**Atmospheric
Chemistry
and Physics
Discussions**

Interactive
Comment

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.
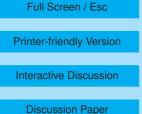
M. Hulkkonen et al.

laura.riuttanen@helsinki.fi

Received and published: 14 September 2012

We thank Referee 1 for his/her critical comments. We see that we have been unsuccessful in explaining both the drawbacks and advantages of the method used.

We have to clarify, that the aim of this paper is not to produce reliable emission fields, but to show what we see at our measurement station in Hyytiälä. We see that air coming from certain sectors is far more polluted than air from other sectors. The features are at some parts different to reported emissions (e.g. best correlation with produced SO2 fields and EMEP emissions is only R2 = 0.11). The reasons might be related either to transport climatology of pollution to the measurement station or chemical sink

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

processes along the transport. We have not taken the dynamical nor chemical processes into account, because it was not the aim of the study. There are far better models already available for that (eg. FLEXPART). Length of the trajectories or characteristic life time of the compounds are of secondary importance in this approach, because we are mostly looking for where the air comes from. Moreover, "life time" is just a mean value and depends strongly on atmospheric conditions.

We want to point out, that the produced Relative Source Contribution Fields do not represent emissions from certain locations, but where the air comes from when we measure high vs. low concentrations in our measurement station. The Referee criticized us from combining air mass studies to measured concentrations. We still think this method is a useful way to see where the concentrations come from, in a statistical sense. High spatial and temporal variations are seen in the produced maps that might not have been visible in other ways. We agree that to produce the actual emission sources, more sophisticated methods, eg. FLEXPART, with relevant removal processes should be used.

Referee #1's main conclusion is that "the method should be considerably improved". He means adding chemistry, effect of sunlight, wet and dry deposition, characteristic life times etc. to the trajectory study. We do not want to start improving the method this way, as our idea was to look only the trajectories and the statistical information that they give regarding air transport. We have tested the method by taking into account HYSPLIT boundary layer height and rain along each trajectory, but it did not improve the method substantially. We did not want to make this method as a chemical model, since we know there are better models for that already existing. Therefore we chose to study only the air mass origins and how they relate to the concentrations measured in Hyytiälä statistically.

Final remarks: We will significantly improve our explanation of the method and interpretation of the results. Also the title will be changed from "source area analysis" to something else (eg. air mass analysis or transport climatology).

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

Specific comments of the Referee #1:

RC: "To my opinion, the method introduced in the manuscript is not relevant for those studies"

We disagree with Referee 1. The method might seem to be inaccurate in case of SO2 where good emission inventories and maps exist, but for other pollutants with not well established sources we think our method gives a meaningful characterization of trace gas and aerosol source areas from Hyytiälä sampling station's point of view. One has to keep in mind that we are not presenting source areas as geographically accurate locations where pollutants are originated, but rather source probability areas in statistical sense. Moreover, not always the result of our analysis is the source area of a given pollutant. Take for example the nano-particle concentration: the highlighted area would represent the area of precursors and clean (low condensation sink) air, the Arctic sea. The actual pollutant, nano-particles, is actually formed during the transportation over Scandinavian boreal forest and is formed uniformly over the big areas with life time of few hours. The source area is not the Scandinavian forest, but rather the Arctic sea. Reasoning is that the same area (Scandinavian boreal forest) would not act as a source for nano-particles when air masses are of continental origin. Similar, but more complex behavior is valid for O3: it has variable precursors, and its sink mechanisms are more complex. In fact so complex that a deterministic trajectory based method has proven to perform worst compared to statistical models (U. Schlink et al. / Atmospheric Environment 37 (2003) 3237–3253).

RC: "I strongly disagree with the statement in lines 63-65."

We agree that we were not able to produce "statistically significant source areas". This needs to be re-formulated, e.g. to "origins of air mass that are statistically relevant for high concentrations".

RC: "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."

