harmful at even lower levels than previously thought (REVIHAAP, 2013). There is an increasing number of studies investigating the health effects of population exposure to specific emission source types such as traffic, industry or biomass burning (REVIHAAP, 2013 and references therein). Although a clear association is not established, there is evidence that living near busy roads substantially increases the total burden of disease attributable to air pollution (Pascal et al., 2013). In Europe, one third of the urban population resides in areas where the legislated target value for PM_{10} is exceeded (EEA, 2013).

The fact that today most of the world's (and Europe's) population lives in cities stresses the need to resolve the variability of pollutant concentrations and provide predictions of future air quality

at the urban scale (Riahi et al., 2011). Up to now the principal focus of relevant research was solely on the global and regional scales utilizing modeling resolutions of a few hundred (global) to a few tenths (regional) of kilometers. Nevertheless, it has been repeatedly shown that coarse resolutions are inadequate to resolve fine scale features (Markakis et al., 2014, 2015; Valari and Menut, 2008; Vautard et al., 2007) due to insufficient representation of chemistry and the use of coarse resolution emission inventories that cannot dissociate the strong emission gradients of the large urban agglomerations from those at surrounding rural areas. There is still practically no information on the climate-air quality interactions at the urban and local scales. A reason is the large computational demand in refining model resolution, while maintaining large spatial coverage. Another is the fact that emission scenarios at fine scale are rarely developed, since long-term projections are constrained by the evolution of energy supply and demand, which is a large scale issue. Air quality projections employing locally developed policy are scarce; a first attempt is described in Gidhagen et al. (2012) who developed air quality projections until the near future (2030s) for the greater Stockholm region in Sweden with a high resolution (4km) modeling system. The impacts were assessed in terms of climate and emissions that were constructed by local experts, however the number of meteorological years included was limited and emissions were projected only for the road transport sector. In Markakis et al. (2014) we describe long-term air quality projections (2050) at urban scale utilizing 10 year-long simulations and fine scale features such as high model resolution (4km) and an emission inventory developed by local experts for the Il-de-France (IdF; an 8-department area including Paris) region in France.

In the present assessment we implement several improvements compared to the works of Gidhagen et al. (2012) for the Stockholm region and Markakis et al. (2014) for IdF, aiming to improve our knowledge on the climate and pollutants emissions driven air quality responses at a refined scale. Here we develop a consistent framework including identical climate and emission scenarios at global and regional scales, horizon of projection (2050), number of simulation years (decade) and pollutants considered (ozone, PM_{10} and $PM_{2.5}$). In this work, Stockholm and Paris cities are used as illustrative examples of large urban agglomerations that have very different origins of influence; Paris is largely affected by local emissions while Stockholm experiences significant contribution by non-local sources. We implement a high resolution modeling grid of 1km for Stockholm and 4km for the IdF region. Here (in contrast to Markakis et al. (2014)) we take into account changes from large-scale global climate and fine-scale local emissions and disentangle their influence in

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shaping local concentrations at the 2050 horizon. For Stockholm we additionally quantify the contribution of the locally enforced emission reduction plan from that introduced by the pan-European change in emissions. To describe the future evolution of pollutant emissions at the city scales we rely on high-resolution bottom-up projections at the 2030 horizon developed by local experts (instead of 2020 used in Markakis et al. (2014)).

Additionally, we employ the coarse applications that have provided the boundary conditions to the fine scale simulations from which we extract the signal for ozone, PM_{10} and $PM_{2.5}$ of future concentration change related to the emission mitigation over the IdF and Stockholm domains. Previous research conducted in IdF (Markakis et al., 2014) indicated a possible overestimation of the ozone concentration response from coarse resolution applications in areas characterized by VOC-limited conditions. More specifically we (Markakis et al., 2014) have identified opposing signals in the projected maximum ozone concentrations, with the regional-scale application to yield large decreases while the urban-scale large increases attributed to the fact that the former implemented top-down coarse resolution emissions and portrayed Paris under NO_x-limited chemistry at present-time conditions, therefore making the city more receptive to forthcoming NO_x emission reductions, compared to the high-resolution simulation portraying a VOC-limited chemistry for Paris. Provided that coarse inventories lack the integration of local policies, this work advances on the work of Markakis et al. (2014) and Gidhagen et al. (2012) by providing the means to identify the differences risen when finer areas are investigated with the refined information of locally developed emission projections and higher resolution. This can help to answer whether there is an added value in integrating local emission-related policy to larger-scale inventories. Specifically for ozone, in order to facilitate the comparison between the scales, we examine the long-term evolution of chemical regimes by employing chemical regime indicators which are a measure of radical production/loss processes (Beekman and Vautard, 2010; Sillman et al., 2003).

2 Materials and methods

The IdF region is located in north-central France (1.25–3.58° east and 47.89–49.45° north) with a population of 11.7 million, more than two million of which live in the city of Paris. The area is situated away from the coast and is characterized by uniform and low topography, not exceeding 200m above sea level. Stockholm is located in south-eastern Sweden, with a population of 1.4

million. Stockholm is located partly on islands where the western coast of the Baltic Sea meets Lake Mälaren. Fig. 1 illustrates the modeling domains of the urban scale simulations over IdF and Stockholm regions and the boundaries of the cities of Paris and Stockholm. 10-year long simulations were carried out over each domain to represent present-time (1991-2000) and mid-21st century (2046-2055) air quality.

2.1 Regional downscaling of climate and air quality data

The air-quality simulations for the IdF and Stockholm regions were conducted to support local scale health impact assessment under the framework of the ACCEPTED ("Assessment of changing conditions, environmental policies, time-activities, exposure and disease") project. Table 1 summarizes the chain of models and configurations utilized for the two case studies. We should note that the range of uncertainty in the results presented here is probably underestimated due to the choice of a single model chain for each case study. To derive projections of the main climate drivers over Europe at 0.11º horizontal resolution (see Giorgi et al. (2009)), we used the IPSL-CM5A-MR (Dufresne et al., 2013) global climate model downscaled with the WRF regional climate model (Skamarock and Klemp, 2008) for the IdF region and the EC-EARTH global climate model, downscaled with the RCA4 regional climate model (Jacob et al., 2014; Strandberg et al., 2014) for Stockholm. In total, 8 (for present and future) meteorological simulations were implemented in this study.

For both case studies, pollutant concentrations at the global scale were simulated (Szopa et al., 2013) with the LMDz-INCA global model (Hauglustaine et al., 2004) at monthly temporal resolution. The regional downscaling of multi-year pollutant concentration averages though, is done separately for each case study, first over 0.44º (~50km) resolution grids over Europe and then with a single nest over a 4km resolution grid over the IdF region and a two-step nesting over grids of 0.11º (~12km) and 1km resolution over Sweden and Stockholm respectively (6 simulations in total were conducted at present day conditions). A thorough presentation of the regional scale air quality simulations used as boundary conditions for the urban scale runs are provided in Watson et al. (2015).

Two sets of simulations (for each scale) were conducted at future conditions; in the first case we implement future meteorology along with present-time emissions in order to isolate the effect of climate change whereas in the second case we utilize future meteorology and projected emissions

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to quantify the combined effect of climate and emissions change. The signal of emission mitigation alone can be subsequently derived from the concentration difference between the two aforementioned runs (the linearity of this relationship was confirmed for the Stockholm simulations and assumed for the IdF simulations). Finally, only for the Stockholm domain we run an additional test case that allows the quantification of the contribution of emission changes at the regional scale compared to the role of the local scale emission mitigation. This is completed using future projections of local emissions for Stockholm but keeping the respective emissions of the regional scale simulation at present-time levels.

Air quality simulations were conducted with the CHIMERE (Menut et al., 2013) and MATCH (Robertson et al., 1999) CTMs for the IdF and Stockholm regions respectively. CHIMERE is used at both urban and regional scales and it has been benchmarked in a number of model inter-comparison experiments (see Menut et al. (2013) and references therein). The MATCH model is applicable to scales from urban to hemispheric and has been extensively used to study the connection between climate change and air quality in Europe (e.g., Andersson and Engardt, 2010; Engardt et al., 2009; Langner et al., 2005, 2012b). Both models are used operationally for emergency preparedness, environmental surveillance and air quality forecasts at France (http://www.prevair.org), Sweden (http://www.smhi.se) and EU (http://www.macc.eu).

