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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantifying the contribution of long-range transport to Particulate Matter (PM) mass loadings at a suburban site in the North-Western Indo Gangetic Plain (IGP)" *by* H. Pawar et al.

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- **Reviewer comment:** This study aims to quantify the contribution of long range transport and local sources to PM loading at a station in IGP using back trajectory model and observational data at receptor site. In general, this paper is well written. The results are informative. The reviewer recommends to accept to publish this study after some modifications and clarifications.

Authors' response: We thank reviewer #2 for this compliment and his/her helpful comments, which have improved the clarity of the presentation further.

- **Reviewer comment:**

  Major comments: Could you address the uncertainty and limitation of the quantifications derived from this study in order to guide the reader to apply the results later? -

  Authors' response: The uncertainties pertaining to figure 9 have already been presented in table 3, which provides the significance level for the difference of the mean for each pair of clusters. We have added the uncertainty of the enrichment above the regional background to table 2.

  Modifications in the text: The revised table(table 2) now reads as detailed below:

Table 1. Lower limit for the contribution of long range transport and local pollution events to PM mass loadings in % $\pm 1\sigma$ of the total PM. "Negative" indicates that the PM mass loadings are not enhanced compared to the local cluster, which represent the regional background levels.

	Fast Westerly	Medium Westerly	Slow Westerly	South Westerly	South Easterly	Calm
			$PM_{2.5}$			
Winter	Negative	7 ± 4	13 ± 9	Negative	Negative	22 ± 15
Summer	20 ± 15	4 ± 3	31 ± 21	10 ± 8	Negative	13 ± 8
Monsoon	Negative	Negative	15	Negative	Negative	Negative
Post-Monsoon	18 ± 10	11 ± 7	Negative	Negative	Negative	Negative
			$PM_{10-2.5}$			
Winter	18 ± 8	28 ± 16	22 ± 15	9 ± 5	Negative	14 ± 10
Summer	57 ± 49	27 ± 21	34 ± 28	34 ± 26	Negative	Negative
Monsoon	Negative	Negative	29 ± 11	Negative	Negative	Negative
Post-Monsoon	27 ± 18	31 ± 21	9 ± 6	Negative	Negative	Negative

- **Reviewer comment:** rewrite the abstract and conclusion parts. The currents ones are long and poorly organized, so it is hard for readers to capture the major signal. To synthesize and refine the results are needed.

  Authors' response and modifications in the text: We have shortened and restructured the abstract. The revised abstract now reads:

  ABSTRACT

  "Many sites in the densely populated Indo Gangetic Plain (IGP) frequently exceed the national ambient air quality standard (NAAQS) of 100 µg m^{-3} for 24 h average PM_{10} and 60 µg m^{-3} for 24 h average $PM_{2.5}$ mass loadings, exposing residents to hazardous levels of PM throughout the year.

  We quantify the contribution of long range transport to elevated PM levels and the number of exceedance events through a back trajectory climatology analysis of air masses arriving at the IISER Mohali Atmospheric Chemistry facility (30.667 N, 76.729 E; 310 m a.m.s.l.) for the period August 2011–June 2013. Air masses arriving at the receptor site were classified into 6 clusters, which represent synoptic scale air mass transport patterns.

  Long range transport from the west leads to significant enhancements in the average fine and coarse mode PM mass loadings during all seasons. The contribution of long range transport from the west and south west (Source region: Arabia, Thar desert, Middle East and Afghanistan) to coarse mode PM varied between 9 and 57 % of the total $PM_{10-2.5}$ mass.

  Local pollution episodes (wind speed < 1 m s^{-1}) contributed to enhanced $PM_{2.5}$ mass loadings both during winter and summer season and to enhanced coarse mode PM only during winter season.

  South easterly air masses (Source region: Eastern IGP) were associated with significantly lower fine and coarse mode PM mass loadings during all seasons.

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The fraction of days in each season during which the PM mass loadings exceeded the NAAQS was controlled by long range transport to a much lesser degree.

For the local cluster, which represents regional air masses (Source region: NW-IGP), the fraction of days during which the NAAQS of 60 µg m^{-3} for 24 h average $PM_{2.5}$ was exceeded, varied between 22 % and 85 % of the days associated with this synoptic scale transport during monsoon and winter season respectively ; the fraction of days during which the NAAQS of 100 µg m^{-3} for the 24 h average PM_{10} was exceeded, varied between 37 % during monsoon season and 84 % during winter season.

