BACCHUS

Impact of Biogenic versus Anthropogenic emissions on Clouds and Climate: towards a Holistic UnderStanding

Collaborative Project

SEVENTH FRAMEWORK PROGRAMME
ENV.2013.6.1-2

Atmospheric processes, eco-systems and climate change

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Estimated effort contributor(s) (PM):	2.5
Internal reviewer:	Monika Burkert (ETHZ)

Summary of the results:

This deliverable is at the heart of BACCHUS. We devoted the entire project to address the question of the **importance of biogenic vs. anthropogenic emissions on cloud formation and climate** both experimentally and through modelling. Experimentally we divide this assessment into the contribution of biogenic vs. anthropogenic emissions for cloud condensation nuclei (CCN) and ice nucleating particles (INPs). From the modelling side, we conducted an expert meeting on bounding the aerosol radiative forcing as discussed below.

Sources of CCN

In order to address the importance of natural vs. anthropogenic emissions for liquid clouds, we evaluated long-term measurements (> 1 year). In particular, we evaluated the correlation between the concentrations of CCN and four aerosol chemical compounds (organic matter, sulfate, nitrate and black carbon) as a function of supersaturation. All of these compounds have natural and anthropogenic sources, but to varying degrees. Because of the long measurement duration at these sites, these sites are the best for comparison with climate models.

These correlations are shown in Figure 1 for the seven European sites that measure these quantities. They are rather different for the different sites. The variability of CCN at Cabauw is correlated with the variability in ammonium nitrate concentrations, indicating that aerosol growth to CCN sizes is impacted by NOx and ammonia chemistry in strongly anthropogenically influenced European regions. Conversely, the variability of CCN at terrestrial forest sites (Hyytiälä and Melpitz) is triggered by the variability of organic particle concentrations and to a much lesser extent by black carbon, indicating that biogenic sources play a major role in sustaining the concentrations of CCN at high supersaturations in these environments. Finally, it should be noted that at remote mountain sites (Jungfraujoch) all chemical compounds show a similar behavior, as expected for an internally mixed composition. In summary, the results of this analysis, based on long-term measurement records, qualitatively agree with the existing knowledge about the dominant sources of CCN for the above environments. However, the comprehensive CCN and chemical datasets from the European supersites can be used to extract metrics – describing the statistical correlation between CCN and aerosol anthropogenic and biogenic components – which can be used as new constrains for global models, especially with respect to testing parameterization schemes of secondary aerosol formation and growth. Preliminary results from a comparison with a set of 15 global models employing a two-moment description of aerosol properties (size-distribution and composition) indicate that indeed the individual models show very diverse skills in reproducing the observed features of CCN – chemical compounds correlation in the specific environments.

