

WP6: Integration, outreach, and sustainability

Deliverable D6.17: Comprehensive training course on techniques, outreach and integration to improve running a network-4

The training course was aimed at PhD students and advanced young scientists in the field of experimental atmospheric research. The emphasis was on introducing novel methods, including in situ measurements, radars, lidars and satellite instrumentation, and comparing and integrating the results from different approaches using advanced statistical analysis methods. The work done during the training course was organized in small projects, each done in a small group of course participants. There were a total of five groups, combining the topics of ACTRIS WPs 2-6 and 20-22 (Group 1: WP4 & 21; group 2: WP2, 5 & 22; group 3: WP5, 20 & 22; groups 4 & 5: WP3, 6 & 21). The groups collaborated with each other, shared the data and results and came to common conclusions.

The course included three group presentation sessions where each group presented their plans and results. At the end of the course, each group was given the task of writing a scientific report about their results.

The course was given at the Hyttiälä Forestry Field Station in Southern Finland between March 10 and 21, 2014. The course was coordinated by the University of Helsinki.

The lectures covered the following topics:

- Introduction to ACTRIS & the course
- Atmospheric aerosols
- Radar observations
- Aerosol lidars
- Biosphere-atmosphere interactions
- Aerosol-cloud interactions in global models
- EARLINET
- Career and writing in science
- Philosophy of science

The course saw the participation of 30 students of 16 different nationalities, representing 17 institutions in 11 different countries. The course saw the participation of 30 senior staff of 11 different nationalities, representing 8 institutions in 6 different countries.

Course leader :

- Prof. Markku Kulmala (University of Helsinki)

Other professors, senior scientists and assistants:

- Prof. Jaana Bäck (University of Helsinki)
- Prof. Dmitri Moisseev (University of Helsinki)
- Prof. Adolfo Comeron (Universitat Politècnica de Catalunya)
- Prof. Ulrike Lohmann (Swiss Federal Institute of Technology Zurich)
- Prof. Pertti Hari (University of Helsinki)
- Dr. Hanna Manninen (University of Helsinki)
- Dr. Lucia Mona (CNR-IMAA)
- Dr. Oleg Dubovik (Université Lille-1)
- Dr. Mika Komppula (Finnish Meteorological Institute)
- Dr. Nønne Prisle (University of Helsinki)
- Dr. Antti Lauri (University of Helsinki)
- MSc Mikhail Paramonov (University of Helsinki)

- MSc Maija Kajos (University of Helsinki)
- MSc Simon Schallhart (University of Helsinki)
- MSc Juho Aalto (University of Helsinki)
- MSc Susanna Lautaportti (University of Helsinki)
- MSc Roberto Cremonini (University of Helsinki)
- MSc Anna Nikandrova (University of Helsinki)
- MSc Tuija Jokinen (University of Helsinki)
- Dr. Arnaud Praplan (University of Helsinki)
- MSc Nina Sarnela (University of Helsinki)
- MSc Jenni Kontkanen (University of Helsinki)
- Dr. Henri Vuollekoski (University of Helsinki)
- BSc Xuemeng Chen (University of Helsinki)
- MSc Tuomo Nieminen (University of Helsinki)
- Dr Anton Lopatin (B.I. Stepanov Institute of Physics)
- MSc Anu-Maija Sundström (University of Helsinki)
- David Daou (Joint Research Center, European Commission)
- Fabrice Ducos (Universite Lille-1)

Students:

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|---------------------------------|--------------|
| - Mr. Shahid Bhutto | Finland |
| - Mr. Chao Yan | Finland |
| - Mr. Robert Wagner | Finland |
| - Ms. Alisa Krasnova | Estonia |
| - Mr. Antti Manninen | Finland |
| - Mr. Putian Zhou | Finland |
| - Ms. Noora Hyttinen | Finland |
| - Mr. Robert Banks | Spain |
| - Mr. Luis Barreira | Finland |
| - Ms. Stephany Buenrostro Mazon | Finland |
| - Mr. Roberto Cremonini | Finland |
| - Ms. Birthe Marie Steensen | Norway |
| - Mr. Matti Tapio Räsänen | Finland |
| - Mr. Alexander Beck | Switzerland |
| - Mr. Nikolas Papagiannopoulos | Italy |
| - Mr. Faisal Alzawad | Saudi Arabia |
| - Mr. Brandon Hickman | Finland |
| - Mr. Aurelien Chauvigne | France |
| - Mr. Valentyn Bovchaliuk | France |
| - Ms. Alexandra Tsekeri | Greece |
| - Ms. Eleni Marinou | Greece |
| - Mr. Ying Chen | Germany |
| - Ms. Jia Sun | Germany |
| - Mr. Nicolae Sorin Vajaiac | Romania |
| - Mr. Stephane Victori | France |
| - Mr. Mikko Pitkänen | Finland |
| - Mr. Lev Labzovskii | Romania |
| - Mr. Florica Toanca | Romania |
| - Mr. Nikolas Siomos | Greece |
| - Mr. Janne Lampilahti | Finland |