The CHIMERE model includes gas-phase, solid-phase and aqueous chemistry, biogenic emission modeling with the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006), dust emissions (Menut et al., 2005) and re-suspension (Vautard et al., 2005) modules. Gas-phase chemistry is based on the MELCHIOR mechanism (Lattuati, 1997) and includes more than 300 reactions of 80 gaseous species. CHIMERE treats sulfates, nitrates, ammonium, organic and black carbon, dust and sea-salt. The gas-particle partitioning is treated with ISORROPIA (Nenes et al., 1998). The secondary organic aerosol (SOA) chemistry of CHIMERE is described in Bessagnet et al. (2009).

The MATCH model includes options for data assimilation (e.g., Kahnert, 2008), modules describing aerosol microphysics (Andersson et al., 2015) and ozone- and particle-forming photo-chemistry considering ~60 species (Langner et al., 1998; Andersson et al., 2007, 2015) based on Simpson et al. (2012). MATCH also includes SOA formed by oxidation of biogenic and anthropogenic volatile organic compounds (ASOA and BSOA). The SOA modeling is based on the volatility basis set (VBS) scheme in the EMEP MSC-W model (Bergström et al. (2012) with

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modifications from Bergström et al. (2014)). In the present study, primary organic aerosol emissions were considered non-volatile and VBS schemes were only used for "traditional" ASOA and BSOA; BVOC-emissions of isoprene and monoterpenes were calculated in the model, using the methodology of Simpson et al. (2012). A small emission of sesquiterpenes, equal to 5% of the daytime monoterpene emissions, was added (as in Bergström et al. (2014)).

2.2 Urban scale air quality modeling and emissions

In Markakis et al. (2015) we have conducted a sensitivity analysis on a decade simulation over IdF to test the response of modeled ozone and $PM_{2.5}$ concentrations to the refinement of information related to model setup and inputs. On the basis of those findings, in the present study we implement a mesh-grid of 4km horizontal resolution (consisting of 39 grid cells in the west-east direction and 32 grid cells in the north-south direction), vertically resolved with 8 σ-p hybrid layers from the surface (999hPa) up to 5.5km (500hPa). The lowest layer is 25m thick. The 1km resolution domain, covering Stockholm, consists of 48x48 grid cells. The vertical resolution follows the layers of the driving regional climate model, distributed between 20 layers with a 60m thick surface layer.

Present-time emission estimates for the IdF region are available at a 1km resolution grid. Emissions are compiled with a bottom-up approach by the IdF environmental agency (AIRPARIF) combining a plethora of city-specific information (AIRPARIF, 2012). The spatial allocation of emissions is either source specific (e.g. locations of point sources) or completed with proxies such as high-resolution population maps and a detailed road network. The inventory has hourly source specific, temporal resolution. The compilation of present-time emission for the Stockholm region (covering an area of 30 municipalities and 2.2 million inhabitants) is also based on a bottom-up approach e.g., the estimates of total traffic volumes are primarily based on in-situ measurements and variations of vehicle composition and temporal variation of the traffic volumes are described for different road types. Vehicle fleet composition and vehicle exhaust emission factors are based on the Swedish application of the ARTEMIS (Assessment and Reliability of Transport Emission Models and Inventory Systems) model (Sjödin et al., 2006). There are also large non-tailpipe emissions due to road, tyre and break wear. In Stockholm the non-tailpipe emissions dominate and emission factors are estimated based on local measurements (Omstedt et al., 2005; Ketzel et al.,

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2007). The emission database has hourly source specific, temporal resolution. More details on the emission data and how they were compiled can be found in Gidhagen et al. (2012).

2.3 Climate and regional scale emission projections

Climate follows the long-term 4.5 scenario of the Representative Concentration Pathways (RCP-4.5) that exhibits a 20% greenhouse gas emission reduction for Europe, constant population and mid-21st century global radiative forcing at 4W/m^2, increasing to 4.5W/m^2 by 2065 and stabilizing thereafter (Clarke et al., 2007). Shown in previous work (Markakis et al., 2014) this scenario represents an intermediate alternative between the pessimistic and optimistic RCPs (8.5 and 2.6 respectively) in terms of long-term temperature projection in IdF with 0.6ºC increase in the 2050 annual mean temperature compared to -0.5ºC for RCP-2.6 and +1.1ºC for RCP-8.5.

The European scale simulations use anthropogenic emissions developed in the framework of the ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) project (Klimont et al., 2013). It is consistent with the long-term climate projections of the RCPs but also spatial algorithms to improve the representation of short-term continental and national air quality legislations. In this study we used the "Current Legislation Emission" scenario (CLE) for mid-21st century in Europe, which includes both climate and regional air quality policies and assumes full enforcement of all legislated control technologies until 2030 and no climate policy thereafter. CLE projects that NO_x, NMVOCs, PM_{10} and $PM_{2.5}$ emissions drop in 2050 by 43, 35, 32 and 32% respectively compared to the present day. The MATCH simulations include biomass burning emissions as well taken from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) database (Lamarque et al., 2010).

Urban-scale emission projections

The IdF region with the support of the "Direction Regionale et Interdepartementale de l'Environnement et de l'Energie d'Ile de France" (DRIEE-IF), has introduced the "Plan de Protection de l'Atmosphere d'Ile de France" (PPA) enforcing short and long term emission cutbacks in order to comply with the national legislation of air pollution concentration reductions. The 2030 emission projection for the IdF region includes gradual renewal of the vehicle fleet according to the latest emission standards (Euro VI), increased use of public transport, replacement of domestic fuel for heating with electricity and gas, new French thermal regulations

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in buildings, aviation traffic projections and implementation of planned legislation for the industrial sector. The emission projection for the county of Stockholm is founded on vehicle fleet evolution and emission factors for 2030 based on the application of the ARTEMIS model (details found in Gidhagen et al. (2012)). Other emissions besides the traffic-related were not changed from the present to the future in Stockholm.

Fig. 2 illustrates the annual, sectoral emissions of NO_x, NMVOCs, PM_{10} and $PM_{2.5}$ in the IdF domain for the present-time and the 2030 scenario. Present-time NO_x emissions mainly stem from the transport sector (~60% of annual emissions), largely mitigated by 2030 (emissions decline from 60Gg to 20Gg). The leading emitter of NMVOCs at present-time is the "use of solvents" sector accounting for 49% of all-sector annual emissions. Interestingly the emissions coming from this sector are hardly mitigated in the future compared to NO_x; the corresponding reduction reaches only 11%. The transport, industrial and heating sectors have important PM_{10} emission shares at present day. The heating and transport sectors are strongly mitigated (reductions reach ~60%) while industrial emissions are abated by only 18% mainly due to the fact that their primary origin is fugitive dust released during production processes whereas the mitigation plan introduces fuel-based reductions. The main contributors of annual fine particles emissions are the transport and the heating sectors, both strongly mitigated by 2030 (transport sector's emissions drop by 96%). Total present-time emissions are reduced by 55% for NO_x, 32% for NMVOCs, 37% for PM_{10} and 54% for $PM_{2.5}$. For Stockholm about 60 and 80% of present-time NO_x and PM_{10} emissions respectively stems from the road transport sector. The decrease in the future (by 16, 18 and 10% for NO_x, NMVOCs and PM_{10} respectively) in domain-wide emissions is mainly a result of planned renewal of the traffic fleet and stricter emission limits. Finally, as there are no local scale emission projections available for the 2030-2050 period we assume that local emissions are unchanged between 2030 and 2050. Never the less this assumption is in-line with the European scale emission scenario (CLE).

