Eurodelta multi-model simulated and observed PM trends in Europe in the period of 1990–2010

Svetlana Tsyro,Wenche Aas,Augustin Colette,Camilla Andersson,Bertrand Bessagnet,Giancarlo Ciarelli,Florian Couvidat,Kees Cuvelier,Astrid Manders,Kathleen Mar,Mihaela Mircea,Noelia Otero,Maria-Teresa Pay,Valentin Raffort,Yelva Roustan,Mark R. Theobald,Marta G. Vivanco,Hilde Fagerli,Peter Wind,Gino Briganti,Andrea Cappelletti,Massimo D'Isidoro,and Mario Adani

Author comments to Referee #1

Overarching comments:

The manuscript provides a comprehensive evaluation of modeled PM mass and species trends against observations for the 2000 – 2010 time period, with a more limited analysis of trends over the 1990 – 2010 time period. Evaluating whether models can capture historic trends resulting from changes in emissions is of key importance when using such models to predict future changes. The model simulations and analyses presented in the manuscript are sound and of good quality. The manuscript is well written and structured clearly, the presentation quality of all figures and tables is good, and references to other published studies on similar topics are provided where appropriate.

Our reply: We are grateful to the referee for valuable questions and comments, as well as suggestions for editing the text. We have tried to address all issues raised, which we hope has helped to improve the quality of our paper. Our answers to the referee's comments can be found below, in Cursive text format.

My only major comment is that the impacts of not considering forest fire emissions in these simulations should be discussed more prominently in the description of results as well as the abstract and summary. In addition, the list below contains a number of minor comments that the authors may want to consider when revising their manuscript.

Our reply: We thank the referee for this valuable comment. Forest fires were not included in this work partly because of the uncertainties in emissions and modelling of those, but also because we aimed to look at PM trends due to emission regulation in Europe. As we discuss below, there are no model indications that the pollution from the major forest fires had any strong effect on the annual mean PM at the EDT sites. Therefore we do not think that excluding forest fire emissions from the consideration in EDT analysis had any significant consequences on models vs observations comparison.

Though an in-depth analysis of the effects of forest fires on PM trends was not in the scope of this work, it is indeed important to discuss the possible consequences of omitting forest fire emissions in the trend modelling. Since no EDT model ensemble simulations are available in order to address this issue, we made use of trend simulations with the EMEP

model which included FINN (Fire INventory from NCAR) emissions. We looked at the effect of primary PM (which were available as individual tracers) from fires on PM10 and PM2.5 trends and also at the enhancement in secondary organic aerosol due to exceptionally large 2010 Russian forest fires and other major fires and included a short discussion based on the main findings (see further replies below).

EMEP trend simulations show only minor effects from primary PM emissions from forest fire to PM2.5 trends in Russia and Eastern Europe and negligible ones in other parts of Europe (Fig. R1). The EMEP model simulates enhanced concentrations of secondary organic aerosols (SOA) associated with the Russian fires (due to the enhanced amount of pre-existing organics and more efficient VOC oxidation by ozone), but most of the pollution from those fires mostly stayed over European Russia due to particular air circulation in the stagnant high-pressure system. Witte, J. C., et al. "NASA A-Train and Terra observations of the 2010 Russian wildfires." Atmospheric Chemistry and Physics 11.17 (2011) found that elevated levels of aerosols detected by satellites over western Russia corresponded to the time period of peak wildfire activity (22 July–18 August 2010) and a persistent anticyclonic circulation. In this situation air was trapped, allowing smoke pollutants to accumulate as the air mass re-circulated. Also Konovalov et al (2011) wrote that the accumulation of anthropogenic pollution over European Russia was favoured by a stagnant and dry meteorological situation (Konovalov, I. B., et al. "Atmospheric impacts of the 2010 Russian wildfires: integrating modelling and measurements of an extreme air pollution episode in the Moscow region." Atmospheric Chemistry and Physics 11.19 (2011): 10031-10056).