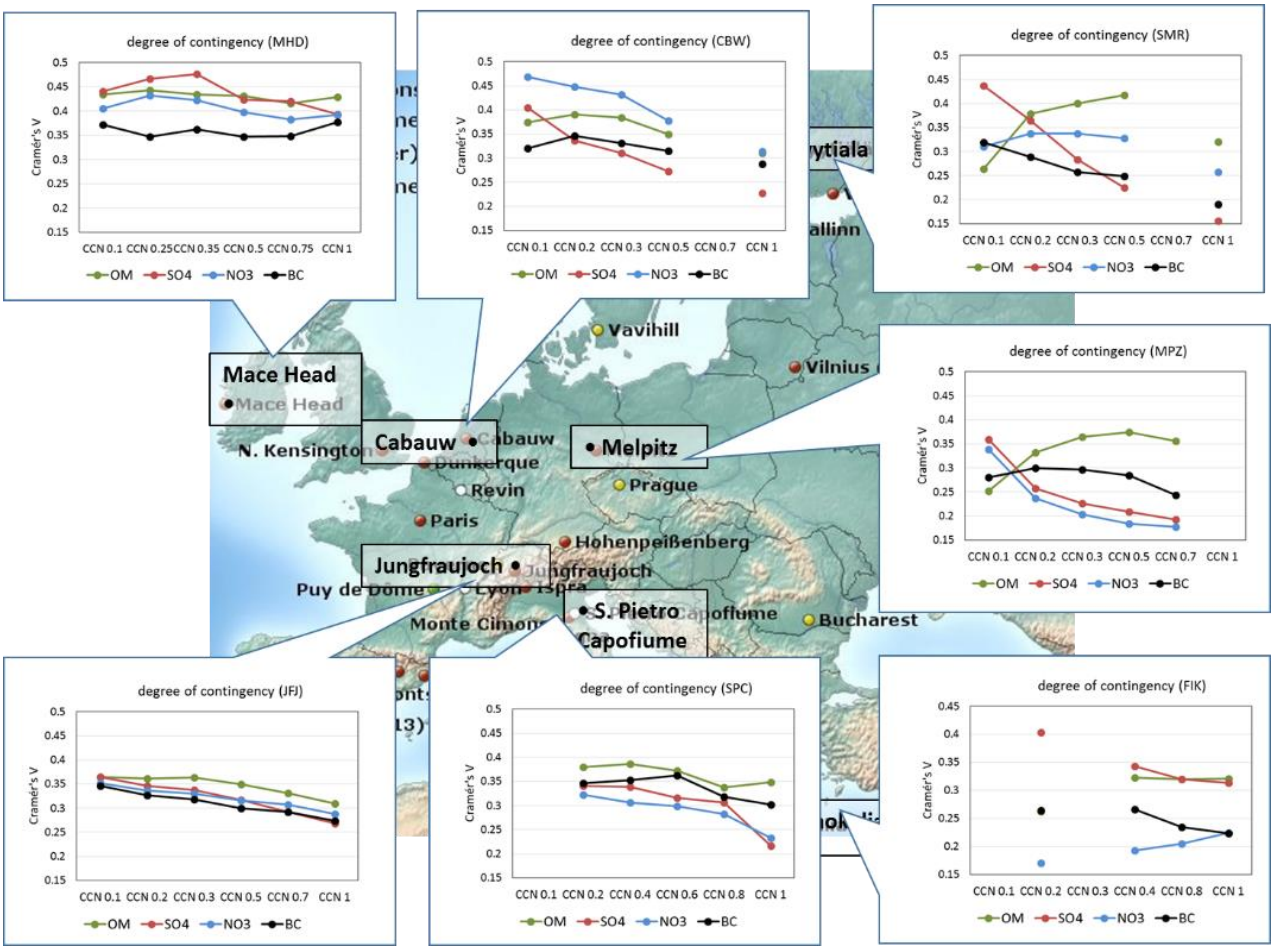

Figure 1: Correlation (expressed by the Cramér's V statistic for 2D frequency distributions), between CCN and AMS chemical compounds (organic matter (OM), sulfate (SO4), nitrate (NO3)) and black carbon (BC) concentrations as a function of supersaturation in % at seven European sites. Figure taken from: Decesari et al., in prep.

If the organic aerosols are further subdivided, then biological volatile organic carbon (BVOC) particles were found to be more important as CCN as previously thought (Gordon et al., 2016, 2017; Arneth et al., 2016).

Accounting for the aerosol formation and growth mechanisms by Extremely Low Volatility Compounds (ELVOC) (Jokinen et al., 2015), projected future increase of BVOC emission in Siberia provide a mean for increased nucleation and aerosol growth rates leading to increased CCN concentrations (Arneth et al., 2016, see also Deliverable 2.3). The importance of BVOCs in new particle formation (NPF) and CCN formation is greater than previously thought since all secondary organic aerosol from BVOC has been calculated to reduce low-cloud-level CCN at 0.2% supersaturation by 26% in the present-day atmosphere and 41% in the preindustrial atmosphere (Figure 2). About 75% of the impact of secondary organic aerosol (SOA) on CCN0.2% is due to the tiny fraction of the oxidation products of BVOCs that have very low volatility and are thus involved in NPF and early growth. Gordon et al. (2017) estimated that ion-induced NPF was more important for CCN0.2% production in the pre-industrial atmosphere (contributing by 40% for NPFs growing to CCN0.2%) than nowadays (27%). This estimate has to be regarded with caution

due to the currently poorly measured ion-induced fraction of NPF involving BVOCs. Because of the simulated production of significant amounts of natural atmospheric aerosol contributing to the baseline pre-industrial aerosol concentrations, these finding could lead to a reduction of 27% in estimates of anthropogenic aerosol radiative forcing (Gordon et al., 2016).
Overall CCN seem to be less sensitive to chemical composition than INP (see below).

