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********** 3rd CATCH Open Science Workshop 9-13 May 2022 **********

Day 4 - 12 May 2022: Cryospheric links to aerosol-cloud interactions (Paul Zieger & Jessie Creamean): What are the sources and abundances of cloud forming aerosols in the polar regions and how do these vary over the seasons? What processes control aerosol emission and transport? What are the current modeling deficiencies?

Please use this etherpad "chat" to leave your general comments and specific questions to speakers. This way we'll keep a record of your contributions and discussion across all three session of today. Please add you name to your comment. Thanks!

Session 1 - 0800-1000 UTC+2

08:10-08:40 Ben Murrary: Arctic ice-nucleating particles and climate

Paul Zieger (SU): Is the hole-punch-cloud caused by addition of INP or by pressure changes?

Luisa Ickes (Chalmers): I think the concept/idea is that it is entrained ice crystals from above. Bur more important here (despite if it an INP or ice crystall triggering this) is that you can see the Wegener-Bergeron Findeisen process there.

Concerning the Southern hemisphere: Does the source/nature of INP matter in the model runs (marine source?)?

Ben: I'm not entirley sure, but I suspect it is ice generated by low pressure and may also be related to mixing air with INP into the cloud. I'd be suprised if it were aerosol produced by the engines (soot is thought to be very poor at nucleating ice, and these soot particles are very small).

Louis Marelle (CNRS-LATMOS): Thank you for the very interesting talk. It would be really useful for modelers if all the INP measurements at the poles could be made available in an aggregate, easy to use dataset with a common format. We can always go back to the original publications but it's

a shame if every group has to replicate the work of obtaining the data, understanding the different file formats and writing the code to read them. Is there already such a dataset? Like the one behind Figure 6 of your 2021 ACP paper that you showed several times in your talk, or Figure 2B of Porter et al. 2022?
Ben: I agree. Alberto Sanchez-Marroquin started working on this, but unfortunately he ended up leaving for a job elsewhere. I can make the data behind those plots available to people.
Paul: The data from Porter et al can be found here: https://archive.researchdata.leeds.ac.uk/932/
Ben: this is only the north pole data, not the entire literature dataset.
Luisa Ickes (Chalmers): There is the initiative to collect and have all INP data collected in one database (BACCHUS database) and we are actually working on finishing this for all data (also historic) until the year 2020 (so Graces publication will unfortunately not be part of it).
Louis: Thank you all for the answers, this is great!

KFossum (NUIG): Great talk, thank you. Is there any information on the physicochemistry of the INP in the Arctic measurements? I.e. what species and sizes are contributing to the high INP concentrations.
Ben: This is something I didn't get time to talk about :) There are a range of types. Some of this material is from the mid and low latitudes, desert dust, bio. But, there are also local sources: high latitude dust (that sometimes has biological material mixed with it), sea spray.
The size of INP in the Arctic also vary a lot. Our SHARK measurements suggest that on many days (not all) that the particles less than 0.25 um were most important. But, in other locations the coarse mode has been highlighted.
KFossum: Thank you, yes sounds like huge variation with the likely suspects again contributing. Any suprises? :)

Thorsten Bartels-Rausch (PSI). Very nice talk, Ben. Thanks. Would you know about studies/information on the location/fate of the INP in the ice clouds, in particluar if those stettle and turn to surface snow (at some moment in time). Are these always in the centre, or could ice from also in a way that the INP end up at th surface of the ice particle?
Ben: Luisa might be able to better answer this. Sometimes the ice crystals will end up on the surface, sometimes they will sublime and release the INP. Generally, I think we'd exect the INP to end up within the ice crystals.

Luisa Ickes (Chalmers): I think that really depends on the freezing mode you are looking at, but in case of immersion freezing (which is dominant) I would expect that the aerosol particle is rather in the middle of the ice crystal.
As Ben said it is very difficult to asses the sources and it does not become easier using models because you still have to decide at the beginning which aerosol types you want to presribe (and how they freeze), but using global/regional modells you can get some idea of aerosol transport depending on the source and the impact on the cloud/freezing.
Thanks Luisa and Ben!