Long range transport was responsible for both, bringing air masses with a significantly lower fraction of exceedance days from the Eastern IGP and air masses with a moderate increase in the fraction of exceedance days from the West (Source region: Arabia, Thar desert, Middle East and Afghanistan).

In order to bring PM mass loadings in compliance with the NAAQS and reduce the number of exceedance days, mitigation of regional combustion sources in the NW-IGP needs to be given highest priority."

We have shortened and restructured the conclusion, the revised conclusion now reads:

CONCLUSION

We investigated the contribution of long range transport and local pollution episodes to the average coarse and fine mode PM mass loadings at our receptor site using two years of high temporal resolution data.The study yielded several results as follows:

1. Long range transport from the west Source region: Arabia, Thar desert, Middle East and Afghanistan)leads to significant enhancements in the average coarse

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mode PM mass loadings during all seasons. The contribution of long range transport from this source region to coarse mode PM varied between 9 % to 57 % of the totals $PM_{10-2.5}$ mass.

2. For fine mode PM the situation is more complex. The fast westerly cluster is associated with a 20 %increase in fine mode PM during summer and post monsoon season but cleaner air masses during winter season. The medium westerly cluster shows moderately enhance PM mass loadings during all seasons while slow westerly transport leads to enhanced $PM_{2.5}$ mass loadings during winter, summer and monsoon season but not during post monsoon season.

3. Local pollution episodes (wind speed $< 1\,m\,s^{-1}$)contributed to enhanced $PM_{2.5}$ mass loadings during both winter and summer season and to enhanced coarse mode PM only during winter season.

4. The south easterly cluster(Source region: Eastern IGP) is associated with significantly lower fine and coarse mode PM mass loadings during all seasons.

5. The number of days during which PM mass loadings exceed the national ambient air quality standard (NAAQS) of 100 $\mu g\,m^{-3}$ for 24 h average PM_{10} and 60 $\mu g\,m^{-3}$ for 24 h average $PM_{2.5}$ (NAAQS, 2009), however is controlled by long range transport to a much lesser degree.

   For the local cluster, which represents regional air masses (Source region: NW-IGP), the fraction of days during which the national ambient air quality standard (NAAQS) of 60 $\mu g\,m^{-3}$ for 24 h average $PM_{2.5}$ was exceeded varied between 22 % and 85 % of the days associated with this synoptic scale transport during monsoon season and winter season respectively; the fraction of days during which the national ambient air quality standard (NAAQS) of 100 $\mu g\,m^{-3}$ for the 24 h average PM_{10} was exceeded, varied between 37 % during monsoon season and 84 % during winter season.

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6. Long range transport was responsible for both bringing air masses with a significantly lower fraction of exceedance days from the Eastern IGP and air masses with a moderate increase in the fraction of exceedance days from the West (Source region: Arabia, Thar desert, Middle East and Afghanistan). The south easterly cluster (Source region: Eastern IGP) is always associated with a significantly lower fraction of exceedance days and the south westerly cluster also leads to a lower fraction of exceedance days during winter and monsoon season.

   Whenever long range transport increases the fraction of exceedance days the increase varies between a few percent and at most 30 %.

7. Fine mode PM ($PM_{2.5}$) contributes most to PM exceedance events at a regional level and $PM_{2.5}$ mass loadings are largely controlled by combustion sources during all seasons. Primary emission and gas to particle conversion of gas phase precursors emitted during the combustion, both contribute to the final mass loadings in varying proportions.

   In order to bring PM mass loadings in compliance with the national ambient air quality standard (NAAQS) and reduce the number of exceedance days, mitigation of regional combustion sources needs to be given highest priority as the number of exceedance days for air masses associated with the source region NW-IGP is already extremely high.

   To devise efficient mitigation strategies targeted at bringing down the number of PM exceedance events, a larger set of tracers needs to be incorporated and alternate source receptor modelling approaches e.g. PMF modelling targeted specifically towards identifying local and regional combustion sources contributing towards the emissions of PM and towards the emission gas phase aerosol precursors need to be adopted.