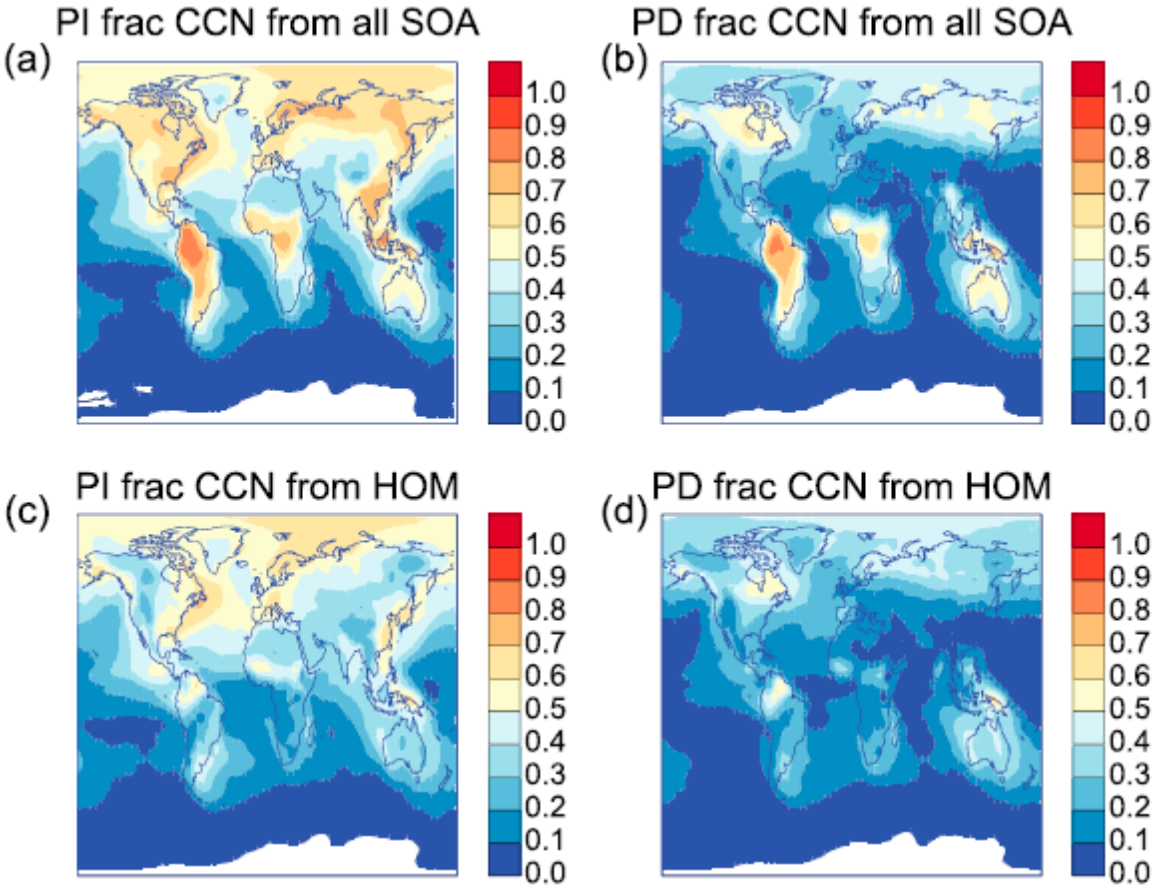

Figure 2: Annually averaged fractions of CCN0.2% concentrations at low cloud level (approximately 460–1100 m altitude) that result from (a, b) all SOA, i.e., all organic vapors that can form or grow particles of all sizes, and (c, d) the most highly oxygenated molecules (HOMs) including BioOxOrg, i.e., all vapors that can form or condense onto particles of less than 3 nm in diameter. The fractions f are approximated by comparing CCN0.2% concentrations in simulations with SOA or HOM formation switched off to the standard model runs, for example, $f_{SOA} = 1 - \frac{[CCN0.2\%(no\ SOA)]}{[CCN0.2\%(with\ SOA)]}$. Figure taken from Gordon et al., 2017.

This dataset is used in a model intercomparison study that was initiated in BACCHUS and has now been opened up also to the global models participating in the AEROCOM intercomparison (Fanourgakis et al., 2018). This study has shown that the spread of models for the cloud droplet number concentration is smaller than the spread for aerosol number concentration and for CCN, because the sensitivities of cloud droplet number concentrations to aerosol number concentrations and to updraft velocity, are negatively correlated.

Composition and abundance of INP

During BACCHUS numerous observational activities regarding INP were conducted in laboratory and field studies, from the Artic to Antarctic region, with shipborne, ground-based, and airborne instrumentation and the buildup of an INP data base (https://www.bacchus-env.eu/in/).

Several well organized, international field campaigns were realized with focus on aerosol chemical (composition), microphysical, and cloud-relevant properties (CCN, INPs), at the west coast of Ireland in mostly very clean marine air and in Cyprus (in every spring of 2015-2017) at highly polluted and dusty air with Middle East/Eastern Mediterranean aerosols.

