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Interactive comment on "Climatology of atmospheric PM$_{10}$ concentration in the Po Valley" *by* A. Bigi and G. Ghermandi

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Sincere thanks to referee #2 for the constructive comments, for highlighting the weak points of the manuscript and giving the chance to improve the study.

Reply to general comments:

1. the referee is certainly correct in stating that a climatological analysis is conventionally defined upon a 30 years or longer series of data, and it is very true that studies similar to ours refer often themselves as "long term trend analysis". We had the audaciousness of using the word "climatology", although not strictly correct, since our study is larger than a straight long term trend analysis, involving

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weekly periodicity, cluster analysis and relations to trend in emissions, therefore we had considered the word "climatology" more inclusive and fitter to the study, although not strictly correct and eventually misleading. For the Revised Manuscript (RM) we might consider to change the title to: "Long term trend and variability of atmospheric PM$_{10}$ concentration in the Po valley"

2. A clearer discussion on weekly cycle has been added in the RM, as shown in the reply to the following comments, including role of secondary and possible outlooks for future analyses. Seasonal trends, being significant over most of the year, furtherly support the hypothesis that estimated trends are scarcely influenced by occasional meteorological conditions (see reply to referee #1) and are driven by emissions. These notes have been added to the RM.

3. Referee #2 is correct in pointing at the lacking description of the inventory used in the analysis. A thorough description of the inventory will be included in the RM, including the key features discussed hereafter.
Methodology & Uncertainty
Methodologies used to build the national inventory "are based on and conform to the EMEP/CORINAIR guidebook, the IPCC Guidelines and the Good Practice Guidance" (Romano et al., 2012). Emissions for SNAP 7 for the national inventory (i.e. not disaggregated at a provincial scale) derive from COPERT 4 v.9.0. For the national inventory "uncertainties in PM emissions stay, even if the inventory accounts for non-exhausts PM emissions" (Romano et al., 2012). "An overall uncertainty analysis for the Italian inventory has not been assessed yet", besides a general assessment of the uncertainty for GHG emissions (Romano et al., 2012). Top-down disaggregation of the national inventory followed the procedure described in De Lauretis et al. (2009).
Secondary particles
Referee #2 inquired about emissions of secondary particles for SNAP 7. Giving to "secondary particle" the commonly accepted definition of particle formed by

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gas-to-particle conversion processes (Raes et al., 2000), secondary particles are not included in SNAP. SNAP provides codes for non-exhaust PM emissions, for "direct" PM emissions and for gaseous emissions (either in exhausts and in non-exhausts).

Clarification on non-exhaust particulate emissions from road traffic

The national (i.e. non-disaggregated) inventory accounts for SNAP sectors 0707 "tyres and brakes abrasion" in PM_{10} and $PM_{2.5}$ emissions (Bernetti et al., 2010). Also the disaggregated inventory accounts both for primary and non-exhaust PM emissions, although not explicitly stated in the Original Manuscript (OM). In line 1 page 153 with "primary" we intended "non-secondary" i.e. primary PM emissions from exhaust and non-exhaust. We agree with referee #2 that the sentence in line 1 page 153 might be misleading and in the RM it will be replaced by "of the role of primary (including non-exhaust) PM emissions on the observed decrease in atmospheric PM_{10} in the Po valley."

4. the decrease in PM_{10} emissions from SNAP7 despite the dieselization of the fleet has been ascribed to the strong renewal of vehicles, forced by driving restrictions to older vehicles since 2002 in the Po valley (page 151 lines 19 and following in the manuscript). This assumption is consistent with the lower PM emission rates for diesel engines compliant to most recent European emission standard (e.g. see figure 1 from Copert 4 v.10.0 report (Katsis et al., 2012)).

5. Referee #2 raises one of the questions left open by the manuscript, which eventually has been not sufficiently discussed in the text. NM-VOC has been largely decreasing over the valley (see figure S2a and note that figure S3 in the OM shows the change in percent contribution by each SNAP sector, not the absolute value, since absolute trends are already shown in figure S2). Also NO_x has decreased, although in fewer provinces (as shown in figure S2b). Therefore it cannot be stated that PM_{10} trends are driven by trends in PM_{10} primary emissions from SNAP 7, given also the large fraction of secondary particles in PM_{10}.

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A combined study of $PM_{2.5}$ and PM_{10} trends might help to highlight the role of secondary and primary coarse particle in the identified PM_{10} trends, although this analysis would deal only with data from year 2006, when $PM_{2.5}$ measurements started. A note on this outlook has been added to the RM.