3 Model evaluation

In this section we evaluate the present day simulations at the study domains. Surface ozone concentrations modeled with CHIMERE and MATCH (averaged over the ozone period which spans from April to August) were compared against all available measurements of the air quality networks included in the high resolution domains e.g., 17 urban, 5 suburban and 8 rural sites in

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IdF and one urban site (Torkel Knutsson) in Stockholm. We also evaluate maximum ozone concentrations calculated from 8-hour running means (MD8hr). Modeled PM_{10} and $PM_{2.5}$ ground-level concentrations in summer (JJA), winter (DJF) and on annual basis are also compared to all available measurement sites in the high resolution domains: 7 urban stations in IdF and 1 urban station (Torkel Knutsson) in the Stockholm region. Results are illustrated with scatter plots in Fig. 3. For Stockholm we additionally evaluate the organic carbon (OC) and elemental carbon (EC) as well as sea salt (as sodium) using measurements conducted during the years 2002-2003 and 2013 respectively at the remote site of Aspvreten, located 70 km south-east of Stockholm. The Aspvreten site is located outside the 1km Stockholm domain therefore we use model results from the 12km resolution simulation to represent the modelled background. We note that the measurements of particulate matter for the period in question was conducted using the Tapered-Element Oscillating Microbalance (TEOM) method that has been associated with negative sampling artefacts depending on the season, location and particle size (Allen et al., 1997).

Fig. 3a shows that over the urban stations of IdF, CHIMERE overestimates daily ozone (overall bias=10%) mostly at the urban sites outside the city center; focusing on downtown monitoring sites the model bias is only 3.7% (not shown). The simulation successfully reproduces MD8hr. Overestimation of daily ozone is observed at suburban (by 14.6%) and rural (by 13.3%) stations. Discrepancies in rural ozone may be due to overproduction of isoprene emissions due to a warm modelled bias (+0.3°, not shown) or enhanced advection from the boundaries.

The evaluation of $PM_{2.5}$ at urban stations (Fig. 3b) shows a negligible mean bias during winter but overestimation by 15.3% in the summer. Simulations in Markakis et al. (2014), where dust emissions were not included, showed an underestimation of both summer and winter period concentrations suggesting that CHIMERE might overproduce dust particles especially in the drier summer period. From the other hand a sensitivity analysis conducted with the use of reanalysis meteorology in Markakis et al. (2015) has revealed that the small wintertime $PM_{2.5}$ bias could be due to model error compensation such as unrealistically high modelled precipitation (not shown) and possible inhibition of vertical mixing or overestimation of wintertime anthropogenic emissions. Wintertime PM_{10} concentrations appear underestimated (Fig. 3c) provided that the enhanced wet deposition affects the larger particles more. While the exaggeration of summertime dust emissions is also valid for PM_{10}, PM_{10} concentrations for the same period are generally well represented. It is possible that the stronger modeled winds in the summer compared to observations

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(not shown) affect the larger particles more, through accelerated dry deposition (Megaritis et al., 2014). The wintertime underestimation of PM_{10} concentrations is compensated by a positive autumn bias (not shown) leading to unbiased annual average concentrations.

For the Stockholm case we have first identified the regional and local contributions to ozone, PM_{10} and $PM_{2.5}$ concentrations utilizing measurements from the rural site of Norr Malma. It is sited 80km north-east of Stockholm and only slightly affected by urban plumes, therefore we use it as an indicator of the regional influence in the area. The local contribution is defined as the difference between concentrations monitored at the Torkel Knutsson (urban) and Norr Malma (rural) sites. To evaluate the modelled regional contribution, we utilize modelled concentrations at the respective sites. We note that Norr Malma site is located in the 12km resolution domain. The Stockholm city exhibits weak titration as the daily mean ozone concentrations measured at the two sites are similar (Table 2). The performance of MATCH is therefore mainly driven by the simulations at the coarser scales which overestimate nighttime ozone (not shown) due to too efficient vertical mixing during the night; this causes the MATCH model to overestimate the regional contribution in Stockholm by 17% (not shown), which also explains the major part of the positive bias at the 1km resolution simulation by 10% (Fig. 3d). On the contrary, the regional contribution in modeled MD8hr is well represented (bias <1%) leading to unbiased MD8hr in the high-resolution modeling.

Annual mean $PM_{2.5}$ concentrations are accurately reproduced (Fig. 3e) by the MATCH model over the city but summertime levels are overestimated by 14% and wintertime by 40%. This is due to a large over-production in total sea salt in the Stockholm domain, during the whole year (+2.1μg/m^3), but mostly during winter (+3μg/m^3). Despite this, an underestimation of PM_{10} concentrations by 26% is observed over the whole year (Fig. 3f). This is due to a large summertime underprediction of PM_{10} (40%), partly explained by the model's lack of aerosols of biogenic origin, which are mainly assigned to the coarse mode of the size distribution. Spores and other primary organic material have an important contribution to the speciation of the organic aerosol in northern Europe (20% to 32% of the total carbon during summer (Yttri et al., 2011)). Another possible reason is the underestimation of OC (by 1.5μg/m^3) and EC (by 0.1μg/m^3), which is probably due to the bias inherited by the regional scale simulations since less than 38% and 26% of city's PM_{10} and $PM_{2.5}$ concentrations respectively stem from local sources (Table 2). The regional contribution to PM_{10} concentrations based on monitor data is about 60% but due to the

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aforementioned reasons 17% lower based on the MATCH simulation (annual mean) mainly stemming from the summer period (-43%).

4 Climate projections for 2050

In Table 3 we show the projected domain-wide values of key meteorological variables. A warmer climate is expected in both regions. Surface temperature in IdF increases by 0.2^{o}C in summer and 0.4^{o}C in winter while in the Stockholm domain this trend is stronger reaching +1.3^{o}C in summer and +1.4^{o}C in winter. During the summer months, when ozone formation mainly occurs, no significant change in solar radiation is observed. Ground-level wintertime specific humidity rises by ~6% in IdF and by +7 and +9.7% in summer and winter respectively over Stockholm. The effect of humidity on ozone levels is ambiguous (see Jacob and Winner (2009) for a thorough discussion); elevated levels are linked with lower levels of background ozone (Johnson et al., 1999) even though some have found a weak effect in more polluted atmospheres (Aw and Kleeman, 2003). Changes in the planetary boundary layer height (PBL) affect pollutants dispersion. In IdF we observe an increase by 3.4% in PBL during the summer and decrease by 5.6% during winter. In the Stockholm domain projected changes in the PBL are less than 2%.

The precipitation rate, a regulating factor of PM concentrations, increase by 6.5% and 3.6% during summer and winter respectively in IdF whereas, summertime precipitation in the Stockholm domain decreases by 6.3% and wintertime levels increase by only 1.7%. Nitrate concentrations are expected to increase with humidity due to shift of the ammonia-nitric acid equilibrium to the aerosol phase (Seinfeld and Pandis, 2006) but to decrease due to the higher temperatures. On the other hand, sulfates increase with the warmer climate while there is evidence that elevated humidity may also lead to decrease in particle concentration by increasing the water content of particles and accelerating dry deposition rates (Megaritis et al., 2014). A warmer climate may also affect secondary organic production since semi-volatile pollutants are more prone to the gas phase under warm temperatures. Furthermore, climate change induced changes to the oxidizing capacity may cause changes to the volatility of organic gases.

5 Air quality modeling analysis

5.1 Present-time

Maps of present-time daily mean ozone concentrations (in the ozone period) and annual mean PM_{10} and $PM_{2.5}$ concentrations are illustrated in the left columns of Fig. 4 and Fig. 5 for IdF and Stockholm domain respectively. Concentrations that are spatially averaged over the cities of Paris and Stockholm (see Fig. 1) and domain-averaged concentrations that are representative of rural areas, are discussed separately. Consequently, lower ozone concentrations are found over the city-centers due to titration while higher levels are modeled at the surrounding areas due to photochemical formation (IdF) or long-range transport (Stockholm). The urban increment of daily mean ozone, defined here as the difference between the urban and the domain-averaged concentration, is -13μg/m^3 in IdF and only -1μg/m^3 in the Stockholm domain. Ozone formation in IdF is VOC-limited and therefore, titration rate over Paris is high (Markakis et al., 2014). On the contrary, ozone levels over the city of Stockholm are mainly due to transport from the boundaries and much less affected by local NO_x emission and titration (see also discussion in the previous section). Annual $PM_{2.5}$ and PM_{10} concentrations (Fig. 4e,i) are high over areas of intense anthropogenic activity such as the Charles-de-Gaulle international airport (north-east in the IdF domain), the city-centre and the suburbs of Paris due to road transport and wintertime heating emissions while local dust contributes with PM_{10} emissions to the south. The spatial pattern of $PM_{2.5}$ and PM_{10} concentrations in the Stockholm domain mainly reflects major roads, i.e. traffic emissions (Fig. 4e,i).