An example of the INP measurements during the intense field campaign at Mace Head (Ireland) in August 2015 is shown in Figure 3 (McCluskey et al., 2018). These measurements allowed a thorough characterization of the INP population over mid-latitude oceanic regions, in terms of number concentration, nucleation efficiency and size distribution. The obtained results contribute significantly to fill the current gap of INP observations in the remote marine boundary layer.

Average INP number concentrations active at -15 °C were 0.0011 L^{-1} and large variability (up to a factor of 200) was observed for INPs active warmer than -22 °C. Highest INP number concentrations in the clean sector occurred during a period of elevated marine organic aerosol from offshore biological activity (INP number concentration of 0.0077 L^{-1} at -15 °C). A peak in INP number concentration was also observed when the air mass was dominated by terrestrial organic aerosol (with INP number concentrations of 0.0076 L^{-1} at -15 °C). The impacts of heating and hydrogen peroxide digestion on the INP number concentration indicates that INPs at Mace Head were largely organic and that INPs observed during offshore biological activity and in a terrestrial organic aerosol plume were of biological origin (i.e., protein-containing). In addition, pristine sea spray aerosol is a dominant regional source of ice nucleating particles at this remote North Atlantic coastal site. Compared to mineral dust, sea spray aerosols are, however, much less efficient INPs. On average, they have a factor of 1000 fewer ice nucleating sites per surface area. No evidence was found for anthropogenic aerosols to be a good INP. However, anthropogenic activity could change dust sources and with that affect INP concentrations.