6. Answer to this point has been addressed together in the answer to observations by referee #1

Replies to SPECIFIC COMMENTS AND SUGGESTIONS are listed below (comments are in italic, answers in plain text):

- Page 139, line 13; Check the use of "renown", which is a substantive, not an adjective.

"Renown" has been changed to "well-known"

- Page 139; the works of Matta et al., (2003) and Putaud et al., (2010) present similar results and these two references could result somehow redundant not providing added information. Perhaps they should be included in the same sentence by giving a range, i.e. 40-44 % for the contribution of those species.

The two sentences have been merged in the following: "In Bologna urban background Putaud et al. (2010) and Matta et al. (2003) found a concentration range of 40–44% of ammonium, nitrate and sulphate in $PM_{2.5}$ and PM_{10}."

- Page 140, Line 9; studies on CO_2, which is not an atmospheric pollutant but an atmospheric component, are focussed on global atmosphere composition trends and for this reason should not be referenced in an atmospheric pollution study. On the other hand CO_2 is usually measured at remote sites not comparable to the local or regional scale ones used for air pollution studies. Ciattaglia et al., (1987) and Artuso et al. (2009) references should be withdrawn from the text for that reason.

We agree with the referee that CO_2 is an atmospheric component, as PM_{10} from deserts or SO_2 from volcanoes. Seinfeld and Pandis (2006) include CO_2 among atmo-

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spheric pollutants, whom they define as "any substance that result from anthropogenic activities and that is present at concentrations sufficiently high above their normal ambient levels to produce a measurable effect on humans, animals, vegetation, or materials" (in agreement with the 2008/50/EC). In the OM references about CO_2 studies have been cited since, to our knowledge, are among the very few investigations on long term (e.g. $\geq$ 10 years) trends in the atmospheric constituents in Italy, along with Bigi et al. (2012) and a just published study by Tositti et al. (2014) on long term trend of ^7Be and ^{210}Pb at Mount Cimone (added in the RM). Eventually the references on CO_2 could be removed in the RM.

- Page 140, Line 12; What is the time period analysed in Bigi et al (2012)? It should be mentioned in the text.

That study investigated the 1998–2010 period. In the RM the sentence has been changed to "Bigi et al. (2012) found a decreasing trend for many pollutants in a urban background site in Modena, Po Valley, over the period 1998–2010. "

- Pag 143 Line 7; "Indipendent" should be "independent". Check spelling.

Amended in the RM.

- Pag 147. Line 12; Although it appears in the title of the paragraph, it could be specified better also at the beginning of the first sentence: "Emission time series. . ." just for clarification.

Amended in the RM.

- Pag 148; Although in the referenced work of Harrison et al., 2008 is mentioned that no decreasing trend in PM_{10} concentrations has been observed in other regions of Europe during the period 2002-2011, there is no reference on this in this work, which deals with UK observations. Have the authors found any other work that illustrates such flat behaviour of PM_{10} concentrations in Europe during the same time period?

The work of Harrison et al (2008) has been cited since the authors addressed their

point to a large region. Steady or eventually increasing PM_{10} concentration over Europe have been observed at single sites in several studies, although some of these trends might be highly site-dependent. Anntila and Tuovinen (2010) showed that mean hourly PM_{10} concentration resulted steady at several sites in Finland over the period 1994/1998–2007/2008. Barmpadimos et al. (2012) showed a steady daily average PM_{10} at the EMEP site of Langenbruegge/Waldhof over the period 1999–2010 (both for measured and meteorologically-adjusted concentrations) and still Barmpadimos et al. (2011) showed a steady trend for annual average PM_{10} at few sites in Switzerland (period 1991–2008). The sentence in line 10 page 148 has been changed to ", contrarily for instance to the trends observed in the U.K. (Harrison et al., 2008)."

- Pag 149. Line 1; Similar trends as those resulting from this work are mentioned in the text have been found for other sites in Europe. Could the authors specify the period analysed by Barmpadimos et al 2012?

In the RM the sentence has been changed as follows: "Trend slopes are similar to other sites in Europe: Barmpadimos et al. (2012) found a PM_{10} decrease ranging between -0.5 to -1.3 $\mu g/m^3$ in five rural sites within the EMEP network over the period 1999–2010 and ascribed most of decrease to a change in $PM_{2.5}$ concentrations."

- Pag. 149 Line 20; The text includes a comment about observed weekly cycle during specific seasons (winter). Can this affirmation be supported by any figure?