A discussion on this is included in Section 7.2 "Uncertainties in emissions" (see PMtrends_diff.pdf). The issue is mentioned in Summary ("Not accounting for forest fires in EDT simulations should also affect the accuracy of simulated PM trends, at least in the regions of large fires, whilst this does not appear to have a major impact on the modelled trends at the EDT sites") and in Abstract: "Further discussions are given with respect to emission uncertainties, implications of not accounting for forest fires…"
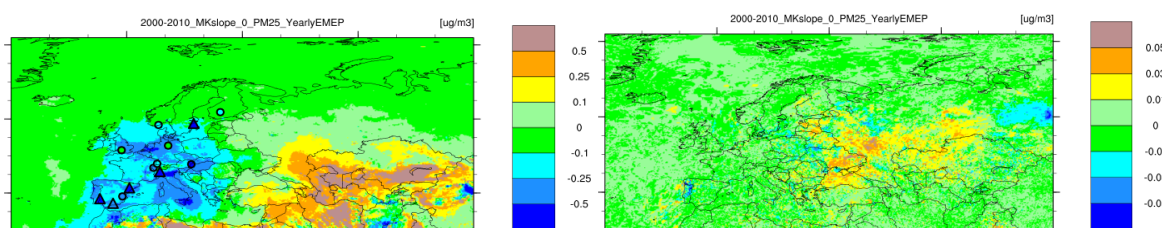

Fig R1. EMEP modelled trends for PM2.5 including forest fires emissions (left) and the difference in PM2.5 trends when PM emissions from forest fires are included and excluded (right) (shown are both significant and insignificant trends)

Specific comments:

Line 25: maybe mention that the exclusion of forest fire emissions and (in some models) windblown dust emissions may have contributed to a higher fraction of modeled trends being

significant due to the lower amount of interannual variability being present in such model simulations

Our reply: The referee is right that excluding forest fire emissions in EDT simulations and not accounting for windblown dust by some of the models (CHIM, MATCH and POLR only included dust from Boundary conditions) may have smoothed PM annual variations, which could have resulted in a larger fraction of modelled significant trends in the areas affected by those emissions.

Regarding forest fires, most of the EDT trend-sites were not significantly influenced by those emissions (see our comments above) so that model evaluation against observed trends in this respect was not feasible. Effects of mineral dust on PM trends, namely stronger PM modelled trends and larger fraction of significant ones compared to observations, should be seen more obviously for Spanish sites influenced by mineral dust. However, Figure 6 shows that the model ensemble trends of PM10 and PM2.5 are not significant for all Spanish sites, whereas observations indicate that there were significant trends at 4 out of 6 Spanish sites for PM10 and 3 out of 4 sites for PM2.5. Further, we could expect to see weaker and more of not significant trends simulated by EMEP, LOTO and MINNI, which accounted for both African dust (from boundary conditions) and windblown dust within the simulation domain, still the results are rather mixed (Figure A8). For example, both EMEP and MINNI include windblown dust; while EMEP simulates significant PM10 and PM2.5 trends for the most number of sites compared with the other models, MINNI doesn't recognise any significant trend at any of the Spanish sites.

Line 61: suggest inserting "over the" before "last years"

Our reply: The suggestion is followed.

Lines 80 – 83: as noted below, to more fully accomplish this goal, it would be beneficial to provide more information on the speciation of primary PM2.5 emissions (especially EC and OC) and the representation of secondary organic aerosols in the models, and then reference this information when discussing speciated results in section 5.4

Our reply: We agree. The details regarding PM2.5 emissions speciation to elemental and organic carbon, as well as SOA representation in the models are included in the new Table A16: Main features of the Chemistry-Transport Models involved in the Eurodelta-Trends modeling exercise.

Line 96: insert "that" before "participated"

Our reply: Done

Line 109: insert "was" before "performed"

Our reply: Done

Lines 105 – 116: Please provide information how the ECLIPSE PM2.5 and PM10 emissions were speciated into different compounds. If this was handled differently for each model, please provide a summary of the approach for each model to better understand the results presented in section 5.4

Our reply: The models were not required to use the same split of the Eclipse PM2.5 and PM10 emissions. The details regarding PM emissions speciation to elemental and organic carbon is included in the new Table A16: Main features of the Chemistry-Transport Models involved in the Eurodelta-Trends modeling exercise, and are referenced to in the discussion in Section 5.4.

Lines 113-114: Please provide more information on the temporal resolution the ECLIPSE inventories for the different sectors – are these all annual total emissions, and did each model then apply the same EMEP profiles to perform monthly, weekly, and diurnal allocation? Did the vertical distribution of emissions depend on the meteorology used by each model to account for the effects of meteorology on plume rise, or are these EMEP profiles static? Given the effects of atmospheric stability on near-source PM concentrations, uncertainties in representing plume rise may be an important factor of model error especially during wintertime.