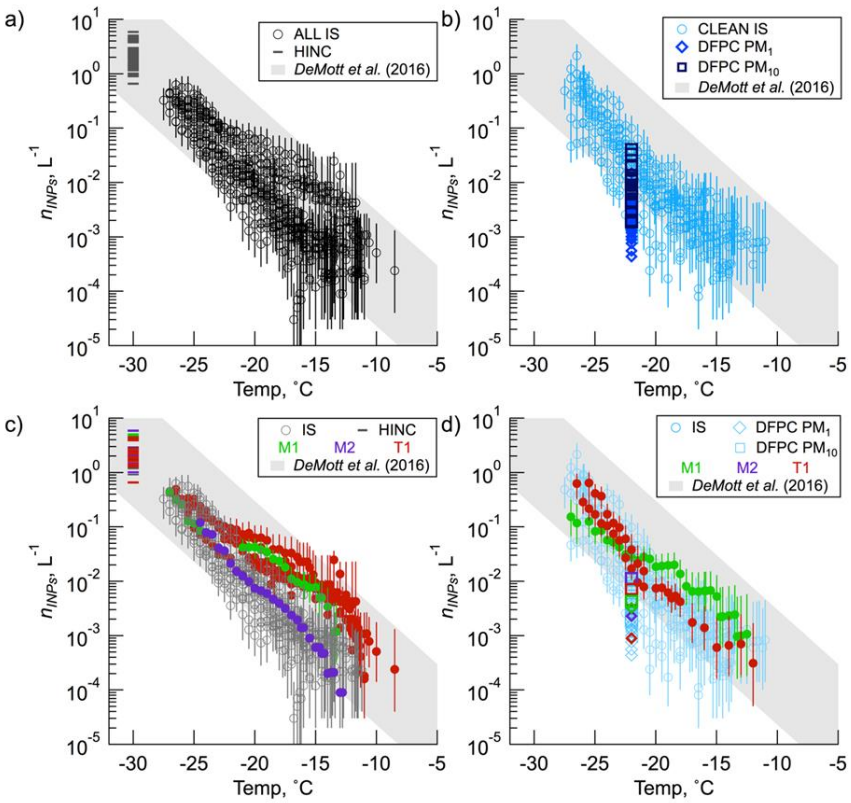

Figure 3: INP temperature spectra for a) ALL samples; b) CLEAN sector samples; c) ALL samples with highlighted events; and d) CLEAN sector samples with highlighted events. Data include those from IS (circles), HINC (dash markers) and DFPC PM1 (diamond markers) and PM10 (squares). Figure taken from McCluskey et al. (2018).

In summary, the results obtained during BACCHUS confirm earlier results that biological particles are the best INPs, initiating ice nucleation already at temperatures larger than -10 °C. Dust particles are almost everywhere (together with haze and smoke) and control heterogeneous ice formation in the temperature range for about -18 to -35 °C. In the absence of dust, INP at a remote North Atlantic coastal site were largely comprised of organic carbon and/or heat labile material from biological activity but sea spray aerosols also contributed. No evidence was found for INP from anthropogenic activity to be an important source for INP. In summary, anthropogenic activities mainly affect CCN concentrations while they only indirectly affect INP concentrations.

Anthropogenic aerosol radiative forcing

A more specific question related to just the anthropogenic aerosol radiative forcing since pre-industrial times was discussed during a one-week expert meeting in Schloss Ringberg, Germany that took place from February 26th to March 2nd, 2018 to which 36 experts were invited. The aim of this meeting was to discuss unlikely strong and weak anthropogenic aerosol radiative forcings (aerosol forcing for short) by providing lines of evidence for why the aerosol forcing cannot be more negative or more positive than certain bounds. The bounds are meant in terms of likelihood rather than certainty. The Ringberg meeting was organized by Nicolas Bellouin, Sandrine Bony,

Olivier Boucher, Jean-Louis Dufresne, Piers Forster, Jim Haywood, Ulrike Lohmann, Gunnar Myhre, Johannes Quaas, Bjorn Stevens, and Philip Stier, i.e. by 3 BACCHUS Principal Investigators. Stephanie Fiedler, David Neubauer, Stefan Kinne and Ken Carslaw from the BACCHUS consortium attended the meeting as participants. The program is in the appendix.

During this week structured discussions were held with the aim to bound global aerosol radiative forcing. Each aspect was first introduced in half an hour, followed by 1.5h discussions in break-out groups and a final plenary. The break-out groups were randomly assigned in order to mix the participants and to have an active participation also from the early career scientists. This concept worked extremely well. Interestingly enough often the key messages were similar in the different break-out groups, which by itself showed the consensus amongst all workshop participants.

The following lines of evidence were considered and supported by the BACCHUS activities: process-based (bottom-up) approaches from large-eddy simulations, field observations, ship tracks, volcanic tracks, global climate model (GCM) simulations, satellite statistics and contrasted them with top-down approaches involving energy balance models, Earth system models of intermediate complexity, GCM ensembles, emerging constraints and observed trends in relevant variables.