Main conclusions of the group work

Aerosol Group 1 – Mass Spectrometry

Aiming to a better understanding of gas-to-particle conversion in Hyytiälä, the contributions of different gas-phase compounds to the daytime NPF and the nocturnal clustering events were studied, based on 3-year spring time data. Among all the compounds, we were focusing on H_2SO_4 , ammonia, dimethyl amine and ELVOCs in this report, in both ion and neutral forms. Cluster analysis was done with the neutral molecular cluster data and it can be used to group compounds whose chemical composition is not known with the known compounds showing similar diurnal patterns. Known compounds form only 28 % of all different neutral compounds in air during the event days. Also the total concentrations in almost every group contain more than 50 % of unknown compounds in both event and non-event days.

With cluster analysis that includes particle size data we can see how different particle sizes correlate with different compounds. The analysis indicates which compounds are involved in each stage of NPF and also the size-dependent chemical composition of the particles. The smallest particles measured with PSM (0.9 – 1.1 nm) correlate with sulphuric and iodic acid and several ELVOC monomers and organonitrates. This indicates that sulphuric acid is essential in the beginning of NPF these specific compounds play role in the beginning of NPF. Particles of size range 1.1 – 1.5 nm correlate with the radical group and the largest particles (1.5 – 2.1 nm) correlate with the monomer group and ELVOC monomer with m/z 224.

In the daytime, the sulphuric acid molecules are necessary for initial growth while the ELVOCs play a more and more important role in the following growth. Both of them show the similar relation with the growth rate from 1.5 nm to 3 nm. However, from the ion analysis, H_2SO_4 -ions seem to be more correlated with NPF. A threshold of 1 molec/cm³ for H_2SO_4 ion concentration seems to be defined to separate event and non-event daytime cases. Moreover, in some special cases, which are defined as non-event cases but with high concentration of H_2SO_4 -ions, ELVOC ion clusters seem to act as an inhibitor of NPF.

The contributions of NH_3 and DMA to the daytime NPF are studied by measuring the ion clusters $\text{H}_2\text{SO}_4\text{-NH}_3$ and $\text{H}_2\text{SO}_4\text{-DMA}$. Due to their limited atmospheric concentration, in most cases they are not driving the NPF events in Hyytiälä. One strong $\text{H}_2\text{SO}_4\text{-NH}_3$ clustering event occurred in April 10, 2013 and clusters up to $(\text{H}_2\text{SO}_4)_9\text{-(NH}_3)_9\text{-HSO}_4$ were detected. The growth rate of the clusters can be determined by their appearance time, which was about 0.23 nm/h. This value is a quarter of the reported growth rate via neutral pathways, supporting the results from the other instruments that the ion-induced nucleation is less important than neutral nucleation in Hyytiälä.

In previous work (Junninen et al. 2008) small molecular clusters have been observed to form in Hyytiälä during some nights. Our work shows that the ELVOCs should be the major contributors. Firstly, the dominant ions in these events were ELVOC monomers, in contrast to the non-event nights in which the nitrates were the major ions. More convincingly, in comparison between ELVOCs (both monomer and dimer) and particle measurements, their starting times, peak times and ending times were exactly corresponding with each other.

To summarise, for the NPF in daytime, the initial nucleation is possibly driven by H_2SO_4 (both ion and neutral clusters), and both H_2SO_4 and ELVOCs contribute to the further growth. Sometimes ELVOC seems to be able to inhibit NPF, but the reason is still unclear. Neither DMA nor NH_3 plays an important role in NPF in Hyytiälä. For the night time events, ELVOCs were unambiguously the contributor, but the reason why those clusters seldom grow to larger than 3 nm is still mysterious, further study on this issue is needed.

Aerosol Group 2 – Aerosols and Ions

The concentration of primary ions in Hyytiälä (2011 - 2013) is estimated to be around 750 cm⁻³ varying seasonally between 500 and 1250 cm⁻³ with a minimum in spring. The agreement between cluster ion production rates calculated from measured ion concentrations to those calculated from ion source rates due to radiation has substantially improved with introducing the loss rate to the canopy. The difference is now down to a factor of 2 - 3.

The observation that the ion source rate $Q_{<0.8\text{nm} \geq 10.8-1.7\text{nm} \geq 11.7-7\text{nm}}$ could be confirmed with the calculations done in this study. However, ion induced nucleation rate can just be compared to ion production rate in steady state (when no new particles are formed) due to the steady state assumption in ion balance equations. Comparing the values of 4-Apr-2013 in Hyytiälä excluding the new particle formation event there is a factor of 4 - 5 difference. The constant production of 2 nm ions could explain why the observed nucleation rate J_2 is always larger than zero.