We sincerely thanks the referee for forcing us to double check once more the weekly cycle analysis. In line 21 of page 149 is stated that Forlì and Sannazzaro were the only two sites without a weekly cycle *in winter*, whereas these two sites do not show a weekly cycle if the *full year* is considered (along with Febbio, line 13 page 149). We apologize for this oversight, which came along with another quite gross errata. i.e. the extra word "winter" in line 20 page 149: as shown in table 4, PM_{10} exhibit a weekly cycle in winter at few sites, consistently with the discussion from line 24 in page 149. Amendments included in the RM are as follows:

1. a figure with for winter and summer 7 day week mean PM_{10} anomaly has been added as figure S2 in the supplementary material in the RM and enclosed in this reply as figure 1.

2. Page 146 line 14 has been changed accordingly "while graphs of 7 day week mean anomaly for all sites are presented in Fig. S1 and Fig. S2 for full year and seasons respectively."

3. Paragraph from line 20 of page 149 to line 1 of page 150 has been changed accordingly in the RM:
"Results from the two tests for weekly periodicity are highly similar. Considering the whole year, a significant weekly periodicity is present during 7 day week at all sites besides Febbio (accordingly to weekend effect magnitude only), Forlì and Sannazzaro (see Figure S1). As shown in Figure S2 and table 4, most of shorter time series show a weekly periodicity in summer and not in winter, whereas many of longer ones still exhibits a weekly periodicity in both seasons. The lack of weekly periodicity in winter might be due to the large fraction of Secondary Inorganic Aerosols (SIA) in PM_{10} in this season (Larsen et al., 2012), uncoupling the weekly fluctuations of primary anthropogenic emissions (non-exhaust included) and PM_{10} concentration. This behaviour has been observed by Bernardoni et al. (2011) in Milano urban background conditions, where relative contribution of direct human-related particulate sources (e.g. re-suspension, traffic, industry) to PM_{10} is higher in summer than in winter, consistently with a significant periodicity in summer weeks. Possibly this buffering effect by SIA is dimmed in longer time series by a higher primary/SIA ratio in the late 90s early 2000, leading more likely to significant weekly cycles in winter, although this hypothesis should be substantiated by further analyses. Test of weekly cycles for 6 and 8 day weeks resulted non- significant for all sites besides Magenta in winter 6 day week and Voghera in the complete series 8 day week."

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- Pag 151. Line 7; The text mentions the thematic maps of emission trends for NO_x, CO and PM_{10} appearing in Fig S2. It must be a mistake as maps in such figure are for NM-VOCs, NO_x and PM_{10} pollutants.

Thanks to referee #2 for pointing this oversight out. The RM lists the pollutants effectively corresponding to figure S2 (S3 in the RM).

- Pag 151; How do the authors support the conclusion that despite no correlation has been found between PM_{10} emission trends and PM_{10} observations at background sites, the drop observed in the latter derive from an overall decrease of emission in the Po valley?. What about other causes (meteorological) or specific sources? Must be this uncorrelation attributed to the emission inventory uncertainties?

Even if we looked for a linear correlation between PM_{10} emissions and PM_{10} atmospheric concentration trends, being the simplest model to test, we do not think it is so surprising this lack of direct proportionality, since the PBL and the atmosphere represent such a complex system where linearity is hardly present, particularly in PM_{10} formation. We therefore consider that the linearity between PM_{10} concentrations and emissions is not missing because of an uncertainty in the inventory, notwithstanding this is inevitably large. Moreover, as pointed out by the several studies cited in the introduction, secondary PM_{10} in the Po valley (e.g. nitrate, sulphate, ammonium) can be as large as 50% of total PM_{10}. We have discussed already about the effect of meteorology on trends in the answer to referee #1. We have no knowledge of other specific sources which can have influenced PM_{10} atmospheric concentration at a valley-wide scale and on a long-term scale.

- From figure S2 overall PM_{10} emission decrease is very low and only in a few provinces of the Eastern sector of the Po valley. On the other hand, total numbers for PM_{10} emissions (Fig S3) would explain a decrease only from the SNAP7 (Road Transport sector), but it is not the case for the SNAP2 (Commercial, institutional and residential combustion plants), which exhibits a significant increase of PM_{10} emissions.! What

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is the source apportionment (SNAP sectors) for the total PM_{10} *emissions in the Po valley?*

The use of biomass burning tracers as levoglucosan has revolutionized source apportionment of atmospheric PM_{10} in the Po valley and subsequently also inventory of PM_{10} emissions by domestic heating and biomass combustion plants. It is also worth noting that estimate of PM_{10} emissions by biomass burning for domestic heating suffers from large uncertainty due to the difficulties in collecting data on biomass consumption and existing domestic sources. The source apportionment of PM_{10} emissions in the Po valley for each SNAP sector is exactly the lowest left panel in figure S3 (S4 in the RM): the ordinates in the barplots in figure S3 are the percentage contribution to total emissions (i.e. these are not "total numbers"). Indeed, the disaggregated inventory estimates a doubling or even a tripling of absolute emissions of PM_{10} by SNAP 2 at several Po valley provinces over the period 2000–2010. However for each province either a decreasing or a steady trend for total PM_{10} emissions is observed, since the absolute increase in PM_{10} emissions by SNAP 2 is compensated by the decrease by other SNAP sectors.