To narrow the uncertainty range in aerosol forcing, the first step was to bound the perturbation strength, using aerosol optical depth (AOD) as a proxy. The total AOD was assessed to be between 0.12 and 0.16. The anthropogenic contribution was assessed as roughly 30-40% of the total AOD. A previous community assessment (Bond et al., 2013) consolidated a low bias of global model simulated AOD in comparison to near-source measurements through emission scaling. However, this approach was not supported in this expert assessment as it ignores indications that global models tend to overestimate black carbon lifetimes (e.g. Kipling et al., 2013; Samset et al., 2014). This led to a downward revision of the importance of the positive direct radiative forcing by black carbon. It amounts to roughly 10% of the anthropogenic AOD.

The effective radiative forcing due to aerosol-radiation interactions (ERFari) was assessed to be between -0.2 and -0.5 W m^{-2}. Adjustments were assessed to be negligible in the global mean so that ERFari is roughly the same as RFari. ERFari in the clear-sky is roughly twice as large because clouds mask that forcing to approximately 50%.

The discussion on RFaci (radiative forcing due to aerosol-cloud interactions) was split into aerosol-cloud interactions associated with liquid clouds and aerosol-cloud interactions associated with mixed-phase and ice clouds. This distinction was done because we have a better scientific understanding of RFaci on liquid clouds than on mixed-phase and ice clouds. RFaci in liquid clouds was bound to be between -0.3 and -1 W m^{-2} to -1.3 W m^{-2} (the lower bound was not further refined during the meeting). RFaci for clouds containing ice and ERFaci were not bounded because of too many unknowns, but can be found in the BACCHUS publication by Lohmann (2017).

With that, the expert assessment for the final bound for the all-sky ERFaci+ari concluded it to lie in the range of -0.5 and -1.5 W m^{-2}. To have either a more positive or more negative ERFaci+ari

would require strong adjustments for which we find no evidence. The uncertainty range in ERFaci+ari from the Ringberg meeting is reduced by 50% as compared to the expert solicitation by Granger Morgan et al. (2006) but wider than can be found in some recent literature. The likelihood that ERFaci+ari falls within one of the four 0.5 W m^{-2} ranges, as assessed at the end of the Ringberg meeting and is shown in Figure 4. The results of this expert assessment are still preliminary and may need to be adjusted. A paper entitled "Bounding aerosol radiative forcing" by Bellouin et al., (2018) is in preparation.

