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- **Start comments with your first and last name, example:**
  - **Jennie Thomas: Question for Antoine, what is the main impact of your figure on slide 6?**
- **We have prefilled the talks that are occurring in each session, please note questions for the interactive discussion during the talk.**
- **You can respond to questions during the interactive discussion and also pose extra questions during the discussion time.**
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********** **3rd CATCH Open Science Workshop 9-13 May 2022**

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Day 2 - 10 May 2022: Ocean-ice-snow-atmosphere fluxes (Markus Frey & Jen Murphy): Presentation of the current approaches to quantify trace gas and aerosol fluxes above ocean, ice and snow surfaces. Discussion of the strategies to overcome limitations/uncertainties in measurement strategies and how to use this flux information within models.

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.

Today's Poster & Social Session is for 1hr prior to each oral session, but posters are online all week so stop by in GatherTown to discuss / leave your comments:
https://app.gather.town/app/ueKHvojBKqY9wBgq/Catch%20Science

Markus Frey (maey@bas.ac.uk) | Jennifer Murphy (jen.murphy@utoronto.ca)

Session 1 - 0800-1000 UTC+2

Welcome and introduction - Markus Frey

Instructions from Jennie: Insert comments and questions for Markus here

Trace gas flux in/above snow - Detlev Helmig

Joel savarino - partly agree with Detlev, chamber enclosure can be difficult to conduct. The bag experiment does not respect the actinic flux radiation, light in snow is largely dominated by scattering. It might also be important to characterise the flow conditions through the snow sample such as residence time, are there channels, what fraction of the sample is flushed by the air/gas. At least in the lab this is (sometimes) an issue (thorsten bartels-Rausch)

Joel Savarino - The diurnal cycle show that after the first a sterady regime is reached for the NOx concentration. This means that the chamber is entirely flushed, macroscopically there is no area where NOx accumulate. Also given the porosity of the snow, snow is at 70 % of volume is air. Only close to ice density, flow can channel.

Thorsten Bartels-Rausch - What about air fluxes when sampling snow. Mixing/dilution with air, flow..

Joel savarino: volume pumped is very small so the dilution effect is negligible. For the snow tower at Concordia, each inlet pumps at 1l/min for 5 min. 5l of intersticial air is a volume of 8 L at 0.3 density, a small sphere around the inlet. there is also two connect inlet at 180° for each inlet to further decrease the dilution effect.

Just for interest: In the lab we sometimes check by adding a non-reactive gas the partitioning of which is well known. Then - based on the residence time - and the known partitioning and known surface area of snow sample - we can confirm that the gas is in contact with the entire sample. IS that something useful for the field?

Eddy accumulation HONO flux measurements - Jörg Kleffmann

Water vapour fluxes above snow in conditions of drifting and blowing snow - Armin Sigmund

Byron Blomquist - Did you consider making the flux measurements higher than 2m? Say, 10m height? This would have limited the atrifacts in the EC measurement from blowing snow.
answered during session.
Armin S.: Yes, that would be beneficial. Additionally, that would allow us to capture flux contributions from blowing snow particles above a height of 2 m because the flux corresponds to the net exchange happening below the sensor height.

Jennie T.: Can you let us know the main important differences between cryo-WRF and polar-WRF?
The cyrowrf publicaiton is here for the reference of others:
https://gmd.copernicus.org/preprints/gmd-2021-231/
Armin S.: As far as I know, the main differences are that (i) CRYOWRF uses a more detailed model of the snowpack as the land surface model and (ii) CRYOWRF models drifting and blowing snow processes using a 'fine mesh' with additional model levels between the surface and the first WRF level. The main idea is to achieve more realistic properties of surface snow such as surface temperature, grain size, and bond strength that are needed for a good representation of drifting and blowing snow and the exchange of latent and sensible heat and ultimately the surface mass balance.

Xin Y for @Armin: although partiles in sub- and micron size mode is small in the observation, the fration distribution does not represent their original production flux. In fact, it is at the steady state after production and loss of sublimaiton. Due to the relative short life-time, thus, the actual water flux from small particles could be very significant. This is what we derived from our blowing snow sea salt work, see Yang et al., ACP 2019. We can discuss it further if you like.
Armin Sigmund: Thank you for the comment, I was not aware of this special role of small drifting and blowing snow particles but I will take a look at your paper. With the default setting in our LES simulations, we only simulate particles with diameters between 50 and 2000 microns.
I remember you mentioned you underestimated water flux, I am wondering if it is due to the small particles contribution.
Armin S.: The underestimation I mentioned is a problem of the MOST bulk parametrization, not the LES. But if we compare the LES-based water vapour flux with the EC measurement, the LES indeed underestimates the flux. This is at least partly due to the fact that my LES domain is not big enough to

capture the contribution of the largest eddies observed in the field. Another part of the underestimation can be explained by the sensitivity to the upper boundary conditions for temperature and humidity and the instrument uncertainties, which make it hard to reproduce exactly the same conditions as in the field.
Xin Y.: Note that the curvature effect works even at a saturated moisture condition, therefore, it keeps working at RH=100% or above it. If my understanding (or work) is correct, then a large part of the water vapour should come from fine particles, whihc has not been "observed" so far. I just derived the BS particle produciton flux based on a top-down mehtod, thus needs further field or lab data to confirm.
Armin S.: That's interesting, so far I have only heard about the curvature effect in the context of cloud droplets. I will read more about your work, thanks for the input.
Markus F: Very interesting discussion. If there was a high vapour flux from very small particles they also should disappear very quickly after formation. To maintain a significant flux over a drift event would you not have to constantly resupply small particles? at steady state there seem to be very few of them at the lower end of the size range (<50um, beyond the SPC observation). so does it matter on longer time scales for the vapour mass budget?
Armin S.: I could imagine that small particles particularly form in the beginning of a drift event through fragmentation of drifting particles when they hit the snow surface. However, some studies argue that the drifting snow particles soon become rounded, which may limit the formation of small particles. Just some speculation.
Xin Y.,: If the momentum (winds) stays, then there will be supply of fine partiles form the saltation layer, right? Also, the rounding effect for large pariticles is also driven by the curvature effect, not the classic moisture gradient, right?
Armin S.: I agree that the curvature effect should contribute to the rounding effect. In the LES simulation, we assume spherical particles and it is an open question how realistic this assumption is.
Xin Y.: agree, there are many unknown issues re ice particle sublimaiton process. I am happy that you are looking into this issue. As most "recent" study on this topic is several decades ago (this conlcudion may not be correct :).
Armin S.: Thanks again for the comments. For me it is hard to judge whether there will be continuous supply of fine particles from the snow surface during a drift event. It could also be that the fine particles are

eroded and sublimate in the beginning, leaving the larger particles behind.
Xin Y.: yes, the quick and fast sublimation rate from fine particles may prevent large particles further sublimaiton due ot the release of water vapour (in a close to saturated condition).
Armin S.: Yes, I agree, at least as long as there is a supply of small particles.
Markus F.: there are no observations, the Byrd Experiment in the 60s collected on formvar plates, which led to the gamma distribution model. @Armin could you update the model including the Kelvin effect of (in)finite supply of small particles? just thinking on my feet here.
Armin S.: Good question. I guess the Kelvin effect is also modeled in cloud models. So it may be possible but I have to look more into it.
Xin. Y.: worthy trying. anyway, thanks for the discussion. I got to leave.

The role of the atmospheric boundary layer above snow/ice for vertical exchange fluxes - Timo Vihma

Jennie T.: Is sea smoke a comment wording for this effect over leads?

NOx flux chamber measurements in Antarctica - Joel Savarino

Jan Kaiser: How do you explain the O3 mole fraction increases at Dome C in summer?
Joel Savarino - HONO is also emitted by snow forming OH, OH oxidizes CO and gives HO2, HO2 reacts with NO without consumming O3 --> prod O3
Jan Kaiser: Do you see the corresponding high O3 concentrations in your chambers?
Joel Savarino - no actually we see the reverse, almost zero O3 because we are injecting zero air and O3 takes time to be formed. with respect to the flushing time of the box, ca. 5min, there is no time for O3 to be formed. Also I don't known the CO concentration. The feeding air is zero for NOx and O3, don't know if the purification system also traps CO.
Jan Kaiser: I would have thought that photolysis was the rate-controlling step, not the subsequent secondary reactions (or flushing), but I haven't done the calculations. Presumably CH4 would also produce O3 (there is much more than CO) and is probably not destroyed by zero-air generator? Maybe the HONO photolysis is too slow?

Joel Savarino - the controlling rate for ozone production is indeed the photolysis of NO2 but a pure NOx chemistry will give you zero O3 prod because as soon as it formed it is consumed by NO. To produce O3 you need to short cut NO+O3 reaction and it takes time to produced the right components to lead to O3 formation. In urban environment where ozone is largely produced, the production rate are at few ppb/h. At DC it is also around this range, too long for the chamber expriment as it is set up. Actually we didn't want O3 to be formed if you want to quantify NOx production, better to keep alive your primary emissions as long as possible.

Jan Kaiser: Yes, that's a good point. Did the towers measure vertical O3 gradients above the snow surface? (maybe a question for Detlev)

Joel Savarino - despite 10 years of record, and may be for only few rare cases (but never explore in detail all the data), we never observed NOx gradient up to 5m (our highest inlet) so for ozone it will be even worst. In fact the atmosphere is highly turbulent in the BL, mixing is faster than emissions. even when you compare the inlet at few cm above the snow and the one at 1m you don't observed a difference.

Jan Kaiser: Just found the paper by Helmig et al. (2020), Fig. 8 (https://doi.org/10.5194/tc-14-199-2020). [I tried posting the figure here, but EtherPad doesn't allow this]. Looks like the +45 cm O3 mole fractions are higher than at +15 cm, and that there is some O3 enhancement at -10 cm (within the snow). No difference between -30, -50 and -70 cm though. Presumably, above +45 cm, mixing takes over and blurs the signal. I guess the title ("Impact of exhaust emissions on chemical snowpack composition") suggested that the source of nitrate is station pollution, but the mechanism should be the same.

Joel Savarino - Be careful with O3, the production is in the air not in the snow so the flux is reversed, O3 is penetrating the snow and the gradient is very weak, just few ppb, almost the precision of the instruments. Indeed too bad we can't show figures, I could have shown the detailed plots with a srong in snow gradient of NOx and almost nothing in the atmosphere. It all depends how turbulent is your atmosphere. Only few m/s is enough to well mix the BL.

Regarding the title of the paper, Detlev focussed on the station contamination bt it is only few episodic events. Nevertheless we are still questioning the local snow. It is obvious that it is contaminated by the BC (filter are grey black after few hundreds ml of filtration) but apparently this does not change the optical properties of the snow, at least for nitrate photolysis, scaterring still largely dominates.

Regarding nitrate concentration in snow, station may have influenced it but we need to process in more detail snow pit profiles. One interesting thing about NOx pollution is its sharp profile. It is almost a dirac impulse in the atmosphere and so its propagation into the snow pack can completely characterize the diffusivity of the snow as a function of the wind shear. Figure 6 show a clear event like this. You have a time-depth dependency, all you need to slove the fick's law of diffusion.
Markus F: Our measured firn air profiles of O3 in summer (Jan) show a slight increase of O3 with depth (Frey et al.,2015; doi:10.5194/acp-15-7859-2015). Summer NOx gradients above snow are strongest between 0.1 and 1m, but are still significant up to 4.0m above snow, especially in the evening when a shallow BL forms. Just for polar day: mixing during the day at DC slight convection/ unstable conditions, at night strong decrease of Eddy Diffusivity but you continue some mixing driven by wind shear.
Joel Savarino - @Markus, yes the snow shows a strong and significant gradient of NOx and O3, it is much less evident in the atmosphere, mixing prevails except during few singular events when wind is close to zero.
Jan Kaiser: Who is modelling this (i.e., the O3 enhancements above the surface with your measured NOx production and transport rates)?
Joel Savarino Modeling what? The chemistry? the flux? the transport in the snow? Lennie Thomas has a snow pack chemistry transport model. In Kukui et al., 10.5194/acp-14-12373-2014, you will find a box model that deal with short life species. In Legrand et al., 10.5194/acp-16-8053-2016, 2016. you will find the basic calculations of the ozone production. You don't really need a complex model for that.
Jan Kaiser: Thanks. Lots of papers to read. BTW, congratulations on DOC-PAST!

Jörg Kleffmann: Did you also quantified HONO by the IBBCEAS technique?
Joel savarino - unfortunately IBBCEAS NOx does not have access to the HONO absorption region. IO, CHOCHO, and O3 are also accessible but not HONO.

Jörg Kleffmann: Did you covered the chamber by different filters to confirm the nitrate photolysis e.g. by plexi glass filter (380 nm) or window glass (340 nm)...?
I catched that with your special plexi glass...
JoelSavarino - It was may be not very clear on the figure but the special plexi glass we are using is transparent up to 200 nm, more than the fall off of the solar spectrum. We did dark experiments by covering the chamber

snow with a plastic bag and the concentration reached DL after the flushing time. Actually it is how we know the flushing time of the chamber at a given flow rate, just cover it but did not change the solar spectrum with filter. Mau be a good idea to test in the field.

GHG flux chamber measurements above sea ice - Daiki Nomura

Jennie for @Daiki - I think one difference in the way we think is that atmopsheric boundary layer vertical mixing is not important for CO2/GHG exchange becuase they are too long lived. While for atmospheric chemists, one important factor in how I think about chemical fluxes is how fast air is pumped away from the ice/snow surface. What do you think about this? Does the closed chamber impact how we should think about/use this flux compared to open atmopserhic systems? Actually same comment for @Joel.
Joel Savarino - I don't think so. The difference between long life species and short ones is in the first case you are pouring your products in a infinite reservoir while not in the second case. The gradient concentration method in the atmosphere cannot work for the first case. Until you don't find a gradient somewhere (snow vs air, soil vs air water vs air), the chamber experiment will be the only solution. e.g. it can be difficult to probe a soil at different depths.
@Joel - good point. I think there are cases where dynamics (such as wind pumping) plays an important role in determining emissions fluxes to the atmosphere. I'm not sure how to resolve this to use these fluxes in models.

John Prytherch - @Daiki, do your measurements show ice-atmopshere fluxes persisting through long periods, or could the substantial fluxes be limited to the initial onset of the melt / freeze seasons? I can imagine ice-atmosphere trace gas fluxes being significant on a regional level, but I was surprised to see the per m2 flux was comparable to that of water-atmosphere flux (maybe I misunderstood that slide).

Heavy metals and pollutants particles deposition in northern Tibetan_ Plateau glaciers - Zhiwen Dong

GENERAL DISCUSSION/ QUESTIONS

Session 2 - 1500-1700 UTC+2

Introduction and summary of session 1 - Jennifer Murphy

Keynote: Air-Sea Trace Gas Fluxes - A Focus on Turbulent Flux Methods for the Cryosphere - Byron Blomquist

John Prytherch - Thanks for the nice talk Byron. It was interesting to see the sigma_w / ustar ratio from Polarstern. There was some variation there even in the'good' wind sectors, between about 1 and 1.5 perhaps. I wonder if you are planning or have any way to quantify effects of this flow distortion on the measurements, perhaps with a comparison of the ship-based measurements with those from the on-ice mast(s)?

Yes, as I said, I'm not sure how to intrpret the small variations from the Similarity relationship. Fluxes measured at the two towers differed in measurement height and footprint, so there are many potential sources of variability between the two. Something to think about.

Jen Murphy – you mentioned the importance of “knowing your boundary layer”. Are there any sites in snow/ice-covered regions that are well-characterized with respect to boundary layer characteristics and energy fluxes so that guidance could be provided to atmospheric chemists about requirements for their trace gas/aerosol measurements and inlet design?

I suppose the best option is to work at a site that is already well instrumented for ABL characterization. Otherwise, you will need to bring along the measurement capabilities to do the ABL profiles yourself. For specifice recommendations for how to set up a flux system, this is certainly a worth while objective of an overview paper, since many may not be familiar with all the details.

Keynote: On-line sea-air fluxes of sea spray and of VOC using ship-borne interfacial enclosures - Karine Sellegri

Gabriel Freitas: From which depth did you sample the water for your mesocosm experiments?

GENERAL DISCUSSION/ QUESTIONS

Session 3 - 1900-2100 UTC+2

Introduction and summary of sessions 1 & 2 - Jennifer Murphy | Markus Frey

Arctic sea spray flux measurements - Matthew Salter

Kerri Pratt: Nice talk Matt - those sure were hard measurements! When thinking about the context of the Held et al. study, I think it's important to consider that the wind speeds were <3 m/s, whereas Nilsson et al 2001 shows a strong wind speed dependence on lead-based SSA formation.

Matt Salter: Thanks! Yes I totally agree. Going forward I think we will need to combine aerosol flux chamber measurements in combination with eddy covariance measurements to fully grasp the range in the source flux under different conditions.

Parameterization of sea salt from blowing snow in GEOS-Chem - Betty Croft

Helene Angot: Thanks Betty for this great talk! You showed two model outputs for the November storm during MOSAiC (with and without blowing now). How does this compare to in-situ observations? (I may have missed that) (I won't be able to stay until the end of the session, I'll read your reply tomorrow)

Kerri Pratt: Thanks for this talk! Have you considered the complexity of the snow composition and whether other types of aerosols in the snow could

be produced from blowing snow (beyond sea salt - from the deposition of soot, etc - e.g. Erin's & Nicolas' talks)?

Measurements of chemically-resolved aerosol fluxes by eddy covariance in Alaska - Erin Boedicker

Kerri Pratt: Excellent, beautiful measurements Erin! Great talk too!

Jen Murphy: It seems like the wavelet approach works very well, but could it also be possible to relax the stationarity criterion if the co-spectral density is mostly at higher frequencies? E.g. maybe averaging for only ten minutes is enough?

@Jen - Yes, you could absolutly do that. The reason we didn't is that the cospectra were noisey (which is often the case with particle flux) and so we wanted a route that would be more independent than that.

Ice cores record deposition of perfluorinated pollutants in the Arctic - Cora Young

Black carbon and other light-absorbing impurity flux to snow in the Andes - Nicolas Huneeus

Jen Murphy: Does the majority of black carbon deposition during the winter result from scavenging by falling snow, or dry deposition to the existing snow/ice on the glaciers?

GENERAL DISCUSSION/ QUESTIONS

Review paper or special issue on surface fluxes in the cryosphere ? Is it time? Is there any interest?