Julia Asplund (SU): Thanks Ben! Jessie Creamean has published some work suggesting that thawing permafrost is a significant source of INP, is this something you have considered to explain the high concentrations coming from the russian coast?
Ben: yes, we folded Jessie's paper into our discussion on this in the Porter paper :) : https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2021JD036059

Markus Frey (BAS): Great talk! What are the limiting factors for INP from Arctic surface sources (e.g. sea ice, open water) to reach a level where they can impact clouds? You mentioned verical mixing, are INP small enough or fall back to the surface quickly? what is known about their size range in the Arctic? could potentially small ice crystals, which should be good nucleators, from blowing snow play a role?
Luisa Ickes (Chalmers): Markus one comment to your question: seeding ice crystals (ice crystals falling from above) for example can play quite a big role - we can see a lot of multilayer clouds in the Arctic (see for example https://acp.copernicus.org/articles/19/5111/2019/). And yes blowing snow could be important if lifted up (in the surface mixed layer).

Marc Mallet: Following from Markus' question and the discussion in zoom, here is the link to Griesche et al: https://doi.org/10.5194/acp-21-10357-2021. "Contrasting ice formation in Arctic clouds: surface-coupled vs surface-decoupled clouds".
Luisa Ickes (Chalmers): Great, thanks!

08:40-08:55 Luisa Ickes: Challenges of modelling aerosol-cloud interactions

Paul Zieger (SU): Thanks for your nice talk! A kappa of 0.2 is not so low (similar to organic aerosol you would expect over boreal regions) and as such I wonder if you could have also used much lower kappa's (close or equal to 0)? What would change?
Luisa Ickes (Chalmers): Paul, thanks to bring up that point! We actually did simulate the case also for a kappa value of 0.002 (which was the lower limit at the beginning, but then we did change because it might not be realistic for the Arctic case). In that case for the bigger aerosols the curve looked quite similar (it does a bit of a jump to even lower LWP compared to kappa=0.2), for smaller aerosols the cloud was still! sustained and the LWP a bit lower compared to the kappa=0.2 simulation but not a huge difference.

Marc Mallet: Thanks very much for the talk! Did you test the sensitivity of the cloud droplet number concentration on the shape of the aerosol size distribution (sorry if I missed it!)? I suppose where the activation diameter is in relation to the peak/edges of the Aitken or accumulation mode could make a large difference? I see this to an extent with Antarctic coastal aerosol.
Luisa Ickes (Chalmers): Good point! As I said before modelling aerosols is often very "idealized". In our case we only use the median and width of distribution of the size distribution and assume it is log-normal. We did some tests changing the width of the distribution (and the median obviously) but not really the shape (which would be really tricky to change). But assuming the activation diameter is not really the median of the distribution is kind of caputured by the simulations where the median is shifted (so comparison of bigger/smaller aerosols might give a hint what is happening). We were quite surprised that the cloud case with small aerosols is still so stable (however we start with a cloud and not from zero). I think it would be interesting to do more tests in this direction! And testing cases where more than one aerosol distribution is present would also be interesting.
KFossum: We have tested this in simple cloud parcel models (liguid regime). It is complicated by the chemistry but makes a large difference in relative change in albedo depending on updraft velocity. Can show you general results (if anyone is interested) treating only NaCl and nssSO4 species and altering nssSO4 aitken mode median diameter - kirstennicole.fossum@nuigalway.ie

08:55-09:10 Yutaka Tobo: Insights from a year-round record of ice_

nucleating particles over Svalbard during MOSAiC

Joel Savarino: Mixing the aerosols in solution may change the individual properties of aerosols and thus their ice nucleation properties. How do you deal with this possible bias? By the way very nice data set and clever approach.

Yutaka: I think the impact of soluble coatings on aerosols would be relatively small, because we have measured INPs active in mixed-phase clouds. This means that INPs would be actived as CCN in mixed-phase clouds before ice nucleation (immersion freezing).

Paul Zieger (SU): Thanks for your nice talk! How did you define the seasons? The coarse mode fraction is mainly determined by the sea spray contribution (wind speed dependent). The summer usually is mainly influenced by the fine and not the coarse mode.