We agree with referee that much longer trajectory than life time of the pollutant adds uncertainty to our results, but will not make the results wrong. The method is based on the idea that those map grid cells that get many high concentration "hits" will have an increased importance in the source area maps. A significant area will get more "hits" when a trajectory passes it again but from a slightly different direction. Areas behind the source areas will have smeared concentration hits and will be mixed also with clean trajectories that have gone around the source area. Moreover the life time of a species is not well defined and is only an average value for average atmospheric conditions. Our study covers all seasons, leading to very variable conditions. Most important meteorological variables that define the life time of the pollutants are, along-side precipitation, temperature and radiation, that both have huge differences during the time considered. Temperature difference between time moments is more than 50 oC and also radiation varies greatly from 0 during polar nights to 600 W/m2 . During winter months the Nordic air masses that arrive to Hyytiälä before sun rise have never seen the sun, which rules out all photochemical loss mechanisms for this time periods. Thus, average life time in average conditions has no meaning in this case.

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

We will improve the language.

RC: " 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?"

See explanation above.

RC: "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 de-pend 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."

We agree with referee that, similarly to nano-particles, ozone does not have clear ground sources. The source areas shown are not actual source areas for ozone, but rather its precursors (NOx, VOC) and photochemically favourable conditions. We think Fig. 9 shows nicely that our method works: most of the high O3 concentrations come from the vicinity of Hyytiälä. Further away, the RSC field consists more or less of statistical noise. However, also there differences in O3 production are seen between marine and continental air, with air masses arriving from east having higher probability to result in high O3 concentration in Hyytiälä (as discussed in the paper).

RC: "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 modiïňĄed, you should show the equations used."

We will improve the model explanation.

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

Decrease in reliability of trajectory has been taken in to account, so that the concentration value has been "distributed" to a wider area, leaving lower concentration to a given cell, see line 100-105.

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

We have conducted the studies for SO2 also by taking HYSPLIT rain into account (Hulkkonen 2010). This allowed us to see pollution sources also in the Central Europe. However, taking HYSPLIT rain into account did not change the general feature of high Eastern pollution levels measured at Hyytiälä. It is possible that the reasons why we do not see western pollution in Hyytiälä are climatological (e.g. more wet scavenging

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

in the western air masses). We should write out more clearly that we were not able to produce the actual emission fields, but the climatology of the transported pollutants.

RC: "the best correlation R2=0.11" means there is practically no correlation.

We agree that we were not able to reproduce the actual emission fields. This needs to be stated clearly. We agree, that the maps should be related and differences should be explained. We will improve discussion on that (see explanations above).

RC: "How the method was proved useful? Not clear"

The method is useful from the measurement station point of view. In Hyytiälä case we e.g. see that we do not measure Central European pollution in Finland, but the pollution transported from the East. In the future, the method will be used as part of research on air pollution measured in Hyytiälä, because it provides a statistical view to the origins of air masses related to high/low concentrations.

RC: "line 87 - used for what?"

. . . were included in the analysis.

RC: "Normalization (line 107) is not clear for me."

We will reformulate the method explanation.

RC: "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?"

Here we were trying to set physical boundaries to our statistical model. However, we noticed that there was little effect to the produced RSCFs, which is why the mixing height was not taken into account in the figures of the manuscript.

RC: "Site-speciïňAc 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 ïňAgure 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 signiïňAcance of local sources is enhanced"

We agree with referee that the term "cleansing effect" is not scientific. We will reformulate this and explain better.

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

We will add the major local sources around Hyytiälä station to the manuscript (from EPER database, some largest sources of each of the components). Study of Karvosenoja (2008) shows that 70 point sources contribute of 64% of SO2 emissions in Finland. These point sources certainly contribute to the obtained PSCF's to some extent, but they can not explain the strong eastern contribution to the measured concentrations. The North-Eastern emission sector seen especially in February in Fig. 7 probably includes a contribution from a couple of power plants and paper mills 50-300 km in that direction. Also the point sources of Southern Finland are seen in the Fig. 7. However, there are several strong point sources also in the western coast of Finland that are not visible in these figures. Modelled NOx emissions by Karvosenoja (2008) are in line with our results, with most of NOx produced in Southern Finland.

RC: "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."

These are HYSPLIT calculated wind speeds along trajectory. Note that these are not ground wind speeds.

RC: "line 169 – what does "median of trajectories" mean?"

We will explain this in a better way.

RC: "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?"

Pollution sources of a certain air parcel are either on the ground (boundary layer) or they come via chemical transformation. As majority of the sources are within the boundary layer, most important is to know if the air parcel is inside boundary layer or not. However, taking this into account in the model did not change the results much since high majority of the trajectories stayed below 1000 metres all the studied 96 hours.

