

Interactive comment on “Aerosol immission maps and trends over Germany with hourly data at four rural background stations from 2009 to 2018” by Jost Heintzenberg et al.

Anonymous Referee #1

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In this paper, hourly measurements of aerosol and gases at four rural sites in Germany during ten years have been analyzed by combining the observations with back trajectories. The goal was to identify potential source regions by connecting back trajectories and emission inventories. The back trajectory approach was chosen because chemical transport models are difficult to use for such long time periods. The paper is certainly of interest to the readers of ACP. However, the paper is in my view lacking de-tailed information about the applied data analytical methods making it very difficult or even impossible to understand and rate the conclusions of this study. In my view, this manuscript requires major revisions before it can be published in ACP (see main comments below).

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Main comments:

The methodical part of the back trajectory analysis is explained in section 3. There is, however, no description how exactly the back trajectories are used and how the maps are calculated. This methodical chapter must be much more detailed (and might also include the most important equations) so that the reader can reconstruct what exactly has been done. This is necessary for understanding the meaning, limitations and the interpretation of the maps as given by the authors later in the paper. Some examples: It remains unclear how the authors treated back trajectories that did not interact with the planetary boundary layer. How exactly were source regions defined? On page 8, line 172 the authors state that "Precipitation along the trajectories was used in the interpretation of the immission maps". How has this been done? Without this information it is not possible for the reader to evaluate the quality of this work and the relevance of the findings. Also for the second approach (Page 11, lines 259/261), there is no information how exactly the emissions were summed up. I expect that emissions were only summed up when the air parcel was close to the surface, but the reader doesn't know. The applied approach must be explained in more detail.

On page 10 the authors write that the calculated immission maps have little in common with the emission data as shown in Fig. 2. Later in lines 239 and 240 it is said that "dilution through mixing with cleaner air masses and wet scavenging through in-cloud and sub-cloud processes" are two major atmospheric processes during transport. This is certainly true, it remains however unclear how such processes are incorporated in the back trajectory analysis. As the paper is now, I have the impression that the calculated immission maps are largely dominated by such atmospheric processes and the interpretation in terms of potential source regions is difficult. The signal from the potential source regions seem to be masked by such processes, e.g. strong dilution of emissions during westerly winds.

For the second approach (connection of emissions and aerosol observations), an optimization algorithm was applied. There is again hardly any information what exactly

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has been done, except that the name of the used solver is given. This should be changed, please explain the applied optimization method in more detail. Maybe provide the relevant equations.

Page 7, lines 139-141: The authors say that equivalent black carbon was measured using a MAAP and converting the measured absorption signal into black carbon by using the default mass absorption cross section of the instrument ($6.6 \text{ m}^2\text{g}^{-1}$). For using the terminology of equivalent black carbon (eBC) it is to my knowledge required that the mass absorption cross section is determined specifically for the site of interest, e.g. from parallel measurements with elemental carbon (EC) as determined using an analytical method such as the thermal optical method (see cited paper by Petzold et al 2013). The authors should therefore not call their measurements equivalent black carbon (eBC) but instead black carbon (BC).

Page 13, lines 298 – 300. The authors write that contributions from wind erosion of agricultural soils are not incorporated in present anthropogenic inventories. What about anthropogenic precursor emissions of secondary PM? How are they included or not included in this study. The authors should provide the corresponding discussion and information.

Page 28, Table 2. It is surprising that the calculated percentage change in PM10 emissions relative to 2009 is highly sensitive to the considered time period (for 2009 – 2017 percentage change is -16%, for 2009 – 2018 percentage change is -6%). This deserves some explanation: Can this be a matter of limited robustness of the applied method, what are the uncertainties of the trend estimations?

Minor comments:

Page 7, line 143 – 144: At three sites TEOM1400 instruments were used for PM10 measurement, at one of the sites daily PM10 was gravimetrically determined. The authors should say a few words about the comparability of the two methods as it is known that there can be (and typically are) systematic differences.

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Page 7, lines 157-158: "Through combustion processes the trace gases NO_x and SO₂ are related to anthropogenic aerosol formation". This is not correct or at least unclear. Of course, NO_x and SO₂ are in Germany mainly emitted from anthropogenic sources and processed in the atmosphere to form aerosols. However, the sentence should be re-phrased to something like "NO_x and SO₂ emitted by anthropogenic combustion processes are transformed in the atmosphere and add to the anthropogenic aerosol".

Page 9, line 204: What is the "Southeastern half of the map"? Please be more accurate. Page 9, line 207: Typo, "PM10and". Page 9, line 212: It is unclear what countries/region is meant here? Rephrase so that this is clear Page 12, line 279: Delete "are". Page 12, line 288: I cannot see an increase in PM10 in 2010, neither in Fig. 4 nor in Fig. 5. Please explain or revise.

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