- **Reviewer comment:** Specific comments. - figure 2. A. What kind of wind do you plot here? Surface, 2m?

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Authors' response: We thank reviewer $\#2$ for pointing out that the height of the inlets and meteorological sensors was missing in our site description. It is 20 m above the ground level (a.g.l).

Modifications in the text: page 11414 after line 17 we inserted the following text. "The inlets of all instruments and the meteorological sensors are co-located and placed at a measurement height of 20 m a.g.l (Sinha et al., 2014)."

In the figure caption of figure 2 we inserted the following text. "Wind speed and wind direction were measured at a height of 20 m a.g.l."

- **Reviewer comment:** B. Have you checked the representativity of this station to NW IGP in terms of wind? In particular during monsoon season, the prevailing wind direction is south easterly instead of south westerly. Do you have any explanation?

  Authors' response: Yes, we have checked that the station is representative of the N.W. IGP for wind flow patterns. The Monsoon system is called the South-West Monsoon, but this name refers to its wind direction over the Indian Ocean and the southern peninsular. The monsoon cyclone enters the NW-IGP via the Bay of Bengal and the Eastern IGP (Bay of Bengal branch of the monsoon). Occasionally the low level jet enters through the Indus valley as the Arabian Sea branch of the monsoon circulation. The winds in the IGP run primarily parallel to the direction of the plains which are delineated by the Himalayan mountain range and the Aravelli Mountains. This applies to most of the IGP. It can be seen for e.g. in Goyal and Sidharth (2002) that even in New Delhi, at a site located more than 200 km South-Southeast from our site, and far away from the Himalayan mountain ranges W-NW and ESE-SE are still the two predominant wind directions during Monsoon season and south westerly winds are not very frequently observed.

- **Reviewer comment:** page 11418, line 21-22. What is GDAS dataset and which analysis database do you use? –

Authors' response: National Oceanic and Atmospheric Administration's **G**lobal **D**ata **A**cquisition **S**ystem (GDAS) is the meteorological data field used for the trajectory calculation, It originates from the GFS/GDAS (formerly FNL) archive and has been accessed via the Air Resource Laboratory (ARL) server. The full form of this abbreviation "National Oceanic and Atmospheric Administration's Global Data Acquisition System meteorology" has been mentioned on the same page line 11-13.

To make the text more clear, we have rephrased page 11418, line 11-13 and 21-22. Modifications in the text: page 11418, line 11-13 we have inserted the abbreviation (GDAS) after the full form and made the relevant first letters bold to facilitate the connection between the two "National Oceanic and Atmospheric Administration's **G**lobal **D**ata **A**cquisition **S**ystem (GDAS) meteorology."

page 11418, line 21 onwards we replaced "dataset" with "meteorological data field" throughout to improve the clarity of the text

- **Reviewer comment:** page 11419, line 4. Only 3 out of 27 are consistent with the measurement. Do you mean all seasons? How do you explain the low percentage and the representativity of the results from this study?

  Authors' response:

  The model terrain height for a given latitude longitude co-ordinate (place) in the input data (GDAS meteorological data field) of the NOAA hysplit model is independent of season. The actual true terrain height of the same place is well known. However, the meteorological data field used as model input has a 1 resolution and the terrain height for the latitude longitude pair entered as the starting coordinate for the trajectory run is interpolated from the closest grid points. This is done by the model itself and the interpolated terrain height does not always agree with the actual terrain height, in particular in the vicinity of mountain ranges. How many ensemble runs will give reasonable terrain heights both for our station in the plains and the mountain site Shimla, located 60 km to the NE of our site, depends on how the terrain is represented in the models input data field. While the modelled air mass origin is strongly affected by how well the mountain range is resolved in the model, we note that this sensitivity of the model does not affect the results of our study. We have selected only those 3 ensemble runs for which the GDAS meteorological data field provide appropriate terrain heights both for our site in the plains and for a mountain site located 60 km to the NE of our site. By doing this we have made sure that the model output we work with is unaffected. To make this clear we have inserted the text below page 11419 in line 7.

Modifications in the text: "To ensure that this sensitivity of the model does not affect the results of our study, we. . .."

- **Reviewer comment:** - page 11421, line 17. In the previous study, you mentioned that the optimum is 6, but you use seven cluster here. Why?