Yutaka: I simply define the seasons by months. As you point out, the aerosol pupulation would be largely influenced by new particle formation by number. On the other hand, the aerosol population would be characterized by coarse particles when we saw the data by volume or mass. The coase mode fraction would be mainly determined by sea spray aerosols, and this is probably the reason why INPs at Zeppelin did not show clear correlation with coase mode aerosols (we will need to see specific aerosol types like dust, biogenic aerosols, etc.)

Ben Murray: These long term datasets are important! Sorry if I missed this, but did you do a heat test on these samples? Also, I was trying to see where your measurements fell relative to those of Wex and others. Do you have a literature comparison plot?

Yutaka: We have not conducted heat treatment because we need to analyze a lot of samples for this kind of long-term monitoring. I have net yet compared the Zeppelin INP data with Wex's data and we will probably need to do it. But I think the values would be similar.

Julia Asplund: Thank you! Did you look at air mass trajectories as well? Could the increased INP concentrations in summer for temperatures above 0 degrees be associated with warm air intrusions and therefore longe range transport, or are they definitely dominated by local sources?

Yutaka: Yes, I did backward trajectory analysis. The results sugget that although air masses were occasionally influenced by long-range tranport from Europe/Siberia, those were mostly influenced by local air masses in summer. Given the measured high INP concentrations throughout the summer,

I assume that local sources would be more important that long-range transport in summer, at least.

09:10-09:25 Max Maahn: Clouds in Northern Alaska demonstrate sensitivity to local industrial aerosol emissions

Paul Zieger (SU): Thanks for your nice talk! I might habe missed it: When you compare your entire MODIS dataset with respect to effective radius, did you put a constrain on the cloud top pressure (filtering out high level clouds)? Is the ShupeTurner classification similar to the Cloudnet target classification?

Max Maahn (Leipzig): 1) The figures I showed are not filtered for cloud top pressure, but I show them in the paper, limiting the data to low clouds enhances the signal slightly 2) Yes, ShupeTurner is very similar to Cloudnet

09:25-10:00 Questions and Discussions

Session 2 - 1200-1400 UTC+2

12:10 - 12:40 Gillian Young McCusker: Introducing QuIESCENT Arctic

Paul Zieger (SU): In the CMIP6-satellite comparison: How much bias comes from the satellite data (which are probably also difficult/uncertain over ice/snow)? Is there an estimate?

Gillian McCusker: Most of the very high latitudes were chopped off of this comparison for this reason, but yes there is still sea ice in some of those areas highlighted. I would need to double check if the paper estimates an uncertainty, but I believe they did impose some corrections to help minimise any artificial effects from clouds-over-ice.

Ben Murray: regarding this bias to too many Arctic clouds in the Arctic in CMIP6, is this a result of the changes that have been made to correct the biases in CMIP5 in the southern ocean where there was not enough liquid water cloud?

Gillian McCusker: yeah could be, there have been a lot of developments along the ACI lines (e.g., actually using CCN activation rather than a fixed droplet number) so they have likely affected our modelling capability. The reduction in ice/enhancement of cloud liquid in Southern Ocean clouds is likely having a similar impact on the clouds of the

northern latitudes, but I think there are other factors at play in the Arctic clouds which are missing in the models. (in my opinion!) the Arctic clouds look a lot more like SO clouds now, but they shouldn't necessarily because there is a more complex vertical structure (the aerosol/cloud/BL structure I mean) in the Arctic which is not reproduced well in the large-scale models. There are a lot of outstanding problems with meteorology, so the UM/IFS comparison I showed has large temperature and moisture biases which would affect when given (e.g., primary ice) parameterizations kick in in the model.

Dominic Heslin-Rees: Models in general underestimate the melting of Arctic sea-ice in terms out future projections of an ice free Arctic, but seem to overestimate low-level clouds, leading to increased radiative forcing - could you explain?
Gillian McCusker: My understanding was that there is still a lot of spread in the model predictions of sea ice decline? Perhaps I missed something there. There are a lot of differences between the different global models, both in terms of performance more generally and how they represent certain processes, so though the majority appear to overestimate low-cloud fraction, not all do. I think this is the danger in using multi-model means - this really needs to be looked at on a model-by-model basis to see how each model individually performs in terms of cloud cover, the surface energy balance, and sea ice decline.
The general figure you showed with the cloud cover from CMIP5 models - the observations are out of the bounds of the model uncertainty (blue shaded region) when freeze-up and melting occur/start i.e. September and March, coincidence?
Gillian McCusker: Sorry I misread your question! No I don't think that's a coincidence, there's a lot of processes going on in those time periods (both in terms of changing CCN/INP sources linked with the changing sea ice and dynamical/meteorological changes) which are not captured well by these models and will likely impact their ability to generate and sustain clouds during the spring/fall (I think that's what you meant?)