5.2 Future air quality at 2050 due to climate change

Fig. 4 and Fig. 5 show the future changes (compared to present-time) in daily mean ozone concentrations (over the ozone period) and annual mean PM_{10} and $PM_{2.5}$ concentrations, due i) only to climate change; ii) only to emission reductions and iii) to the combined effect of climate and emissions for IdF and Stockholm regions respectively. The spatial distribution of the ozone concentration difference between present and future reveal that despite the overall increase of mean surface temperature there is a domain-wide climate benefit for both domains. In Paris reductions in the daily and MD8hr ozone concentrations reach ~5% (Table 4). To some extent this is explained by the locale climate change; decrease in surface ozone despite the warmer climate has been also observed by other researchers (Coleman et al., 2014; Fiore et al., 2005; Lauwaet et al., 2014) and linked with enhanced ozone destruction through the $O_3 + OH \rightarrow HO_2 + O_2$ reaction due to increase in OH radicals triggered by higher surface water vapour ($O(^1D) + H_2O \rightarrow 2OH$).

For Paris this is consistent with the fact that NO_x concentrations are not much affected in the future ($|\Delta c|$=1.2μg/m^3) and therefore the decrease in ozone cannot be attributed to enhanced titration. The increase of the summertime period PBL height could also be responsible for the declining ozone trends through less dispersed primary NO_x emissions. Most probably changes in regional climate are responsible for the observed trend e.g., a weakened outflow from North America which is known to affect Europe through the north and western boundaries (Auvray and Bey, 2005; Lacressonniere et al., 2014). This is consistent with the fact that Paris and the IdF average responses are equivalent (Table 4) also evident in the Stockholm case which is known to have significant regional influence. Overall ozone concentration response in the Stockholm domain is negligible (~2% for daily mean and MD8hr ozone) driven by the respective response at the regional level (Watson et al., 2015).

Changes in future concentrations of particles in IdF are up to 5% and 10% for PM_{10} and $PM_{2.5}$ respectively, depending on season and area of focus (Paris or IdF average, Table 4). There is a weak climate benefit for annual concentrations of PM over Paris and the domain, mainly due to enhanced summertime precipitation. A small increase in PM concentrations over Paris is observed in wintertime as a result of a shallower boundary layer and higher temperatures that positively affect sulfates. PM annual concentrations over the Stockholm domain remain practically unchanged; a weak decrease of 3% is only estimated during winter, and similarly to ozone it is linked to regional-scale changes.

5.3 Local air quality at 2050 due to emission reductions

The spatial distribution of changes in mean daily ozone concentrations due to emission mitigation in the IdF region reveals two opposing trends (Fig. 4c); in Paris there is an overall increase of daily ozone by 4.8μg/m^3 (Table 4) despite the enforced NO_x emission mitigation. Under the VOC-limited photochemical regime characterizing the city, NO_x abatement inhibits the ozone titration process resulting in higher ozone levels. The magnitude of the ozone increase due to emission mitigation outbalances the predicted climate benefit and the combined effect leads to an overall penalty of +1.5μg/m^3 over Paris. In contrast, the domain-wide ozone concentrations decrease by 6.5μg/m^3, since ozone over the rural areas is less affected by titration (Markakis et al., 2014). It is worth noting that the absolute change in the MD8hr concentration over Paris due to climate change is two times higher than due to emission mitigation (Table 4). Therefore, while local emission

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mitigation has a stronger impact on background ozone levels, climate change affects more the ozone peaks (found at around 15:00LT in Paris). This may be particularly interesting from a health impact assessment standpoint where the MD8hr indicator is typically implemented (Likhvar et al., 2015).

Emission reduction policies appear to be more efficient for ozone abatement over the Stockholm region, with reductions reaching ~11 and ~13μg/m^3 for the mean and MD8hr respectively indistinctively for the city and the domain-averaged concentrations (Table 4). Based on the sensitivity simulations we find that the observed ozone decrease is entirely attributed to emission mitigation at the regional rather than the local scale (Table 5). We should note however, that the role of local emission reductions is probably underestimated in Stockholm due to lack of non-traffic emission abatement, although traffic is the main contributor to the Stockholm NO_x emissions contributing by ~50% to the total even after the future reductions.

Particle concentrations are very sensitive to their primary emission changes (Markakis et al., 2015). Therefore, it is not surprising that PM concentration reductions are mainly due to emission mitigation in both domains (Table 4). The domain-wide annual mean in IdF declines by 7.2 and 8.1μg/m^3 and in the Stockholm domain by 1.9 and 1.6μg/m^3 for PM_{10} and $PM_{2.5}$ respectively. In IdF the decrease is higher over areas and seasons with high primary PM, e.g., Paris compared to the rural areas of IdF (Fig. 4g,k) as well as in wintertime compared to summertime (-8.7μg/m^3 vs. -5.8μg/m^3 respectively for annual mean $PM_{2.5}$) due to significant abatement in the heating sector. In contrast, in the Stockholm domain the seasonal and spatial distribution of changes are much less prominent due to the prevailing regional influence (Table 5).

We have assumed unchanged local-scale emissions for the 2030-2050 period. Never the less, the projected concentration change in the Stockholm region is mostly affected by regional emission mitigation that according to the CLE emission scenario is weak. Therefore, further mitigation of local scale emissions would not strongly affect the future concentration change in the Stockholm domain. In contrast additional emission mitigation in the IdF scale would result in further improvement of domain-wide ozone and $PM_{2.5}$-related air-quality at the mid-21st century horizon. However, due to highly non-linear ozone chemistry over Paris, it is difficult to make firm assumptions on the nature of ozone projected changes, under additional mitigation of ozone precursor emissions in the 2030-2050 period.

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Future changes in population exposure to ozone ¶
In this section we discuss future changes in SOMO35 over the two study regions. SOMO35 is ozone related population exposure metric recommended by WHO and typically used in health impact assessment studies. SOMO35 is calculated as the sum of the differences between maximum daily 8h running means and the 70μg/m^3 threshold value. ¶
Present-time levels of SOMO35 in Paris are significantly lower than in the rural areas (represented by the domain average value) due to ozone titration over areas of high NO_x emission. It has been shown already that MD8hr is expected to decline by ~2.5% in 2050 (Table 4). Never the less the corresponding drop in SOMO35 is significantly higher reaching 26% (Table 6) due to the dependence of SOMO35 on its 70μg/m^3 cut-off concentration. In future conditions MD8hr for a considerable number of days will shift below threshold levels substantially reducing SOMO35. Similarly, in the Stockholm city, SOMO35 is expected to drop by 74% whereas MD8hr by only 17.4%. The examples of Paris and Stockholm presented here suggest that the use of SOMO35 as an indicator of population exposure may be misleading, since it is based on the underlying hypothesis that no health effects of ozone are present below 70μg/m^3. In the rural areas the implemented emission reduction policies will have substantial benefits in population exposure; SOMO35 drops by 69% in IdF and by 73% in the Stockholm domain. ¶
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5.4 Future evolution of ozone chemical regimes under local and regional scale chemistry-transport modeling in Paris

In this section we study the long-term evolution of ozone chemical regimes in the city of Paris. This analysis is not performed for Stockholm where ozone concentrations are controlled by long-range transport and less by the local chemistry which determines the regime. For each simulated day in the ozone period, in both present and future decades, we determine MD8hr concentrations of NO_y and the ratios of O_3/NO_y, H_2O_2/NO_y and H_2O_2/NO_z. The threshold values proposed in order to discriminate between the two chemical regimes (i.e., NO_x or VOC-limited) are 7.6ppb for NO_y (Beekman and Vautard, 2010), 5.5 for O_3/NO_y (Sillman et al., 2003), 0.12 for H_2O_2/NO_y (Sillman and He, 2002) and between 0.21 and 0.41 for H_2O_2/NO_z (Beekman and Vautard, 2010). The aforementioned analysis is applied on both regional (coarse-res) and urban-scale (high-res) simulations for present and future decades. Three indicators agree on a VOC-limited characterization of present-time ozone production at the urban scale simulation in agreement to the findings of Markakis et al. (2014) while only two indicators classify the regional scale ozone simulation as VOC-limited (Fig. 6). Despite a similar trend towards a more NO_x-limited photochemistry in 2050 at both high and coarse simulations, still three out of four indicators characterize the high-resolution simulation as VOC-limited at 2050 whereas the coarse resolution is positively NO_x-limited according to all four indicators.