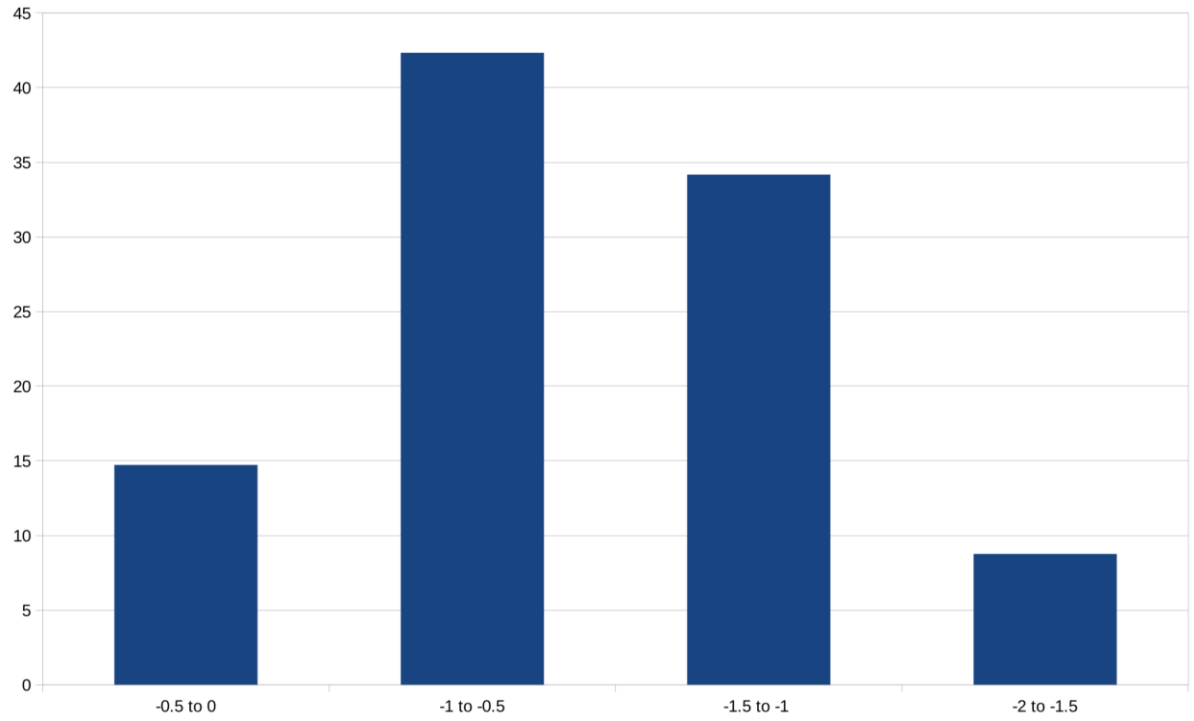

Figure 4: Likelihood in % that the aerosol forcing in W m^{-2} falls in one of the four 0.5 W m^{-2} ranges as assessed from the 36 participants at the Ringberg meeting.

Changes with respect to the DoW

No changes with respect to the DoW.

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Appendix

Programme of the Ringberg workshop

Ringberg meeting "Bounding the aerosol effective radiative forcing"

26 February – 2 March 2018, Schloss Ringberg, Kreuth, Germany

Goal:
The aim[1] is to exclude unlikely strong and weak aerosol forcings (e.g. provide arguments why the aerosol forcing cannot be more negative than -1.5 Wm^{-2}, or why it cannot be positive). We aim for likelihood rather than certainty.
Each participant is invited to submit a 1-page list of theses about likely/unlikely aerosol to the group (ringberg@lists.uni-leipzig.de) before the workshop.

Concept:
One *presenter* per session
- prepares a limited number of theses that quantify or constrain forcings/mechanisms
- all participants are invited to submit theses to the presentations
- theses can (should) be corroborated briefly by explanations/graphics/references
- presenters distribute the theses until late January so participants can prepare for the discussions

At the meeting, one *rapporteur* per session
- moderates the plenum discussions and
- takes notes for the wrap-up discussions on Friday

A large part of the discussions will be in *breakout groups*:
- three breakout groups with 10-12 participants each;
- distribution will be randomly selected and change each time, and
- a rapporteur per breakout group per session will also be randomly appointed

Location:
Ringberg Castle (www.schloss-ringberg.de). Participants are expected to cover their own expenses (travel to Munich + 2 hr train from airport / 1 hr from railway station plus 126 € / night including all meals) and be in residence for the duration of the workshop. If financial restrictions might prevent you from coming please let us know and we will look for ways to help offset some or all of the costs.

Preliminary Programme

Monday 26 February 2018

13.00 h	Lunch
14.00 h	Welcome and goal of the workshop
	Session 1: Possibilities for strong and weak ERFaer
14.30 h	Presentation of theses (*Forster* / rapporteur: *Stevens*)
15.00 h	Discussions
16.00 h	Coffee
16.30 h	Breakout group discussions
18.00 h	Plenum: reports from breakout groups
19.00 h	Dinner

1 We have some funding from the German Research Foundation for the Workshop, and the explanation we provided for them is slightly more exhaustive in case you are interested: http://tinyurl.com/ringberg

Tuesday 27 February 2018

Session 2 : Radiative forcing by aerosol-radiation interactions (direct effect)