The total particle formation rate from NPF days of spring 2011 was found to be higher during the start of the season, the ion induced fraction of the total formation rate also increased during the season. Comparing the particle formation rate to different chemical species in the atmosphere, sulfuric acid showed the highest correlation.

Our analysis suggests that NPF begins simultaneously inside the mixed layer during its vertical growth. There is possibly separate NPF in the free atmosphere. Distribution of newly formed particles is homogeneous vertically but seems to vary horizontally inside the mixed layer. The horizontal variability could manifest as long narrow NPF plumes. Growth rate of all newly formed particles is similar inside the mixed layer. There seems to be less 2.5-3 nm neutral particles at higher altitudes than on the ground during NPF event. Due to this the ion versus total particle ratio was observed to be higher during an event day than during non-event days.

During the spring period of 2011–2013, a preliminary list of 63 nights had nocturnal cluster events, which were further classified into descriptive classes, and compared with NPF daytime events. A second list of all 3 spring months March–May for 2011 to 2013 yielded 111 nocturnal ion cluster events, approximately 40% of data, and used to present basic meteorology and monoterpenes concentrations.

We found a connection between NICE and daytime NPF, with >60% of nocturnal ion cluster events being either preceded or followed by a daytime NPF events. This is clearly something that must be further studied, and investigate a causality, if any, between a nighttime and daytime event, or if they share common environmental conditions. The median GR of nocturnal cluster events was 0.83nm h⁻¹, ~80% lower than the NPF's.

With regards to specific meteorological conditions, we found a clear difference between nocturnal ion cluster events nights and nonevent nights, even if we could not explain the significance of the difference. UVB radiation presented no difference between event and non-event nights; however, daytime temperature, vertical wind component, Obukhov length and sensible heat flux did show discrepancy, which could suggest a more turbulent daytime continental BL, and a stable, near the surface nocturnal BL during NICE. More research is needed to clarify BL dynamics which clearly play a significant role in nighttime events, as they have proven in daytime NPF.

Sulfuric acid was clearly higher on the event-nights, which is in agreement with Lehtipalo et al. (2011) and Junninen et al. (2008), who suggested that although SA concentrations decrease at night, there was increased SA available during nocturnal cluster events compared to nonevents. The concentration behavior of monoterpenes followed that of the ion cluster concentrations, both peaking at around 19:00 hrs, and, additionally, colleagues' (Aerosol Group 1) data have shown an apparent relation between organics and the cluster concentration peaks presented here. Following Lehtipalo et al. (2011) close relation between nano-CNs and alpha-pinene concentrations, a deeper investigation should be done with specific monoterpenes and the appearance of clusters. Furthermore, since both publications of Hyytiälä nocturnal cluster events have shown ion and neutral clusters to both co-occur and be independent of each other, and as our PSM preliminary look at total clusters has shown, it would be most interesting to analyze possible sources (radon, organics) and ambient conditions so as to further understand the mechanisms leading to the various cluster formations, and ultimately the reason why growth is absent or hindered.

Volatile Organic Compounds Group

Long-term measurements are extremely important for the characterization of ecosystems due to their inherent complexity. In this study, the comparison between upscaled emissions from soil and pine and ecosystem flux has shown a clear seasonal and inter-annual variation for all studied compounds.

Pine emissions dominate in the upscaled total emission and contribute over 90%. For the comparison between upscaled total emission and ecosystem flux of SLP method, methanol has the best correlation, but the correlation for isoprene and monoterpenes is lower possibly due to the representative of pine shoot chamber. Upscaled pine shoot emission of methanol showed dependence on photosynthetically active radiation ($R^2=0.52$) and temperature ($R^2=0.48$).

By doing autocorrelation analysis, it was found that ecosystem scale flux had a 3-day cycle. One reason for this could be that the compounds were carried by wind. Other reasons, for instance, human activity effect and other local emissions, need to be studied further.

One reason for the difference between the upscaled total emission and ecosystem scale flux is the fact that the chamber measurements capture more rapid changes in the emissions than can be detected in the ecosystem flux measurements.

The emission potentials of monoterpenes, which are the emission of monoterpenes adjusted by radiation and temperature, were estimated using a nonlinear regression by using a storage pool emission model and hybrid model with an added synthesis term. The models performed equally well and the average contribution of the synthesis term was found to be about 15 percent. In earlier measurements performed with tree saplings, it has been observed that synthesis has a 40 percent contribution to the total VOC emissions (Ghirardo et al. 2010). This means that there is still need for more careful measurements of the VOC emissions especially from older trees.