5.5 Policy implications based on comparison of air quality projections from high and coarse resolution modeling

Air quality projections for 2050 indicate that ozone levels in Paris will increase by 8% and 3% for daily mean and MD8hr respectively as a response to the enforced emission mitigation plan. On the contrary, the coarse resolution simulation yields 7% and 15% decrease in these metrics (Table 6). A similar inconsistency was found in Markakis et al. (2014), where the Global Energy Assessment (GEA) emission projection (Riahi et al., 2014) was used instead of the ECLIPSE inventory. ECLIPSE stands as another state-of-the-art emission inventory, explicitly designed for air quality projections in order to cope with the drawbacks (Butler et al., 2012) of their global counterparts such as the RCPs which were intended for use in global scale climate studies. As discussed in the previous section, ozone production in the coarse resolution simulation by 2050 will shift from a VOC- to a NO_x-limited photochemical regime and therefore more responsive to reductions of NO_x

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emissions compared to the urban-scale simulation where the transition to NO_x-limited conditions is smoother. PM concentrations over Paris under the high-resolution modeling are expected to decrease by 21 to 46% depending on the season and particle cut-off diameter while the coarse-resolution simulation is about 10% more optimistic with reductions ranging from 34% to 55%. Both the evolution of chemical regimes and of PM concentrations are attached to the underlying emission projections. Under the coarse-scale storyline (CLE), annual emissions of NO_x over Paris drop by almost an order of magnitude while the local inventory yields a reduction of 66%. Annual PM_{10} and $PM_{2.5}$ emissions in Paris drop by 76% according to CLE while only by 10 and 38% respectively according to the local projection.

Given that the coarse inventory implements assessment at the large scale, its stronger mitigation over the city of Paris compared to the AIRPARIF projection is due to omission of local policy. The downscaling of coarse inventories on regional scale CTM grids passes through spatial proxies (such as land-use) to distribute emissions and the related bias induced to the air quality simulation over finer areas increases the overall bias of the application as well. The difference in the response of the regional and urban scale simulations is due, at large extent, to the spatial allocation algorithm (inherited by the RCPs) used in the compilation of both GEA and ECLIPSE databases (Riahi et al., 2011), which forces stronger (and possibly unrealistic) mitigation over the urban areas. Additionally, regional inventories assimilate regional/national legislation. In Europe the UNECE/LRTAP convention under the revised Gothenburg protocol (http://www.unece.org/fr/env/lrtap/status/lrtap_s.html) bounds the European member states (EU28) to achieve at a 2020 horizon relative to 2005 an overall reduction by 42% in NO_x emissions and 28% in NMVOCs emissions. Such reductions enhance the shift towards NO_x-limited ozone production. This remark, suggests that coarse-resolution ozone projections may be too optimistic over VOC-limited areas, mainly found in North-Western Europe (Beekman and Vautard, 2010) as well as PM projections over heavily populated urban areas. It is plausible that new updated protocols taking into account regional particularities should be implemented in European emission mitigation schemes and more credible assessments could be achieved by incorporating local policy in large scale inventories. This point is particularly relevant for areas such as Stockholm, where the regional scale mainly drives pollutant concentrations. The transfer of bias from the larger to the finer scale may lead to misclassification of local policy.

Despite the large differences in ozone concentrations simulated at regional and urban scales over the urban area of the city of Paris, rural concentrations are very similar; the projections at both scales show a decrease in ozone at 2050 at comparable magnitudes (Table 6). Therefore, fine-scale information provides little advantage in simulating rural ozone responses in agreement with Markakis et al. (2014). On the contrary, PM rural projections are very different between simulations at different resolutions (Table 6) suggesting that regional scale biases may be transferred to the finer scale run.

A final remark relates to the relative role of climate-change and emissions in future pollutant concentration projections. In contrast to the general conclusion of most recent pan-European scale studies (Colette et al., 2013; Geels et al., 2015; Lacressonniere et al., 2014; Langner et al., 2012b) we find that maximum ozone projections over Paris, modelled at the local scale are more sensitive (based on the absolute concentration change from present day) to climate change than to emission mitigation (Sect. 5.3). This suggests that the coarse-resolution applications could overestimate the magnitude of the contribution of the future emissions mitigation to the overall ozone concentration response.

6. Conclusions

Long-term projections of air quality at the urban scale integrating local emission policies are scarce. In the present study we investigate mid-21st century ozone and particulate matter concentrations focusing on two European cities: Paris, France and Stockholm, Sweden. Using a fine resolution modeling system (4km for the IdF region and 1km for Stockholm) we quantify the contribution of emission reduction policies and of climate-change to pollutant concentration changes at the 2050 horizon. For the Stockholm region we distinguish the role of locally enforced mitigation from that of regional-scale changes in emissions (European policy). Local scale emission changes rely on 2030 projections compiled by authorized air quality agencies at Paris and Stockholm.

The analysis of present-time ozone concentrations reveals very different photochemical conditions in the two case-studies; ozone formation in Paris is characterized as VOC-limited, with ozone titration being the main driver of concentration levels over the city, while both PM and ozone concentrations in Stockholm depend on long-range transport of pollution (96% and 70% of the local MD8hr and annual PM concentrations respectively originate from non-local sources).

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Overall we identify an ozone (daily mean and MD8hr) climate benefit up to -5% in IdF and -2% in Stockholm city despite the overall increase in the mean surface temperatures. For IdF this is not related to changes in local titration (as NO_x concentrations are little affected by 2050) but to changes in the regional climate. Provided the dominant regional influence in Stockholm, it is not surprising that the climate change contribution to the final PM concentrations follows the weak trend observed at continental scale simulations. In IdF, PM concentrations are expected to decrease due to the wetter climate predicted for the region although the trend is very weak.

We find that the mitigation of ozone-precursor emissions implemented in the IdF region instigates spatially irregular ozone concentration changes with a benefit over the rural areas (-9% and -12% for daily mean and MD8hr respectively) while, over the urban area we observe a penalty of +8% and +3% in daily mean and MD8hr ozone concentrations respectively due to titration inhibition. Under VOC-sensitivity ozone benefit may be attained by either pushing NMVOCs mitigation over NO_x or by enforcing enough mitigation on NO_x emissions that will allow a shift of the photochemical regime towards NO_x-limited conditions prior to 2050. In contrast the local emission projection enforces NO_x over NMVOCs reductions while according to the long-term evolution of chemical regimes, studied with the use of chemical regime indicators, NO_x mitigation is not strong enough for the aforementioned shift to take place by 2050.

In Paris, the increase in the daily mean ozone due to emission changes counterbalances the climate benefit to such extent that the combined effect is an overall penalty of +2%. In contrast changes in MD8hr concentrations due to climate (Δc=-4.1μg/m^3) are larger compared to those introduced by emission abatement (Δc=+2.2μg/m^3), indicating that the local maximum is more sensitive to climate change while background ozone concentration levels are more sensitive to emission changes. In the Stockholm city and the domain, emission mitigation is largely influential, with reductions several times higher than those introduced by climate both for ozone and PM. Contrary to Paris, we show that this response is entirely attributed to changes at the regional scale. Finally, the cumulative effect of climate and emissions in the city of Paris reaches +2.3% for daily mean ozone, -2.4% for MD8hr ozone, -33% for PM_{10} and -45% for $PM_{2.5}$ while for the Stockholm city, -17% for daily mean ozone, -18% for MD8hr ozone, -20% for PM_{10} and -20% for $PM_{2.5}$.