- Pag. 151; Figure S3 shows averaged emissions for the whole Po valley after different SNAP sectors. As PM_{10} *emissions trends have been performed showing differences at province level, it could be interesting to do the same analysis of SNAP sectors variations at province level.*

Po valley is shared by 25 provinces and we analysed emissions of 8 pollutants, leading up to 200 barplots. It could be certainly interesting to investigate variations at a province level in contribution to emissions by each SNAP sector for a highly involved reader, but we fear it could result in a excess of information and ultimately being almost useless or eventually confusing for the majority of readers, even providing "only" 25 barplots for a single species. Eventually, if required, we might add thematic maps of trend for the 5 pollutants not included in figure S2.

- Pag. 152; Line 4. Which is presently the degree of dieselization of the Po valley fleet?

In the OM information on trends in fuel consumption have been preferred to those on dieselization of the fleet. Available statistics on vehicle typology, fuel and province do not overlap perfectly to SNAP 7: based on Italian Automobile Club statistics and considering passenger cars, LDV and HDV, in 2002 diesel vehicles in the Po valley were ∼26%, in 2011 diesel vehicles were ∼42%. In the RM at page 151 line 18-19 we added this information: " [. . .] LDV. This increase occurred along with a dieselization of the fleet, with the rate of diesel vehicles (considering passenger cars, LDV and HDV) raising from ∼26% to ∼42% over the period 2002-2011, along with a renewal of the fleet. Changes in vehicular fleet composition [. . .]"

- Pag 152. Line 6. The study of Meij et al., 2009 refers to $PM_{2.5}$ *(for the Lombardia region). As no information on* $PM_{2.5}/PM_{10}$ *ratios has been included in this study, results on trends could not be comparable.*

We agree with referee #2 that the study of Meij et al (2009) is focussed on $PM_{2.5}$, as stressed in the OM, even if Meij et al. provide few simulation outcomes also for PM_{10}. Perhaps, in the OM it was not sufficiently stressed that the two simulation studies have been cited for a rough comparison. These studies have been used because, to authors knowledge, were the only two published studies on simulation on PM concentration change by emission scenarios in the Po valley. A further note about this has been added to the RM. In page 140 line 6 there is a note on common $PM_{2.5}/PM_{10}$ ratios in the Po valley, which has not been reminded in the Results and discussion paragraph. In the RM we added a note on these cited ratios. We also added the mean $PM_{2.5}/PM_{10}$ ratio in table 5; these range from 0.61 (Parma) to 0.94 (Cerano).

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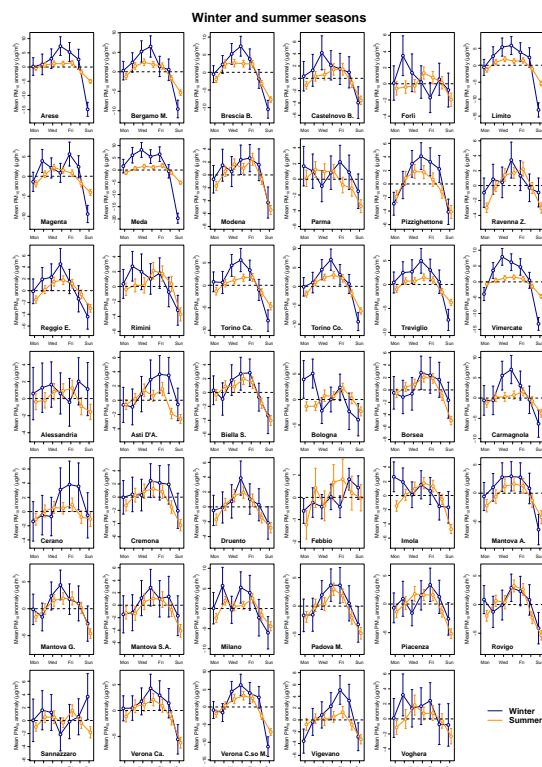
Fig. 1. 7 day week mean PM10 anomaly for winter and summer seasons for all sites listed in table 1 (vertical bars indicate standard deviation)