9.00 h Presentation of theses: anthropogenic aerosol perturbation (*Schulz* / *Bellouin*)
9.30 h Plenum discussions
10.30 h Coffee
11.00 h Breakout group discussions
12.30 h Plenum: reports from breakout groups
13.00 h Lunch
14.30 h Presentation of theses: radiative efficiency and cloud masking (*Kinne* / *Stier*)
15.00 h Plenum discussions
15.30 h Coffee
16.00 h Breakout group discussions
17.30 h Plenum: reports from breakout groups
19.00 h Dinner

Wednesday 28 February 2018

Session 3: Radiative forcing by aerosol-cloud interactions (Twomey effect)

9.00 h Presentation of theses (*Gettelman* / *Quaas*)
9.30 h Breakout group discussions
11.00 h Coffee
11.30 h Plenum: Report from breakout groups, and discussion: Total radiative forcing

Session 4: Adjustments (effective forcing)[2]

12.30 h Presentation of theses: Liquid cloud fraction, liquid water path (*Gryspeerdt* / *Haywood*)
13.00 h Lunch
14.30 h Presentation of theses: Ice clouds "and other wild cards" (*Storelvmo* / *Lohmann*)
15.00 h Coffee
15.30 h Breakout group discussions
17.00 h Plenum: reports from breakout groups (both afternoon), discussion on cloud adjustments
18.00 h Guided tour of the castle
19.00 h Dinner (Bavarian evening)

Thursday 1 March 2018

Session 5: Implications for climate sensitivity and top-down constraints

9.00 h Presentation of theses (*Forest* / *Myhre*)
9.30 h Discussion in plenum
10.30 h Coffee
11.00 h Breakout group discussions
12.30 h Plenum: reports from breakout groups
13.00 h Lunch
15.00 h Hike
19.00 h Dinner

Friday 2 March 2018

Session 6: Conclusions

9.00 h Session summaries by session rapporteurs
10.00 h Discussions
11.00 h Coffee
11.30 h Final discussions

13.00 h Lunch
14.30 h End of meeting

2 These sessions intentionally have a little less time since the workshop intends to give more time to what is better known than to what is unknown.

Participants (invitation only):

1 Nicolas Bellouin, University of Reading
2 Olivier Boucher, Institut Pierre Simon Laplace/CNRS, Paris
3 Ken Carslaw, University of Leeds
4 Matt Christensen, RAL Oxford
5 Anne-Laure Daniau, Université de Bordeaux
6 Jean-Louis Dufresne, Laboratoire de Météorologie Dynamique/CNRS, Paris
7 Graham Feingold, NOAA Boulder
8 Stephanie Fiedler, Max Planck Institute for Meteorology, Hamburg
9 Chris Forest, Penn State University
10 Piers Forster, University of Leeds
11 Andrew Gettelman, NCAR Boulder
12 Edward Gryspeerdt, Imperial College London
13 Jim Haywood, UK Met Office, Exeter
14 Norman Loeb, NASA Langley
15 Ulrike Lohmann, ETH Zürich
16 Stefan Kinne, Max Planck Institute for Meteorology, Hamburg
17 Florent Malavelle, University of Exeter
18 Thorsten Mauritsen, Max Planck Institute for Meteorology, Hamburg
19 Daniel McCoy, University of Leeds
20 Johannes Mülmenstädt, University of Leipzig
21 Gunnar Myhre, CICERO Oslo
22 David Neubauer, ETH Zürich
23 Anna Possner, Carnegie Institution for Science, Stanford
24 Johannes Quaas, University of Leipzig
25 Maria Rugenstein, ETH Zürich
26 Yousuke Sato, University of Tokyo
27 Michael Schulz, met.no Oslo
28 Steven Schwartz, Brookhaven National Laboratory
29 Brian Soden, University of Miami
30 Odran Sourdeval, University of Leipzig
31 Bjorn Stevens, Max Planck Institute for Meteorology, Hamburg
32 Philip Stier, University of Oxford
33 Trude Storelvmo, Yale University
34 Velle Toll, University of Reading
35 Duncan Watson-Parris, University of Oxford
36 Dave Winker, NASA Langley