Another aim of this work was to quantify the plausible added value of the assimilation of local policy into regional scale inventories. To do so, we compared pollutant concentration changes modeled over the two cities at urban scale against regional-scale simulations over the same areas

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forced by ECLIPSE, a state of the art emission inventory designed to cope with the drawbacks of inventories such as the RCPs, by assimilating air quality policy at a continental scale. Over Paris the regional scale simulation is more optimistic than its urban scale counterpart. The fine scale modeling yields increase in ozone over the city of Paris (by 8% and 3% for daily mean and MD8hr respectively) while the regional scale modeling yields a 7% and 15% drop respectively. Regional scale simulations are more optimistic for PM concentrations as well with about 10% larger reductions compared to the urban scale projections. These discrepancies are a direct effect of the much stricter mitigation of primary anthropogenic emissions under the ECLIPSE scenario.

Overall our assessment suggests that the long-term evolution of atmospheric pollution solely based on regional scale emissions may lead to misclassification of the effect. The stricter mitigation in ECLIPSE projections is mainly due to the spatial allocation algorithm, which assigns unrealistically high mitigation over urban areas. It is plausible that new updated protocols taking into account the particularities of regions should be implemented in European emission mitigation schemes and that more credible assessments could be achieved by incorporating local policy to those inventories. An effect, overlooked by the coarse scale modeling, is the response of MD8hr ozone, a crucial input of health impact assessment studies: for Paris this metric is more prominent to climate change rather than to emission mitigation.

For Stockholm the comparison of regional and urban scale simulations shows small discrepancies given the major role of long-range transport over the area. This stresses the need to better understand the mechanism of bias propagation across the modeling scales in order to design more successful local-scale strategies.

Acknowledgements

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Table 1. Models (and their implemented resolutions) used for the simulations over the study regions.

	Climate[a]		Air quality[b]	
	IdF	Stockholm	IdF	Stockholm
Global	IPSL-CM5A-MR	EC-EARTH	LMDz-INCA	
	1.25º x 1.25º	1.125º x 1.25º	3.75° x 1.9°	
Regional	WRF, 0.11º	RCA4, 0.11º	CHIMERE, 0.44º	MATCH, 0.44º/0.11º
Urban	Same as regional	Same as regional	CHIMERE, 4km	MATCH, 1km

a IPSL-CM5A-MR: Institute Pierre Simon Laplace-Climate Model 5A-Mid Resolution, WRF: Weather Research and Forecasting, EC-EARTH: European Centre-Earth, RCA4: Rossby Centre regional atmospheric model

b LMDz-INCA: Laboratoire de Météorologie Dynamique Zoom- INteraction avec la Chimie et les Aérosols, MATCH: Multi-scale Atmospheric Transport and Chemistry

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Table 2. Quantification of the regional and local contributions to the present-time concentration levels at the city of Stockholm.

	City concentration levels (µg/m^3) [a]	**Local contribution [b]**	**Regional contribution [c]**
Ozone daily mean	62.5	-0.8	63.3
Ozone MD8hr	78.5	-3.3	81.8
PM_{10} annual mean	14.7	5.7	9.0
PM_{10} JJA mean	13.1	3.5	9.6
PM_{10} DJF mean	12.7	4.4	8.3
$PM_{2.5}$ annual mean	7.3	1.9	5.4
$PM_{2.5}$ JJA mean	6.5	1.5	5.0
$PM_{2.5}$ DJF mean	7.7	2.0	5.7

a based on the only available urban background station in the domain (Torkel Knutsson).

b calculated from the concentration difference between the Torkel Knutsson and the Norr Malma sites.

c based on measured concentrations at the Norr Malma site.

Table 3. Future changes in key meteorological variables in the study regions under the RCP-4.5 climate scenario. Seasonal averages include both day-time and night-time values.

IdF	**Summer (JJA)**		**Winter (DJF)**	
Variable	**REF**	**2050**	**REF**	**2050**
2m temperature (ºC)	18.8	+0.2	4.2	+0.4
Specific humidity (g kg^{-1})	7.9	+0.3	3.4	+0.2
Precipitation (kg m^{-2})	118	+7.1	130	+4.7
Radiation (W m^{-2})	262	-6.5	50	-1.9
10m wind speed (m s^{-1})	4.0	+0.2	6.8	-0.2
Boundary layer height (m)	643	+22	727	-41
Stockholm domain	**Summer (JJA)**		**Winter (DJF)**	
Variable	**REF**	**2050**	**REF**	**2050**
2m temperature (ºC)	12.9	+1.3	–1.2	+1.4
Specific humidity (g kg^{-1})	7.7	+0.6	3.1	+0.3
Precipitation (kg m^{-2})	223	-14	159	+2.7
Radiation (W m^{-2})	232	-0.4	28.2	-0.7
10m wind speed (m s^{-1})	3.2	-0.1	4.3	-0.1
Boundary layer height (m)	673	+6	574	-11

Table 4. Changes in pollutants concentrations (in μg/m^3) between present (REF) and 2050 for the IdF and Stockholm regions due to climate change, emission reduction policies and their combined effect. Results are presented separately for the urban centres (Paris and Stockholm cities) and the domain averages. Ozone is averaged over the April-August period.

	Ozone		**PM_{10}**			**$PM_{2.5}$**		
Paris	**mean**	**MD8hr**	**JJA**	**DJF**	**annual**	**JJA**	**DJF**	**annual**
REF	60	79	22	25	25	15	19	18
clim.	-3.3	-4.1	-1.1	+0.6	-1.1	-1.5	+0.1	-1.2
emiss.	+4.8	+2.2	-4.7	-8.1	-7.2	-5.8	-8.7	-8.1
clim. + emiss.	*+1.5*	*-1.9*	*-5.8*	*-7.5*	*-8.3*	*-7.3*	*-8.6*	*-9.3*
IdF Domain	**mean**	**MD8hr**	**JJA**	**DJF**	**annual**	**JJA**	**DJF**	**annual**
REF	73	92	22	21	23	14	14	15
clim.	-3.7	-4.2	+0.2	-0.2	-0.7	-1.0	0.0	-1.0
emiss.	-6.5	-11.4	-4.0	-6.6	-6.3	-4.1	-6.0	-5.9
clim. + emiss.	*-10.2*	*-15.6*	*-3.8*	*-6.8*	*-7.0*	*-5.1*	*-6.0*	*-6.9*
Stockholm	**mean**	**MD8hr**	**JJA**	**DJF**	**annual**	**JJA**	**DJF**	**annual**
REF	72	81	7	12	10	5.3	10	7.4
clim.	-1.3	-1.7	+0.1	-0.4	0.0	0.0	-0.3	0.0
emiss.	-11	-12.7	-1.3	-2.2	-2.0	-1.0	-1.8	-1.6
clim. + emiss.	*-12.3*	*-14.4*	*-1.2*	*-2.6*	*-2.0*	*-1.0*	*-2.1*	*-1.6*
Stockholm domain	**mean**	**MD8hr**	**JJA**	**DJF**	**annual**	**JJA**	**DJF**	**annual**
REF	73	81.5	6.6	11	9	5	9.5	7
clim.	-1.3	-1.1	+0.1	-0.3	+0.1	0.0	-0.3	0.0
emiss.	-11.4	-13.1	-1.3	-2.3	-1.9	-1.0	-1.9	-1.6
clim. + emiss.	*-12.7*	*-14.2*	*-1.2*	*-2.6*	*-1.8*	*-1.0*	*-2.2*	*-1.6*

Table 5. Contribution of the emission reduction policies implemented at the local and regional scale to the future concentration changes of ozone, PM_{10} and $PM_{2.5}$ in the Stockholm domain.

	Ozone		**PM_{10}**			**$PM_{2.5}$**		
Stockholm domain	**mean**	**MD8hr**	**JJA**	**DJF**	**annual**	**JJA**	**DJF**	**annual**
REF	73	81.5	6.6	11	9	5	9.5	7
local	+0.1	+0.1	-0.1	-0.1	-0.1	0.0	0.0	0.0
regional	-11.5	-13.2	-1.2	-2.4	-1.8	-1.0	-1.9	-1.6
local+regional	-11.4	-13.1	-1.3	-2.3	-1.9	-1.0	-1.9	-1.6

Deleted: Table 6. SOMO35 (in µg m^{-3} days) in the two study regions for the present day simulation (REF) and the future projection (accounting for both climate and emission changes). The relative change between present and future is also given in parenthesis. ¶

Table 6. Future concentration response relative to present (in %) under the high and coarse-resolution applications over the city of Paris and the IdF domain.