12:40 - 12:55 Kirsten Fossum: CCN over the Southern Ocean in the austral summer of 2015

Paul Zieger (SU): Very nice talk, Kirsten! The cloud parcel modelling study was very interesting and it seems that it compares well to our recent work from the Arctic. Could you post the link to your paper here again? How much did the Hoppel minima change for the air masses (in

general)?
KFossum (NUIG): https://www.nature.com/articles/s41612-020-0116-2
Thank you, the Hoppel minima ranged from 54-65 nm (dry electrical mobillity D) for cAA to 64-82 nm for mP, and 79-83 nm for mT cases. median cAA hoppel minima was 56 nm, and 82 nm for mP.
Thanks, I will look at your paper. Did you include the measured coarse-mode in your simulation? I assume they are the ones that make the difference, right?
No directly measured coarse mode. The APS failed at the start of the campaign. So we mostly modelled sea-salt and nssSO4 with a theoretically scaled jet-drop mode (2um D) based on the literature for sea-salt, and that did make a difference, however, we did see difference in sensitivity when changing CMD of nssSO4 modes and holding sea-salt modes (6 in total) constant.

12:55 - 13:10 Katye Altieri: Impact of sea ice and snow on aerosol nitrate in the Southern Ocean boundary layer

Santiago: Katye, I wonder in those transects SA-Antarctica, has anybody measured volcanic aerosols downwind from the S. Sandwich Islands? There is weekly activity in those volcanoes so one would expect a contribution to the background atmosphere.

Katye Altieri: We sometimes see high nss-sulfate concentrations, which we attribute to DMS emissions usually but could be more careful and evaluate potential volcanic inputs. I'd have to see what other tracers would indicate that and will make a note to do it. Thanks for the suggestion!
cool, let me know . I monitor those volcanoes so I have a pretty good idea when they were most active and when they were not.
Brilliant, I may be in touch to discuss.

Joel Savarino - Very nice work and dataset, complete very nicely what we did with Samuel Morin years ago. Did you not measure NOx & O3? 15N-NOx = 15N-NO3- only if O3 >> NOx (which I will assume it is the case in the middle of the ocean), otherwise 15N exchange btw NO and NO2 may interfere. Regarding the NO+HO2 hypothesis, I do not agree that it is so direct with 18O budget. NO+HO2 will also lead to O3 production and thus NO will still be massively oxidized by O3. It is very rare that HO2 dominates NO oxidation over O3.In fact HOx on the coast is usually pretty low compare

to inland sites.

Katye Altieri: Yes, [O3] >> [NOx] although I didn't present that. The oxidation of NO by HO2 or RO2 will still lead to the NO2 having an influence from the low d18O of HO2. There is no other way to really account for the low d18O observed. Unfortunately, we don't have D17O data for summer, only for winter and spring. The springtime HOx concentrations from air masses that have circulated over sea ice can be very high - the Brough 2019 paper (GRL) was one of the first to document this, it is a bit unusual but can happen.
I'd be happy to discuss this more - I glossed over a lot of isotope details as the talk was for a general audience. But we really struggled to explain such low d18O values and invoking RO2/HO2 seemed the most likely suspect. The Brough paper is from spring, but the mechanism would be just as robust in summer when we are sampling over the sea ice.

Markus Frey - very nice results. did you sample & analyse by any chance snow on sea ice? This would allow to better understand sea ice sources. The low atmospheric N15(NO3-) observed nearby in coastal Antarctica at Neumayer (Wagenbach,JGR,103(D9),1998) is attributed to katabatic outflow, consistent with equally low values on the plateau (Winton, 2020; doi:10.5194/acp-20-5861-2020).
Katye Altieri: Unfortunately no, but the plan is to collect the snow on sea ice in future campaigns.