Our reply: For temporal distribution of ECLIPSE annual emissions, the models applied the same monthly and hourly profiles prepared by TNO (Kuenen et al., 2014); also the same static emission vertical profiles (per sector) provided by INERIS were used. This information is included in section 2.1 (Models, runs setup) and Table A16.

Lines 126-127: The exclusion of forest fire emissions (and in some models dust emissions) may have important implications for the interpretation of modeled and observed trends, specifically the smaller amount of interannual variability the exclusion of such emissions causes in the models which in turn leads to a tendency for more significance in the trends estimated from the models. The current version of the manuscript does not reference the exclusion of fire emissions in any of the discussions in sections 4 – 7.

Our reply: We thank the referee for this valuable comment. A discussion regarding possible implications of omitting forest fires on modelled trends has been included in Section 7.2 Uncertainties in emissions: "Finally, as described in ~\ref{sec:models}, pollution from forest fires were not accounted for in EDT simulations mainly because of considerable uncertainties in forest fire emissions and modelling of those, but also because we aimed to look at PM trends due to emission regulation in Europe. An in-depth analysis of the effect of forest fires on PM trends is beyond the scope of the paper, but we have tested whether the discrepancies between the modelled and observed trends, in particular in terms of a relatively larger fraction of significant trends from the model results, could be due to not including forest fire emissions in the EDT simulations. Additional simulations suggest that the effects from even large fires during the studied period (like 2010 Russian forest fires) were mostly negligible outside the regions where wildfires occurred. In fact, the pollution from major forest fires did not seem to have any large impact on simulated annual mean PM at the EDT sites in the 2000-2010 period. Therefore we believe that not accounting for forest

fires in EDT analysis did not have any significant consequences for models vs observations comparison." References to this discussion have been added where relevant.

Section 2.1: It would be useful if this section also included a summary of the aerosol treatment of the different models (e.g. number of modeled species, sectional vs. modal size distribution representation, representation of biogenic and anthropogenic secondary organic aerosols, etc.) to help with the interpretation of results from individual models in subsequent sections.

Our reply: This definitely useful information has now been included in the new Table A16: Main features of the Chemistry-Transport Models involved in the Eurodelta-Trends modeling exercise.

Lines 136 – 146: Please clarify if the modeled annual mean values at the observation locations were computed by discarding any time periods for which observations were not available at that site. In other words, if a given site had 80% data completeness in a given year, was the corresponding model mean for that site and year computed over the same 80% of all modeled time periods in that year? Such temporal matching of observations and model values at the underlying temporal resolution and completeness of the observations would be the preferred approach for ensuring consistency between the observed and modelled annual means.

Our reply: Very relevant question. Annual mean concentrations were calculated individually for observations and model results. We recognise that including only common days would give a more accurate comparison between the models and observations, but we believe that the uncertainties associated with the adopted approach are in general small enough and that most of the results from the presented trend analysis are valid.

Lines 175 – 176: If possible, it might be interesting to discuss which meteorological and/or emission features (e.g. precipitation, dust or fire emissions) may have caused the elevated PM levels in 2003, 2006, and 2010

Our reply: Though an in-depth investigation of the causes of PM inter-annual variations is beyond the scope of the paper, we made a closer look at the timeseries at the individual sites. It appears that that the enhanced PM in 2003, 2006 and 2010 were mostly due to high annual PM at Central-European sites (primarily in Germany, Austria, Switzerland), rather far away from major forest fires.Those elevated annual PM were due to heat waves in summers 2003 and 2006 (no PM removal by precipitation in dry weather conditions, efficient BSOA formation) and extensive PM episodes in winter 2010. Furthermore, based on the results from independent tests with the EMEP model, we do not believe that PM increases in 2003, 2006 and 2010 were due to forest fires as the trend sites are situated in the locations not (or negligibly) affected by the fire emissions. The year 2010 was already discussed in Section 5.1 (lines 265-273), a short discussion was added concerning 2003 and 2006.

Line 226: consider changing "does not probably indicate" to "hardly indicates"

Line 254: change LOTOS to LOTO for consistency

Line 261: change "they are an underestimation" to "they are underestimated"

Lines 283 - 284: remove double parentheses

Our reply: Thank you for spotting the typos and suggesting better formulations

Lines 338 – 343: This essentially seems like a repeat of the results in lines 291 – 296. Both paragraphs discuss the results averaged over all sites.