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	Ozone		PM_{10}			$PM_{2.5}$		
	mean	**MD8hr**	**JJA**	**DJF**	**annual**	**JJA**	**DJF**	**annual**
Paris high-res	+8	+3	-21	-32	-32	-38	-46	-44
Paris coarse-res	-7	-15	-34	-47	-42	-43	-55	-52
IdF high-res	-9	-12	-18	-32	-27	-29	-42	-39
IdF coarse-res	-9	-16	-29	-41	-37	-39	-52	-49

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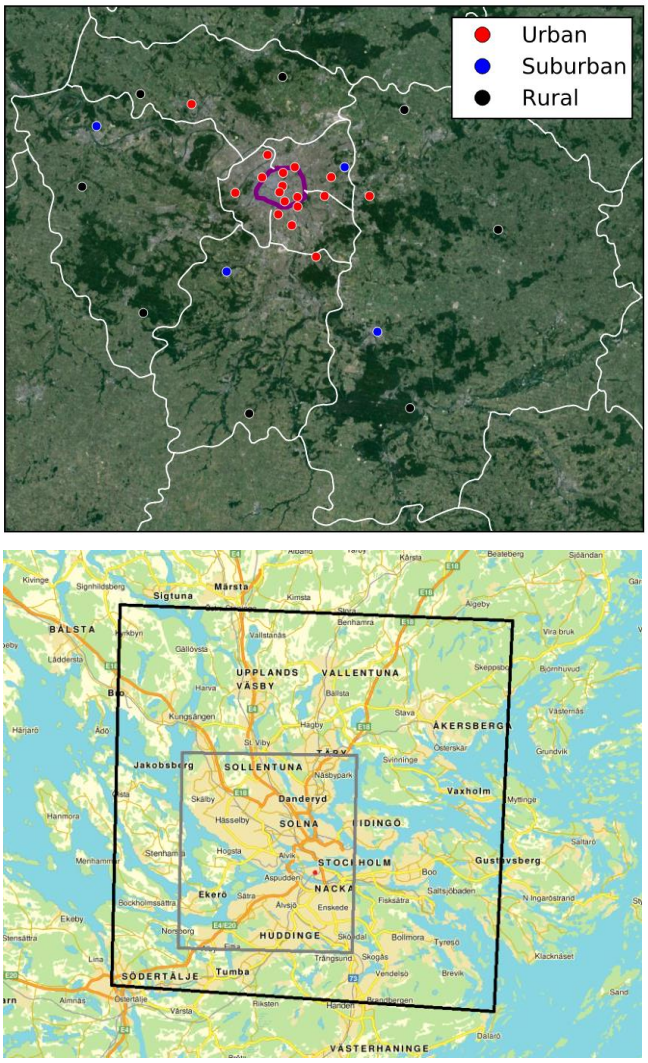
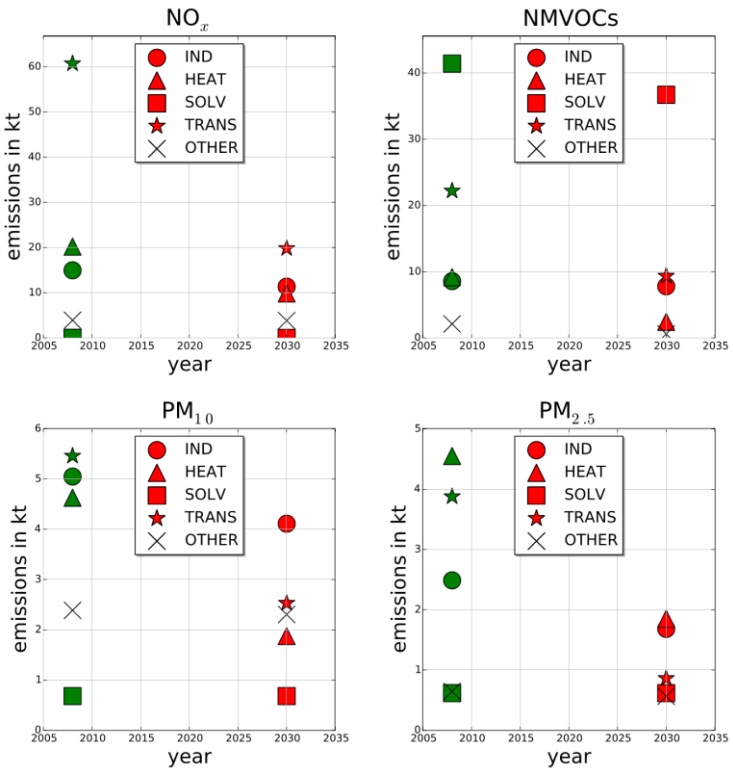


Figure 1. Top panel illustrates the IdF 4km resolution modeling domain, with the city of Paris in the centre (area enclosed by the purple line). Circles correspond to sites of the local air quality monitoring network (AIRPARIF) with red for urban, blue for suburban and black for rural. Bottom panel represents the Stockholm 1km resolution modeling domain (black outline) with the urban area enclosed in the grey rectangle. The red circle corresponds to the urban monitoring site.



1

Figure 2. Annual present-time emissions of NO_x, NMVOCs, PM_{10} and $PM_{2.5}$ in IdF and their projections for 2030. IND corresponds to industrial emissions (SNAP1,3 and 4), HEAT to heating activities (SNAP2), SOLV to solvents use (SNAP6), TRANS to road and non-road transport (SNAP7 and 8) and OTHER represent the remaining source sectors (SNAP5,9 and 10).

2

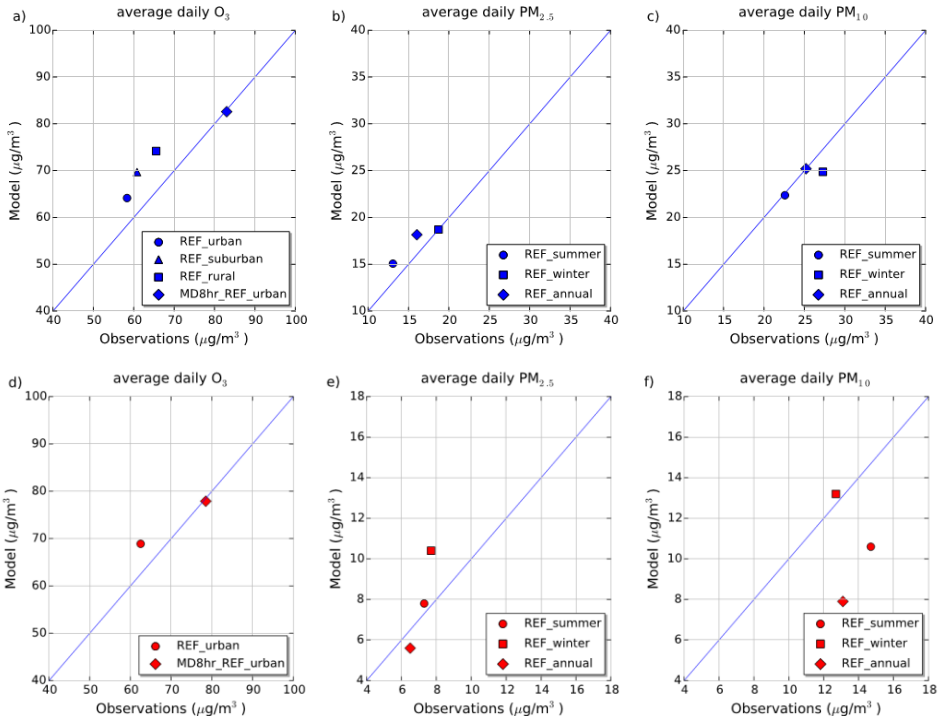


Figure 3. Ozone period (April-August) average ozone concentrations at urban, suburban and rural stations in IdF (panel a) and one urban station in the Stockholm area (panel d). The MD8hr values at urban locations are also shown (MD8hr_REF_urban). Average $PM_{2.5}$ and PM_{10} concentrations in wintertime (DJF), summertime (JJA) and on annual basis over urban stations in IdF are shown in panels b,c (panels e,f for Stockholm).