Paul Zieger (SU): Thanks for your nice talk, Katye! Out of curiosity: do you also observe that fine mode (sea spray) particles are enriched in calcium? (in case you measure it)
Katye Altieri: We do measure it, and yes fine mode calcium concentrations are detected. Jess is analyzing these data for CRiceS. I don't have a plot off hand of nss-calcium concentrations. Interesting to check if you also see this enrichment we saw in the lab and the Arctic: https://agupubs.onlinelibrary.wiley.com/doi/10.1002/2016GL070275 I will forward to Jess - something to explore!

13:10 - 13:25 Andreas Massling: Trends in Arctic aerosols and implications for Arctic cloud formation

Santiago: Andreas , I wonder have you seen any evidence of resupended glacier dust?. I have ocassionally seen dust activity in the NE corner of

Greenland (satellite images). But I think it happens further south from that site. Also, not sure if your sizing instrumentation sampled coarse mode particles.
Dear Santiago, we have OPS measurements, but unfortunately we have not been looking in the data. I will have a student soon to do that. We have also the problem with Villum being few weeks snowfree in summer, then we measure probably local resuspension!

Dominic Heslin-Rees: When you stated that the positive trend in the nucleation mode was a result of increased transport from the open ocean in Baffin Bay during the summer - did you confirm this with back trajectories? basically, Villum station is seeing an increase in air masses originating from the south-west?
Dear Dominic, yes that is based on back trajectories. The paper by Pernov et al. will come out soon.

It is interesting that the accumulation mode also increases in summer, and therefore the condensation sink is increasing - wouldn't you expect a decline in nucleation given the increase in the sink?

The two issues are definitvely working qualitatively against each other, but for now the accumulation number increase is not significant. Quantitatively, it is difficult to say how much that affects. We will have to look into more data in the near future.

Hyojin Kang (KORPI): It is interesting to me! because I'm calculating and analysing on the Hygroscopicity (kappa) on Zeppelin observatory, Arctic region nowadays. So, I am just wondeing that how to calculate critical diameter (Dc) to get the kappa value. Did you use only CCNC and PNSD data, not used CN data for Activation ratio? If you used CN data as well, what did you use the CN data? CN more than 10? or 3nm? and please mention about reason.

Dear Hyojin, we did only use CCNC and PNSD. The method is of course not perfect, but you cannot place a DMA in front of a CCN counter at these low concentration values. We integrated the size distribution from larger particles to smaller ones until we match the number measured with the CCN counter.

Yes, the data I am using one is also not connected with DMA with CCN

counter. I am just used to calcuate Dc, https://doi.org/10.1016/j.atmosenv.2007.09.024. I'm just wondering when you take this method to calculate Dc, What did you use the CPC data. :) Thank you for your reply!

I think I remember our PHD student had for some time a CPC running in parallel, but maybe not all the time. I guess, he used the CPC for comparison to the SMPS and partly also for normalization, but I have to check that. The paper will come out in ACPD hopefully in the next few weeks:-)

Paul Zieger: Thanks for your nice talk, Andi. Is also Villum becoming more marine, like Zeppelin Obs? https://acp.copernicus.org/articles/20/13671/2020/

Dear Paul, the new paper from Jakob will come out soon and then I can direct it to you:-)

These small cloud seeds are seen in direct measurements at Zeppelin (https://acp.copernicus.org/articles/21/8933/2021/) but also at the North Pole using CVI measurements (paper still in review).

It is still a mystery how the very small ones can do it. Very interesting.

13:25 - 14:00 Questions and Discussion

Session 3 - 1800-2000 UTC+2

18:10 - 18:40 Megan Willis: Insights and ongoing questions from the NETCARE project on summertime Arctic aerosol and CCN

Kerri Pratt: To anyone interested, we recently published an observation of solid organic-coated sulfate particles in the summertime Arctic. These particles are consistent with the formation mechanism of sulfate NPF with organics contributing to growth that was explained in Megan's talk from NETCARE results. The impacts of this unexpected particle phase on clouds still needs to be explored. https://www.pnas.org/doi/10.1073/pnas.2104496119

18:40 - 18:55 Hannes Griesche: Surface-coupling effect on heterogeneous ice formation in Arctic mixed-phase clouds during MOSAiC

