Atmos. Chem. Phys. Discuss., 12, C536–C541, 2012
www.atmos-chem-phys-discuss.net/12/C536/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trajectory-based source area analysis of atmospheric fine particles, SO_2, NO_x and O_3 for the SMEAR II station in Finland in 1996–2008" *by* M. Hulkkonen et al.

Anonymous Referee #1

Received and published: 13 March 2012

The manuscript presents the trajectory-based source area analysis of atmospheric fine particles, SO2, NOx and O3 for the SMEAR II station in Finland in 1996-2008.

To my opinion, the method introduced in the manuscript is not relevant for those studies I strongly disagree with the statement in lines 63-65. The length of the air mass back trajectory should not not be longer in time than the lifetime of the traced naturally or anthropogenically emitted species. If the emission sources are not local, with the long air mass back trajectories one can study the transport of the long-lived species only, or the meteorological characteristics of the air masses, which might lead to the chemical processes in the atmosphere Two approaches should not be mixed - pollutants source

studies, where you should account for the amount of emissions and the life time of the transported species and air mass source studies, where the air mass properties arrived at the area of interested are studied with respect to air mass quality and processes within. Agree, that the back trajectories is widely-used method to study the air mass origin for different air components measured. The concentration measured at some location may be explained by the air mass history, but the assigning the measured concentration to the back trajectory points is not that easy

Some parts of the paper was not easy to read (to follow the ideas) because of the poor English language.

In Kaiser (2007) the air mass back trajectories are restricted to -24h because of the short life time of NOx. In your paper : line 284: ... lifetime of NOx is typically of the order of 1 day Table 1: lifetime of NOx is 1.5 day My question : why the 4-days back trajectories are used?

Same question should be asked for the other gases.

I would not recommend to use back trajectories for Ozone source area analysis at all. Ozone is not emitted but formed and decayed at certain conditions, which depend on meteorology and air chemical composition. Disagree with the O3 life time you mention – it might be several days in winter and few hours in summer at warm sunny conditions.

There are no formulas in the paper. From the text it is not easy to follow what and how it was done. If the methods you refer to were modified, you should show the equations used.

The concentration is not scaled by the traveling time (decay, deposition), right? – big mistake

Hysplit gives the rain along the trajectory – wet deposition is not accounted for in those studies

"the best correlation R2=0.11" means there is practically no correlation. Means, your

method can't reproduce the main ground emission sources. Even though it was not the aim of the paper, the main emission sources (EMEP) and the main sources for the gases measured should be related. I guess the reason for that low correlation is that the author tries to correlate the results from two different approaches (see my comments to introduction)

Abstract:

How the method was proved useful? Not clear

Materials and methods:

line 87 - used for what?

Normalization (line 107) is not clear for me. Were the grid cell values normalized by highest for certain averaged period (month) or by yearly maximum? Or 13-years maximum? What then RSCF=1 mean? After normalization, there is no intensity anymore, it is not clear anymore how strong the source is.

Line 116 – how big?

Line 127 – again, not clear, are you studying the emission sources (e.g. EMEP) and the air mass transport directions? Or back trajectories and related measured concentrations?

Site-specific features line 154 – instead of degrees better give the direction (south-west) of geographical name of the area Another conclusion can be done from Figure 1 – the long-range transport was happening more often in south-west and north-west directions. Why you show the total figure here if seasonal differences are studied later on? line 157 – what is the "cleansing" effect? If you conclude about the cleaning effect close to the measurement point, why do you study the long-range trajectories? The cleaning of the air masses over the Baltic sea, is that due to the deposition/decay, or this is because of the mixing with the clean (practically no anthropogenic emissions) marine air? You say that "after that (cleaning) the significance of local sources is enhanced"

. What are those local sources? How strong they are? You mention the Tampere population only.

The wind/distance statistics presented here, are they used in the paper for the explanation of the source areas? Median 14 m/s is very strong wind speed. How often those cases happen? Guess, they should be considered as special long-range transport class when the (European) pollution can indeed reach Hyytiälä.

line 169 – what does "median of trajectories" mean? You show the seasonal statistics for air parcel hight. How the air parcel hight relates to the subject you study – source area – besides checking that the trajectory is inside boundary layer hight? Seasonal differences in direction and travel distance are much more important for the current studies, but I do not see the discussion on that topic in the manuscript.

If "Hyytiälä can be considered as a background station" (line 175), would be interesting not to make 13-years monthly averaging, when the peaks in concentrations are smoothed (or, let say in other words, when the "polluted" trajectory is lost among many "clean" trajectories), but to study the air mass transport for the pollution episodes which could be chosen as a periods of elevated concentration measured in Hyytiälä and compare them with the trajectories for clean episodes.

Results and discussions

Have the emission sources along air parcel path (line 197) been studied? If yes, what was the source? As far as I understood, you, together with air mass trajectories combined the concentrations measured at Hyytiälä. EMEP was used only as a comparison to the RCSF, right? Again, no formulas to check

line 212 – how do you see (define?) concentration level from Figure3? Not clear

Line 214 and later on – how can you explain the RSC increase/decrease? Guess, this is because of the year-to-year difference in the frequency of the trajectories from certain directions and year-to-year difference in the concentrations measured at Hyytiälä– not

studied here No clear conclusions done from Table 2. What is the relation between the SO2 measured in Hyytiälä (96-99 vs. 05-08) and measured nucleation mode particle concentration?

Are RSC for nucleation mode particles for 96-00 and 05-08 significantly different?

Explain, why periods 96-99 and 05-08 were chosen.

From your conclusions and Figure 5 I expect that the years with widest yellow-red areas over the North Atlantic (1999, 2002, 2003, 2005) were the years with the highest nucleation mode particles concentration measured in Hyytiälä; highest concentration of accumulation particles should have been observed in 2002, 2003, 2006. Is that true?

Why RSC is that low for 2008? Was the measured particle concentration much lower comparing to other years? That year looks exceptional. It might make the difference in the statistics between 96-99 and 05-08.

Do you make any other conclusions related to nucleation mode particles seasonal/ yearly sources than already done in the papers you refer to?

Line 216 – check the name of the sector B

line 231 – any explanation for that?

I am not going top discuss Section 4.2 here because to my opinion the method introduces can't be applied to the short-lived trace gases

Conclusions

the discussion of the particle formation mechanism should be in the discussion part. In conclusions, the results obtained from current studies should be clearly formulated, if they improve or disagree with the early published material

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My conclusions to the whole manuscript : method should be considerably improved; manuscript should be resubmitted

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 1653, 2012.