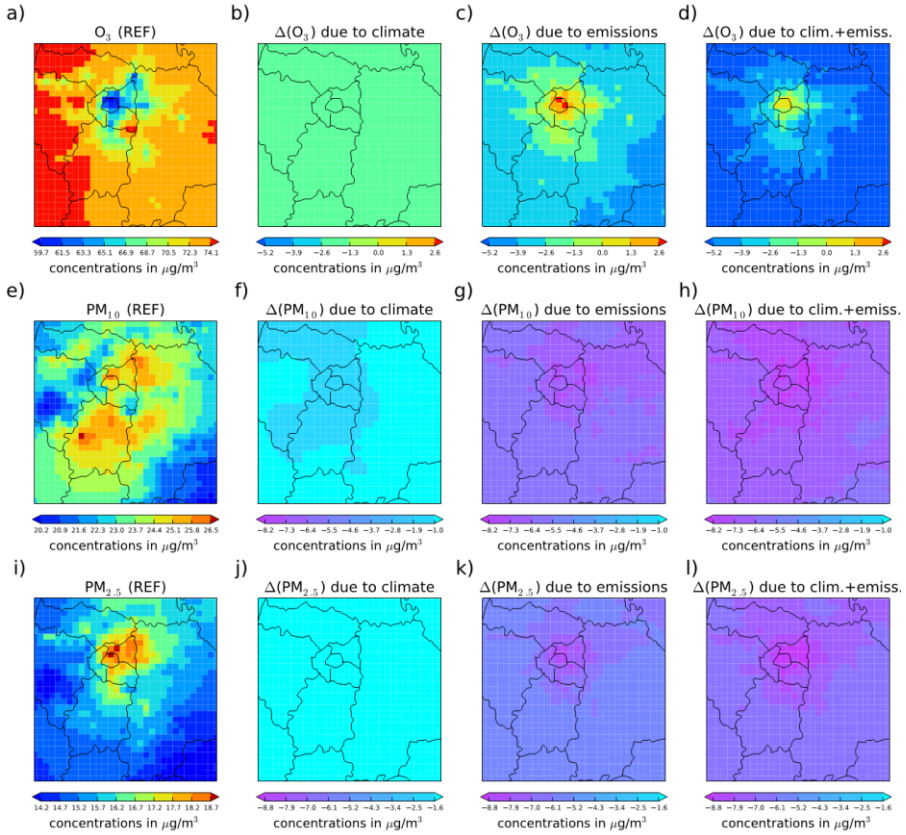


Figure 4. April-August mean ozone, annual mean PM_{10} and annual mean $PM_{2.5}$ concentration maps ($\mu g/m^3$) for IdF, expressed as absolute values at present-time (a,e,i) and as deltas between present-time and 2050 due to climate change (b,f,j), emissions changes (c,g,k) and the cumulative effect (d,h,l).

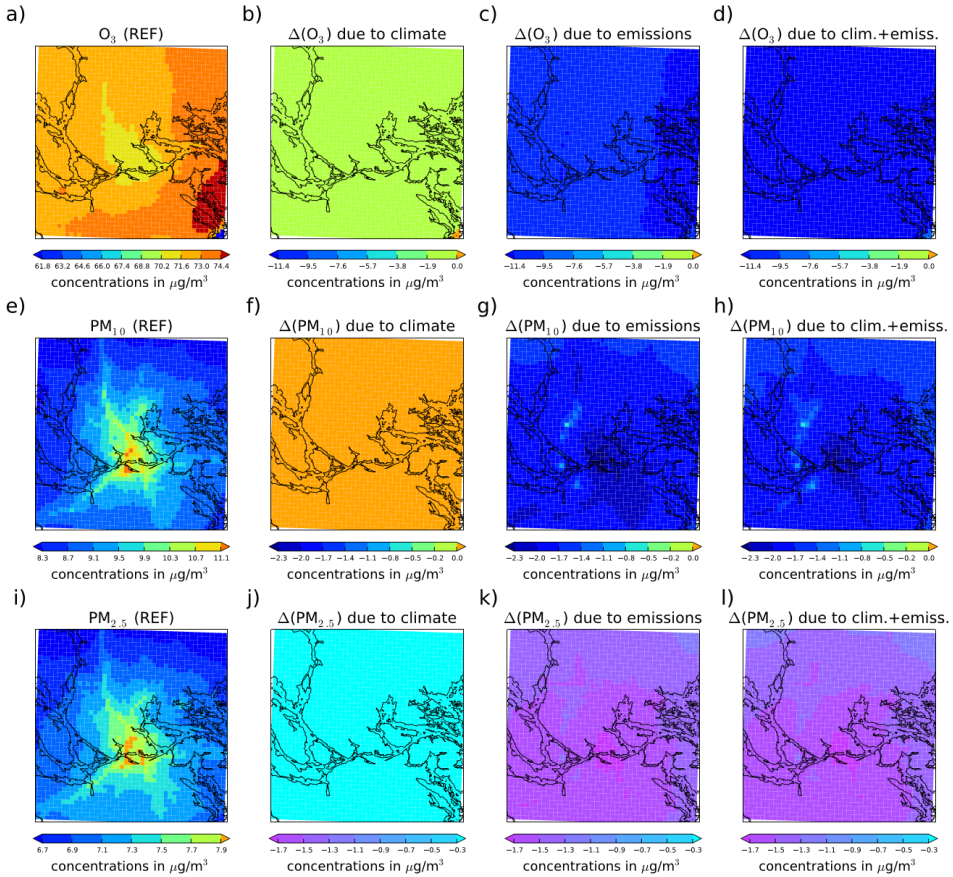


Figure 5. Similar to Fig. 4 for Stockholm.

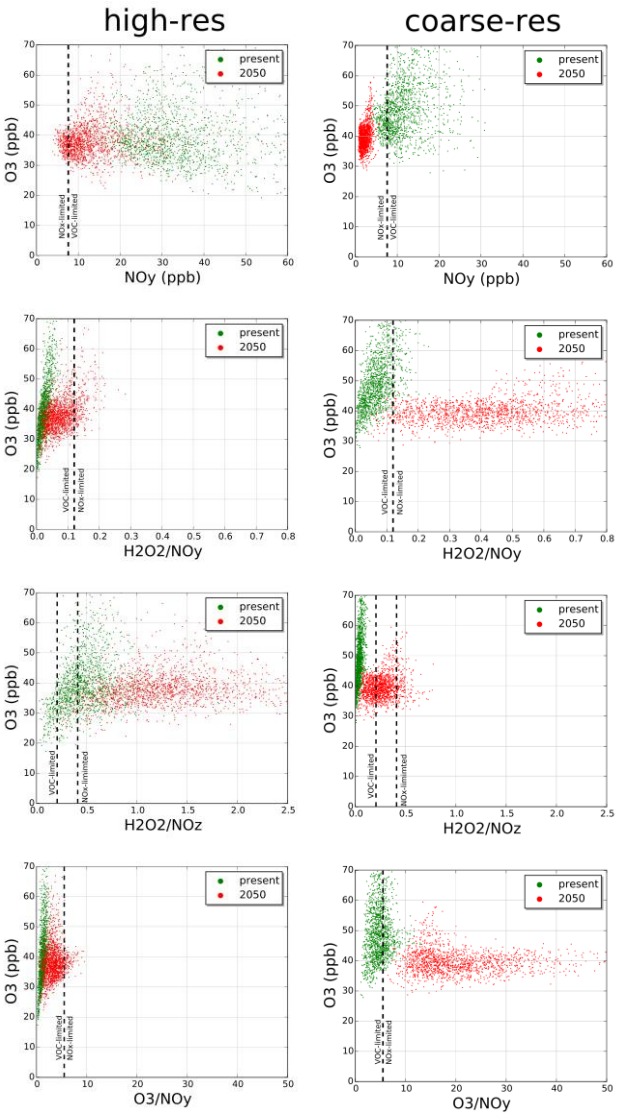


Figure 6. Scatter plots of MD8hr ozone concentrations (y-axis) against chemical regime indicators (x-axis) for the present and future runs in Paris. Results are presented for the high-resolution (left panels) and the coarse-resolution (right panels) applications. All chemical compounds are represented by their MD8hr values. Dots correspond to MD8hr concentrations for each day of the ozone period. For each indicator the limit value that separates the regimes is also depicted with a dashed line.