Our developed method based on vertical gradient measurements allows to calculate NO_x fluxes during calm nighttime conditions. Higher NO_x flux values were accompanied by higher NO_x concentrations at 4.8m height, lower soil temperatures and water content. Because of the sensitivity to the frequency of NO_x accumulation "events" the method was valid for 23% of the studied period measurements.

LiDAR Group

The Planetary Boundary Layer (PBL) height was derived from Kuopio Lidar system. These results were compared with radiosonde data in order to evaluate their accuracy. The lidar results are in good agreement with the radiosonde PBLs. The capability of the lidar system in deriving PBL height, in a site like Kuopio, with low aerosol concentrations and occasional low level clouds is examined with the use of 3 months lidar data. The lidar has 50% to 80% successful PBL derivation, with the larger percentages in summer days, the lower in winter days, and stable percentages at nighttime conditions (approximately 55%).

The new GARRLIC algorithm that uses the synergy from coincident lidar and sunphotometer measurements was tested successfully for various locations and cases with different aerosol loadings. Comparison of the single-wavelength retrievals from two different lidar systems didn't show dependency on the equipment.

Although the EARLINET data based climatology is limited by the number of available measurements, some information over Europe can be obtained. The seasonal PBL cycle seems to vary depending on the location, with Leipzig exhibiting the largest variability of the PBL height depending on season and Potenza showing a relatively flat trend. All of the stations show similar seasonal trends in the seasonal AOD averages of the period 2000-2010 with lower AOD values in winter and the highest during summer. Napoli shows the lowest values of the seasonal Z63 which indicate that most of the aerosol load is contained lower to the ground probably due to Napoli being an industrial area. On the other hand Leipzig shows some of the largest Z63 values, especially in winter possibly due to its continental location which receives numerous synoptic weather systems to help distribute the aerosol load higher in the atmosphere.

The annual AOD values for Potenza and Leipzig both show a decreasing trend for the period 2000-2010 while Napoli shows an increasing trend till 2003 and then a decreasing trend. Thessaloniki annual AOD values show neither an increase or decrease. The annual height Z63 seems to decrease for both Potenza and Napoli indicating an increase in the aerosol load in the PBL over Italy and it seems to increase for Leipzig indicating an increase in the aerosol load in the free troposphere over Germany. Height Z63 also seems to be relatively constant for Thessaloniki. The highest annual Lidar Ratio values in the PBL are observed in Napoli, an industrial region, and the lowest are observed in Potenza, a mountain region. There are indications of a slight decrease in the annual Lidar Ratio in the

PBL for Napoli and Potenza in the period 2000-2010 while Leipzig values seem relatively constant. Furthermore, a slight decrease in annual Angstrom Exponent in the PBL can be observed over Leipzig and Potenza during the period 2000-2010 indicating a decrease in the aerosol size. Taking all into account, there seem to be indications of a slight modification towards less "polluted" and larger particles in the PBL over Italy and Germany during the period 2000-2010.

Cloud Microphysics Group

By combining data from radar and in-situ ground based measurements two specific snowfall cases were examined. The first was suspected to be an aggregation event, and the second case was suspected to be a riming event. With the combination of different instrumentation it was hoped to determine the effects of cloud microphysics and atmospheric dynamics effect of snowflake formation mechanisms. Snowflake formation processes play an important role in precipitation and cloud lifetime and therefore are highly significant in understanding clouds and climate.

Several observations from vertical pointing radar, sounding, wind profiles, micro-radiometer have been analyzed for two snowfall events. Multi-layer clouds and bi-modal Doppler spectra distribution are present in both events, but the riming case shows higher variability of Doppler Spectra in time and height ("snake" shape). The riming occurs in the lowest cloud moist layer. It has not been possible to establish a direct connection between vertical wind shear and riming. Finally, the liquid water path shows clearly higher in case of riming.

Correlations between radar reflectivity, mean weighted diameter and total particle concentration are able to explain variations seen in radar data, along with correlations between vertical velocity on the ground and from the radar we are able to add strength to the radar as a means of identifying riming during snow events.

The in-situ measurements provide a direct measurement for determining whether a riming or aggregation occurred to either support or reject the radar observations. by examining the vertical velocity obtained from the PIP and vertical velocity from the radar we can see that there is a higher velocity obtained during Case 2, which is indicative of riming, while lower velocities indicates aggregation. Snow density further confirms this by showing that the density is larger during Case 2, coinciding with a smaller ratio of liquid water content to snow depth, adding further evidence for riming. We also observed that during the Case 1 aggregation event the clear formation of a bimodal distribution. This appears to be linked to the presence of a liquid water layer about 1 km above the surface. As the snow passes through this layer a new snowflake population is formed, indicating that in the presence of a liquid layer may have an influence on the formation of aggregates.