Paul Zieger (SU): Thanks for your nice talk! In your example from 25 June 2017: Did the potential temperature profile change for the first and second cloud period? Not sure I understood it correctly: Why did you observe (during MOSAiC) so few ice clouds at even very low temperatures (during summer)? Is this real (=missing INP?) or a retrieval issue? Are you planning to do similar analysis for Ny-Ålesund? Would be interesting to link to our NASCENT observations (https://www.aces.su.se/research/projects/the-ny-alesund-aerosol-cloud-experiment-nascent-2019-2020/)
Hannes Griesche (TROPOS): The reason, why the later cloud was identified as decoupled, was the higher liquid layer base height, compared to the earlier clouds (the same potential temperature profile was applied to derive the couping state). And the reason for the few number of ice clouds at low temperatures is definitely an issue of the retrieval. Here I still have to do some work. Once the method works well, I would be very interested to apply it also to Ny Alesund data.

18:55 - 19:10 Christina McCluskey: Simulating Remote Aerosol and Ice Nucleating Particle Populations
Gabriel Freitas: Would the inclusion of bioaerosols in the model have an impact?
Christina McCluskey (NCAR): Thanks for the question! I tend to expect over the SO that the sea spray aerosol (salts + OM) surface area is largely dominated by the sea salt component... but I would also aruge this is still something to test. In know that using simulated organic matter from OCEANFILMS (Burrows' work film drops) showed that marine organics certaintly contributed over the SO (increased surface area over the SO summertime by up to a factor of 2, this is not published work). In terms of how M18 performs with/without bioaerosols in the INP populations, we could consider the results from our obs closure study - see respons to Paul Z. Below!

Paul Zieger (SU): Thanks for you nice talk. In your nephelometer derived parameterization, is this mainly sensitive to the organic-coated SSA particles (in the accumulation mode)? It was a bit fast: What was the difference between PM1 and PM10 in your INP closure study?

Christina McCluskey (NCAR): Thanks for the Q - I agree, I had too much!! Using the PM1 and PM10 nephelometer-derived surface area concentration, we see a we see a -21% and +20% bias, respectively for untreated data. Using the PM1 and PM10 nephelometer data, we see a -9% and +20% bias, respectively for heated data (removes heat-labile INPs)

Jessie Creamean (CSU): What do you think the ice nucleating entities were on the S-rich SSA versus the `normal SSA (from Cindy's measurements)? Is the sulfur basically a tracer for organic materials that would likely be the INPs, since sulfur would not be expected to be correlated to ice nucleation activity?
Kerri Pratt (Michigan): This is a great question Jessie! I wonder if these could be solid particles, like the solid org-coated sulfur particles (marine derived) that we observed in the summertime Arctic. Is that possible? Does Cindy have images of the sulfur particles for comparison? https://www.pnas.org/doi/10.1073/pnas.2104496119
Christina McCluskey (NCAR): Here is Cindy's paper: https://doi.org/10.1029/2020EF001673;
Kathryn Moore (CSU): @Kerri- yes, we/Cindy do have TEM images of the sulfur particles from INP residuals (and the bulk collections). We have more images of total aerosol and fewer of INP residuals because they are hard to collect. We also have STXM-NEXAFS data from (some of) the same grids, if that's of interest: https://www.tandfonline.com/doi/abs/10.1080/02786826.2020.1845296

19:10 - 19:25 Adele Igel: Potential consequences of low aerosol_ concentrations in the Arctic on clouds
Paul Zieger (SU): Thanks Adele! For the MOCCHA/Arctic Ocean 2018 expedition, we have direct measurements of cloud residual size distribution (avoiding indirect measurements using CCNC's), similar as we have done at Zeppelin Observatory (https://acp.copernicus.org/articles/21/8933/2021/). Aitken-mode particles are almost always activating. Some of those Aitken-mode particles indeed need higher SS compared to the Mauritsen figure.
Adele Igel (UC Davis) Thanks, I will check that out.

Jessie Creamean (CSU): I wonder if you would have a better chance finding cloud dissipation cases from MOSAiC, since we often had times where we had very low and very high aerosol concentrations? And it is for a full year :)

Adele Igel (UC Davis) Yes, probably! Our project started before MoSAiC was complete, but we've often talked about looking at that dataset.

19:25 - 19:50 Questions and Discussion