  Authors' response: We did not understand what the reviewer implies by "previous study". Page 11421, line 17 reads the "number of optimum clusters was 6."

- **Reviewer comment:** - page 11422, line 10. Did you mention figure 7 before figure 8?

  Authors' response: Yes, figure 7 is cited first on page 11421 in line 24 i.e. before figure 8.

- **Reviewer comment:** - page 11426, line 23. Should it be figure 9 instead of table3?

  Authors' response We thank reviewer $\#2$ for pointing out that reference in line 23 should be to figure 9. We corrected this and shifted the reference to table 3 to Page 11426 line 25.

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Modifications in the text: Shifted the reference.

- **Reviewer comment:** - page 11426, line 26 and page 11427, line 3. Could you specify what is the aqueous phase oxidation of gas phase precursors?

  Authors' response: Aqueous phase oxidation of gas phase precursors refers to a process wherein a gas (e.g. NH_3(g), SO_2(g), NO_2(g)) is taken up by aqueous phase aerosol and subsequently undergoes reactions that change the oxidation state resulting in the formation of ammonium, sulphate and nitrate ions. Upon drying, these precipitate out as salts which can be coarse in size.

- **Reviewer comment:** Can your study derive it? If not, list reference.

  Authors' response: In figure 10 we provide observational evidence that this process does contribute to coarse mode aerosol whenever the RH is high. We see a good correlation of CO and coarse mode PM at high RH but not at low RH. It is well known, that primary particulate emissions from combustion normally fall into the PM_1 size range and contribute little to PM with an aerodynamic diameter >2.5 μm. The fact that only when RH reaches levels that allow a significant aerosol aqueous phase to exist, a correlation of coarse mode PM with CO appears, indicates, that coarse mode salt formation via such oxidation reactions contributes to coarse mode PM. To make it more clear, that this statement refers to figure 10 we have restructured and modified the text.

  Modifications in the text: Figure 10 shows the correlation of CO with coarse mode PM ($PM_{10-2.5}$) as a function of meteorological conditions. It can be seen from this figure that two sources contribute prominently to coarse mode PM mass loading during winter: aqueous phase oxidation of gas phase precursors and dust. Aqueous phase oxidation of gas phase precursors refers to a process wherein gases (e.g. NH_3(g), SO_2(g), NO_2(g)) are taken up by aqueous phase aerosol and subsequently undergo reactions that change the oxidation state resulting in the formation of ammonium, sulphate and nitrate ions. Upon drying these precipitate out as salts which can be coarse mode in size.

- **Reviewer comment:** page 11436, line 11. Define the exceedance before section 3.4.1

  Modifications in the text: We have modified the first sentence of section 3.4 such that it now includes the definition:

  " The mean PM mass loadings of an air mass cluster represent a poor proxy for the number of exceedance events, that is the number of days on which the 24h average of $PM_{2.5}$ or PM_{10} exceeded the NAAQS of 60µg m^{-3} for 24h average $PM_{2.5}$ or 100µg m^{-3} for the 24h average PM_{10} respectively."

- **Reviewer comment:** – page 11437, line 4-6, page 11438, line 2-4, line 21-22, Give references.

  Authors' response: These statements are based on our own findings; we have revised these statements to make the source of these statements more clear.

  Modifications in the text:

  page 11437, line 4-6: " As discussed in section 3.3.2, during summer season,the ambient RH is usually < 60 % at night and 20-30 % during the day (Table 1) and aqueous phase oxidation contributes less to $PM_{2.5}$ and PM_{10} mass loadings."

  page 11438, line 2-4 we have shifted these lines to the end of the discussion and started the discussion with "Figure 12 shows that the . . ."

  page 11438, line 21-22: "As discussed in section 3.3.4, crop residue burning coupled with aqueous phase oxidation of gas phase precursors is responsible for high PM mass loading during post-monsoon season. It can be seen from figure 12 that it also contributes to a high frequency of exceedance events."

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References

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Sinha, V., Kumar, V., and Sarkar, C.: Chemical composition of pre-monsoon air in the Indo-Gangetic Plain measured using a new air quality facility and PTR-MS: high surface ozone and strong influence of biomass burning, Atmos. Chem. Phys., 14, 5921–5941, doi:10.5194/acp-14-5921-2014, 2014.

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