Our reply: We thank the reviewer for spotting that. We have sorted out the repetitive discussions on the site average ensemble simulated trends and focus analysis in Section 5.2 on the trends from the individual models.

Line 358: suggest changing "calculated" to "modeled"

Line 397: suggest inserting "between species" after "not the same"

Our reply: The suggestions above were followed

Lines 399 – 400: I would suggest converting these results from ugN/m3 and ugS/m3 to ug/m3 for consistency with the analysis of total PM mass

Our reply: Traditionally, ugN/m3 and ugS/m3 have been for decades used in EMEP for sulphur and nitrogen components in model vs observation comparison, but we agree with the reviewer that it's a good idea to use ug/m3 for all species it this study. We have made the unit conversion and new plots are included.

Furthermore, while making the unit conversion, we have spotted that there appeared to be a unit inconsistency in calculations of the contributions of SO4, NO3 and NH4 to PM trends. This has been corrected and the correspondent maps and numbers have been updated in the last manuscript version.

Slightly modified text (lines 415-424): "The decrease in SO_4^{-2} concentrations (Figure 10a) played the dominating role over most of the EDT domain, except from parts of Central Europe and Northern Italy. Namely, relatively large contributions of NO_3^- to PM_{10} trends are seen in Germany (and neighbouring parts of France, Czechia and Poland), Denmark, the Netherlands, and in the Po Valley (Figure 10c). The reduction of NH_4^+ levels, which includes both ammonium sulphate and ammonium nitrate, appears to be quite an important contributor to the PM_{10} decreasing trends, with the largest effects estimated for Poland, Denmark, and the Po Valley (Figure 10b). The reduction of primary PM emissions was according to the model ensemble simulations the dominating factor for PM_{10} trends in Portugal and southern parts of the Balkan; as well as in many European cities (due to emission reductions from traffic and residential heating) (Figure 10d)."

Line 453: change "dependency of" to "dependency on"

Our reply: The suggestion above was followed

Line 578 and 581: what is the estimated uncertainty of VOC and NH3 emissions?

Our reply: NMVOC emission estimates in Europe are thought to have an uncertainty of about ±30% due in part to the difficulty in obtaining good emission estimates for some sectors and partly due to the absence of good activity data for some sources (APE 004Published 15 Oct 2010 Last modified 04 Sep 2015 https://www.eea.europa.eu/data-and-maps/indicators/emissions-of-ozone-precursors-version -2/eea-32-non-methane-volatile-1). NH3 emission estimates in Europe are due largely to the diverse nature of major agricultural sources (https://www.eea.europa.eu/data-and-maps/indicators/eea-32-ammonia-nh3-emissions-1). It is estimated that they are around ±30%. For both NH3 and NMVOC, the trend is likely to be more accurate than the individual absolute annual values. Primary $PM_{2.5}$ and PM_{10} data is said to be of relatively higher uncertainty compared to emission estimates for the secondary PM precursors.

Lines 588 – 593: The authors may want to explicitly discuss that in the real world, these emissions are dependent on meteorology (higher on colder days) while this effect likely is not represented in the current models.

Our reply: That's a very relevant point. A brief discussion has been added. It is also worth noting that the models did not account for the dependence of residential heating emissions on the outdoor temperature, i.e. they increase as it gets colder. This may lead to model underestimation of winter pollution episodes, resulting in under-predictions of annual mean PM (as for 2010, see Sec. 5.1).

Line 589: year is missing for the Simpson et al. reference

Our reply: Thanks for spotting that - the year is inserted!

Line 628: what is the authors' interpretation of this finding? Also see my earlier comments on fire and dust emissions.

Our reply: We have re-written the text on lines 615-628 to provide hopefully a more clear interpretation of the results. Regarding our finding that statement "..large number of sites (and areas) where the models do not estimate significant trends for 2000-2010 period", our explanation is that due to considerable inter-annual variability in PM concentrations and not very strong pM trends, 10-year period is probably not long enough for identification of significant trends with high confidence.

Line 639: consider changing "downsized" to "partially masked"

Our reply: The suggestion is accepted.

Line 665: this section does not include an outlook, consider changing the section title to just "Summary"

Our reply: *Thanks, it's changed.*