RC: "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."

We will add discussion on that.

RC: "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."

We thank the Referee for a good idea.

RC: "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"

Yes, EMEP was only for comparison. Our study only had measured concentrations and trajectories.

RC: "line 212 – how do you see (deïňӐne?) concentration level from Figure3? Not clear"

When the relative sources are clearly stronger in spring months and in September, it is presumable, that also the measured concentrations are relatively higher. This is supported by the observed concentrations. We will improve the formulation.

RC: "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 SO_2 measured in Hyytiälä (96-99 vs. 05-08) and measured nucleation mode particle concentration?"

SO_2 is a precursor for nucleation mode particles but it is not the only factor. The decrease in SO_2 concentration could have effected on number concentration of nucleation mode particles. A clear decrease in SO_2 relative source contribution fields is observed (ca -50% in sectors A-C) and that can have contributed to the slightly smaller decrease in nucleation mode particle RSCFs (-30% to -12% in sectors A-C).

RC: "Are RSC for nucleation mode particles for 96-00 and 05-08 signiïňӐcantly different?"

We will check this.

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

We wanted to choose four years in the beginning and four years in the end of the available measurement period to see if a trend is seen in the relative source contribution fields.

RC: "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?"

Yes, at least partly. Years 2002 and 2003 are the years with highest concentration of nucleation mode particles (Dal Maso et al, 2008) and fraction of nucleation events in Hyytiälä (Dal Maso et al., 2007) and clean Arctic air masses coming from the Northern Atlantic are known to favor these (Sogacheva et al., 2005). Years 2002, 2003 and 2006 experienced high concentrations of forest fire smoke from the South-Eastern sector with high accumulation mode concentrations (Riuttanen et al., 2012). Accumulation mode concentrations were high also in 1998 that is also seen as red areas in the South-Eastern corner in Fig. 5. We will discuss this more in the text.

RC: "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."

We will check this.

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

Seasonal variation in nucleation mode particle source areas in Hyytiälä has been studied by Sogacheva et al, 2005. As far as we know, yearly or monthly variations in the source areas have not been published before.

RC: "Line 216 – check the name of the sector B"

Hyytiälä misspelled, will be corrected.

RC: "line 231 – any explanation for that?"

We will think on that.

RC: "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"

We disagree, see explanations above.

References:

Dal Maso, M., Sogacheva, L., Aalto, P. P., Riipinen, I., Komppula, M., Tunved, P., Korhonen, L., Suur-Uski, V., Hirsikko, A., Kurten, T., Kerminen, V.-M., Lihavainen, H., Viisanen, Y., Hansson, H.-C., and Kulmala, M.: Aerosol size distribution measurements at four Nordic field 20 stations: identification, analysis and trajectory analysis of new particle formation bursts, Tellus B, 59, 350–361, 2007.

Dal Maso, M., Hyvärinen, A., Komppula, M., Tunved, P., Kerminen, V.-M., Lihavainen, H., Viisanen, Y., Hansson, H.-C., and Kulmala, M.: Annual and interannual variation in boreal forest aerosol particle number and volume concentration and their connection to particle formation, 25 Tellus B, 60, 495–508, 2008.

Hulkkonen, M.: Pitoisuuskenttämenetelmän käytettävyys lähdealueanalyysissä: testausta ja arviointia SO2-pitoisuusmittausten ja HYSPLIT 4 -trajektorien avulla, Master's thesis, University of Helsinki, 2010.

Karvosenoja, N.: Emission scenario model for regional air pollution. Monographs of the Boreal Environment Research no. 32, p.55, 2008.

Riuttanen, L., Dal Maso, M., De Leeuw, G., Riipinen, I., Sogacheva, L., Vakkari, V., and Kulmala, M.: Analysis of the effect of biomass burning smoke episodes on atmospheric composition in Finland in 2006, in preparation, Atmos. Chem. Phys. Discuss., 2012.

Sogacheva, L., Dal Maso, M., Kerminen, V.-M., and Kulmala, M.: Probability of nucleation events and aerosol particle concentrations in different air mass types arriving at Hyytiälä, southern Finland, based on back trajectories analysis, Boreal Environ. Res.,

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

10, 479–491, 2005.

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

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper