Properties (and open questions) of Arctic Haze observed by lidar in Ny-Alesund, Spitsbergen

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Outline:

What is Arctic Haze? The site and the (aerosol) remote instruments of Ny-Ålesund **Properties of Arctic Haze:** optical properties, from optical to microphysical properties, hygroscopicity **Open Questions:** Closure ("overestimation"?), pollution pathways Next steps: MOSAiC (pathways), AC3: radiation & clouds

Arctic Haze: spring-time "air-pollution" in the Arctic

What do we know about Arctic Haze: Can reduce visibility Composition: sulfates, organics, few metals, (Ny-Ålesund) sea spray, little BC (Udisti 2016, Tunved 2013)

Origin: mostly anthropogenic (Quinn 2007), but also forest fires (Warneke, 2009)

mixing state: turns into internal mixture during aging (Hara 2003)

Size: around 0.2µm diameter (Tunved 2013)

Max AOD due to size not due to concentration!

Photo: Extreme event, agricultural flaming May 2006 (Stohl-2006)

Tunved 2013, ACP: Arctic aerosol life cycle



Arctic Haze in spring: because particles are larger, have larger scattering efficiency

Max. aerosol number concentration in summer due to marine aerosol

Scattering efficiency, Mie theory:



Arctic aerosol is generally small and at the edge of visibility!

Typical AOD values from Toledano 2012 Atmos. Environm.



Spring: Arctic-AOD > N-European-AOD No Haze in Scandianvia No "easy" direct pollution transport from Europe



Scandinavia

Contrary: Eckhardt 2003 (Flextra, CO Tracer) "NAO + faciliates transport into Arctic"

Aerosol may have different pollution pathways than trace gases!



Ferrero et al, ACP 2016: vertical profiles of aerosol Tethered balloons with particle counters

- a) const. aerosol load – (but convection seldom)
- b) More areorol higher up – advection!
- c) More aerosol close to ground (sources?)
- d) Difference Aitken vs. Accumulation, strange temp. profile



Aitken particles, accumulation mode particles, poten. temp, rel, hum.

division: "circulation of the atmosphere": Runs climate models: global, regional (future: local) For improved understanding of physical and meteorological processes Chemistry of the stratosphere, operate AWIPEV (Koldewey-) station on Spitsbergen



European Arctic warm: Gulf stream, Westspitsbergen current Spitsbergen treaty 1920



Ny-Ålesund, 78.9°N, 11.9°O – one of the northernmost settlements:



Coal mining until 1963

Today science village (I, D, No, Sk, J, Cn, Kor, ...)

- (+) cheap and quick accessible, comfortable
- (-) warm for the Arctic, mountains introduce "micrometeorology"
- (?) testbed for future



The peculiarities of Spitsbergen:



DJF temperature trend at 850hPa using ERA-Interim 1996-2016 "center of wintertime warming" Dahlke & Maturilli, 2017: ¼ of warming due to more efficient advection from Atlantic Maturilli 2015: strong winter warming also in our data (BSRN, surface)

Annual average temp (April 18 – March 19): -3°C

West Coast Spitsbergen is transition between N Atlantic and Arctic. May become "more Atlantic" in future

Koldewey (AWIPEV Station):





Since 1992. 2004 united with frech IPEV

Different projects:

-Biology -Permafrost -Atmospheric research

Automatic stations (T, rh, wind, radiation, cloud altitude) regular balloon launches, Eddy covariance; Remote sensing



Jan 2019: winter-campaign

Tonight!!!! Tue, 21. Jan 17:30

KARL: Koldewey Aerosol Raman Lidar

Backscatter (β) @ 355nm, 532nm, 1064nm Extinktion (α) @ 355nm, 532nm Depolarisation (δ) @ 355nm, 532nm Water vapor (mr) @ 407nm, 660nm



Spectra 290 /50 Laser (10W / colour) 70cm mirror Fov: 1 4 mrad Licel transients, Hamamatsu PMTs Overlapp > 700m Tropo- & stratosphere









Challenges with extinction in lidars:

Regardless of extinction profile in atmosphere: impact on lidar profile is infinitely differentiable

$$\exp\left(-2\,\int_{z_0}^z\!\alpha(\hat{z})\,d\hat{z}\right)$$





Extinction in a lidar:

$$\begin{split} P_r \left(z \right) &= C_r \rho(z) \frac{1}{z^2} \exp(-\int_0^z \alpha_{\lambda_e}(\hat{z}) + \alpha_{\lambda_r}(\hat{z}) d\hat{z}) \cdot \\ \left[O(z) \right] \end{split}$$

Do not smooth or fit your lidar profile !!

Instead you can calculate a "layer-integrated" extinction ($z_{bottom} \rightarrow z_{top}$)

$$P_r(z_t) = C_r \rho(z_r) \frac{1}{z_r^2} \exp(-\int_0^{z_b} \alpha \dots dz) \cdot \exp(-\int_{z_b}^{z_t} \alpha \dots dz)$$
Layer AOD
$$\frac{P_r(z_b) \frac{z_b^2}{z_b^2}}{C_r \rho(z_b)} \text{ If the derivative } \frac{\partial}{\partial z} \text{ harms, avoid it}$$

Or make a statistic from unsmoothed lidar data (first calculation, then averaging)



What does an aerosol lidar deliver:

extensive quantities (dependent on aerosol number concentration):

backscatter (concentration, size, shape, refractive index) extinction (concentration, size, shape, refractive index) !

Intensive quantities (not dependent on aerosol number concentration)

depolarisation $\delta = \frac{\beta_{\perp}}{\beta_{=}}$ (shape) [dipole moment]

colour ratio CR = $\frac{\beta_{\lambda 1}}{\beta_{\lambda 2}}$ (size) [$\beta \sim \lambda^{A}$ -4 < Å <0]

lidar ratio $LR(\lambda) = \frac{\alpha^{aer}}{\beta^{aer}}$ (index of refraction, size, shape)

Knowledge of δ , CR, LR allows a robust classification of aerosol type (dust, smoke, sea salt, cirrus...)

 \rightarrow it's about getting the intensive quantities!











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Intensive quantity: aerosol depolarisation (shape)





Particles more spherical outside haze season! (Mie better)

Extensive quantity





2500m - 3500m 3500m - 5000m 5000m – 7000m

> Size more uniform in Feb?? More uniform in low altitudes



Mixing state of aerosol:



Sort aerosol for size and shape: still very inhomogeneous LR:

Chemistry unrelated to size and shape

On scale 30m/ 10min no individual soot, sulphate, crust ... particles

Color ratio, depol. ratio both intensive quantities



Inverting lidar data:

Aim: estimate size distribution n(r) (r_{eff} , σ , N_0) and refractive index m from lidar data Assume spherical particles, Mie theory, efficiencies $Q_{ext/\beta}$ are known \rightarrow set of Fredholm integral equations for extinction & backscatter

$$\alpha(\lambda) = \int_{R_{\min}}^{R_{\max}} Q_{ext}(\lambda, r, m) \pi r^2 n(r) dr$$

$$\beta(\lambda) = \int_{R_{\min}}^{R_{\max}} Q_{\pi}(\lambda, r, m) \pi r^2 n(r) dr$$

Retrieval of n(r) from Q, α , β is an ill-posed Problem At least 2 α , 3 β needed

But:

Lidar is only able to retrieve aerosol in accumulation mode: $0.1\mu < r < 1.2\mu$





Difficulty: determine index of refraction:

Lidar offers: backscatter and extinction. The refractive index depends on: scattering and absorption. Hence: from lidar alone an index of refraction is difficult to obtain. "Stripes" like this in the probability distribution do occur frequently.



Example case of Arctic haze:





Lidar and contemporary radiosonde: hygroscopic growth?

In-situ define scattering enhancement factor $f(rh) = (1-rh)^{-\gamma}$ Question: apply this to β (instead of σ)? Assumption: all lidar data in a given time / height should belong to "same event"



Open questions:

1. Does remote sensing overestimates extinction?

Tesche et al. 2014 ACP:

Calipso_extinction > in-situ (Zeppelin station)

(what was NOT published in) Lisok, 2016 Atm. Environm:

KARL_extinction > in-situ(Gruvebadet station)And extinction at ground, 1km,2km altitude not correlatedDeviations also at rh =50%



Needs to be clarified during MOSAiC: Less orography!

Open questions: 2. Pollution pathways

Graßl, 2019: Flextra with ERA-interim



5 days trajectories too short Reanalysis products show large differences <u>Slightly</u> higher AOD from Siberia

Sea ice as reduced sinks?



High aerosol load due to sources <u>and</u> sinks

Sea ice: dry, stable BL less vertical mixing, longer aer. life-time

Best conditions for aerosol transport: Air over source regions in BL with enough wind speed Ascend of the air (higher wind speed, 5 days, less precipítation) Advection over sea ice

FLEXTRA 5 days (with photometer) Aprils 2013-2016

MOSAiC: coordinated observations with surrounding stations needed

MOSAiC:

<u>M</u>ultidisciplinary Drifting <u>O</u>bservatory for the <u>S</u>tudy of <u>Arctic</u> <u>C</u>limate



Goal:

To improve the understanding and model representation of coupled atmosphere-ice-ocean-ecosystem-biogeochemistry processes

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Atmospheric observations during MOSAiC



13 stations with year long monitoring program + 2 AWI planes

Transparency from Marco Zanatta

³⁷ Several ground stations for meteorology and aerosol in-situ

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AC3 and PAMARCMiP 2018:



P*r²

1

0.5

Persistent layer of aerosol in 5-7km



Ny-Ålesund, Spitsbergen Range-corrected signal at 532 nm $\times 10^7$ KARL, 05-Apr-2018 8 3.5 7 Altitude [km above ground] 3 2.5 2 1.5

12:30

time [UT]

1

0 11:00

11:30

12:00

Polar5 flight-track towards Station



Compare remote sensing to in-situ

13:00

13:30

Calculate radiative forcing

Summary:

Arctic Haze consists of small particles Max. extinction in spring due to slightly larger particles Effective pollution pathways unknown: MOSAiC Weakly depolarizing particles with moderate LR, chem. "internal mixed" on 10min / 30m scale Maybe in-situ underestimates extinction



A theory is short, concise and complete and is believed by nobody except of its inventor.

Observational data are noisy, strange and incomplete and are believed by everybody except of the one who measured them.

Picture:

Loriot 1923 - 2011

Thank you for your attention!

Available aerosol measurements during MOSAiC

	POLARSTERN	M PAL	PAL VRS ZEP NYA OLI F			BAR	R CBA TIK HOR ERK SUM			Legend			Platforms				
AE	ROSOL BASI	0													Observed	AWI-A	AWI-Aircfrafts (Apr and Sept)
Total number concetration															Not observed	ALR	Alert
Total mass concetration															To be confirmed	сом	COMBLE (Andenes+Bear Island)
Number size distribution															No ifo	PAL	Pallas
Scattering																VRS	Villum
Absorption																ZEP	Zeppelin
Exinction (direct measurement)																NYA	Ny-Alesund
Cloud condensation nuclei																OLI	Oliktok
ChemistryOnline																BAR	Barrow
ChemistryOffLine																СВА	Cape Baranova
EC/OC																тік	Tiksi
	OTHER							•								HOR	Hornesund
refractory black carbon																ERK	Eureka
Hygroscopicity (HTDMA)																SUM	Summit
In siu cloud activation (CVI or interstitial)																	
INP (online-offline)																	
Wet scattering																	
Cloud residual size distribution																	
Cloud resiudal BC																	
Cloud residual composition																	
Cloud liquid water content																	
Droplet size distribution																	
CCN on cloud residuals																	
Aerosol ion spectrometer (NAIS)																	
Single particle BC both in aerosol and cloud residuals																	
Bioaerosol single particle (MBS) botn in aerosols and cloud residuals																	
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Aerosol measurements:

Can be direct ↔ indirect (physical model / other quantities needed)

 \leftrightarrow

Can be In situ DMPS, Filter, (OPC) "what aerosol is there"

In situ and remote sensing must not match closely: Shape of aerosol? Index of refraction? General scattering theory missing! For spherical particles: Mie

"closure experiments"

Note: prefer direct measurements $f(cause) \rightarrow effect$ always given;

 $f^{-1}(effect) \rightarrow cause ??$

Passive.

radiation

without own

remote sensing

"what is the optical impact

of the aerosol"



Active, emits

own radiation:

easier ranging

KARL: Koldewey Aerosol Raman Lidar Since 2001 Aerosol and water vapor 199? – 2008 also a lidar for stratospheric ozone





Spring AOD decreases over time → annual run of AOD becomes flatter 2009 was last polluted year Generally high variability

Principles and equation:

elastic:

$$P_e(z) = C_e \beta(z)^{1/2^2} \exp(-2 \int_0^z \alpha_{\lambda_e}(\hat{z}) d\hat{z}) \cdot [O(z)]$$

 $\beta^{\text{tot}} = \beta^{Ray} + \beta^{aer}$ P : return power [MHz] β: backscatter [m⁻¹sr⁻¹] $\alpha^{tot} = \alpha^{Ray} + \alpha^{aer}$ α : extinction [m⁻¹] O: overlap function

C: lidar constant (laser power, optics: transmission, PMT efficiency ...)

Raman –scattering, inelastic $\lambda_{elastic} \rightarrow \lambda_{Raman}$

$$\begin{split} P_r(z) &= C_r \rho(z) \frac{1}{z^2} \exp(-\int_0^z \alpha_{\lambda_e}(\hat{z}) + \alpha_{\lambda_r}(\hat{z}) d\hat{z}) \cdot \\ &[O(z)] \end{split}$$

 ρ : air density, Raman scattering at nitrogen molecules Extinction principally challenging! It all depends on SNR In total: 2 equations for α , β



Without Raman effect: 1 equation, 2 unknowns

If the green 532nm light is emitted into the atmosphere the strongest Raman return occurs at 607nm (Stokes from N_2), N_2 is proportional to air number density

$$P^{607nm}(z) = \hat{C} \cdot \frac{1}{z^2} \rho(z) \cdot \exp(-\int_{0}^{z} \alpha^{532} + \alpha^{607} d\hat{z})$$

As with photometer assume $A : \alpha_{Aer}^{532} := \alpha_{Aer}^{607} \cdot (532 \text{nm}/607 \text{nm})^{\hat{A}}$ \Rightarrow equation for α^{532} but noisy

$$P^{607nm}(z) = \hat{C} \cdot \frac{1}{z^2} \rho(z) \cdot \exp(-\int_{0}^{z} f \cdot \alpha^{532}(\hat{z}) d\hat{z})$$

does f depend on z?



Evaluation of lidar data:

If Raman channel is available and looks trustful:

- a) Solve Raman lidar equation for extinction
- b) Use this extinction to solve the elastic lidar equation for backscatter

If Raman channel is not available:

Estimate a Lidar Ratio $LR(\lambda) = \frac{\alpha^{aer}}{\beta^{aer}}$

Bring elastic lidar equation in form of Bernoulli Differential equation and solve it for the backscatter

You need (always): 1) air density profile

 $\beta^{tot} = \beta^{Ray} + \beta^{aer}$

2) boundary condition $\beta^{tot}(z_{ref}) = (1+\epsilon) \cdot \beta^{Ray}(z_{ref})$

$$P_r(z) = C_r \rho(z) \frac{1}{z^2} \exp(-\int_0^z f \cdot \alpha_{\lambda_e}(\hat{z}) d\hat{z}) \cdot [O(z)]$$

$$P_e(z) = C_e \beta(z) \frac{1}{z^2} \exp(-2\int_0^z (\alpha^{Ray} + LR \cdot \beta^{Aer}) \cdot [O(z)]$$
ANN

Extinction in a lidar:

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Or make a statistic from unsmoothed lidar data (first calculation, then averaging)



How do lidar signals look like?



Shortcomings of lidar data:

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Phase function missing: only info around \Phi =180 ^\circ
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overlapp: boundary layer difficult

Refractive index challenging: $m = m_{real} + i \cdot m_{imag}$ $m_{real} \sim scattering$ $m_{imag} \sim absorption$ but we only have β , α

Weak absorption $\rightarrow \omega$ insecure

Only trustful info for accumulation mode: Aitken: interacction too small giant mode: Mie efficiency becomes flat

$$\alpha = \frac{2 \pi r}{\lambda} \iff r = \frac{\alpha \lambda}{2 \pi}$$

$$\alpha \in [1 ... 12], \ \lambda=0,5\mu \rightarrow r \in [\frac{1}{4\pi} ... \frac{3}{\pi}] \mu$$

conclusion: aerosol, cloud particles



Results Haze season 2013:

Most polluted March / April Principally more backscatter close to ground Haze season starts close to the ground and lasts longer in higher altitude LR might have an annual cycle with lower values early in the season and closer to the ground During the haze period the particles are more irregular in shape

Below 850m time consuming overlapp corrections with Ceilometer required

Since 2011: Vaisala CL51

Calibrate Ceilo with KARL (Klett) in about 1.5km on clear, stable day Assume Ceilo measured true results (@910nm) Assume Å, LR(z) \rightarrow calculate missing overlapp of KARL

SNR(Ceilo) moderate



Definition "aerosol closure":



- 1) Closure on microphysics (locally)
- 2) Closure on radiative impact (column)

 Bring together different sensors (with different weaknesses and assumptions) in a common evaluation scheme until

 a consistent set of assumptions & properties is found
 a clear gap / mismatch has been identified
 best: homogeneous aerosol (composition, mixing state)
 dry, stable atmosphere, spherical particles
 problems: scattering theory (who does not depend on that, entirely?)
 refractive index & shape, espec. when heterogeneous particles, hysteresis of rh ...

at the end of a successful closure we have a match between "what is there" and its optical impact

2) understand the relation between microphysics and optical properties for the whole column and include integrating values as AOD, radiation values and the resulting forcing by a radiative transfer model problems: air-borne in-situ measurements for validation At the end: knowledge of RF of aerosol, depending on meteorology



Comparison in-situ, remote sensing



Red: AMALI lidar, downward looking

Blue: sunphotometer Extinction from $AOD(z_1) - AOD(z_2)$

So we have: Backscatter from lidar Extinction: from lidar and from photometer 1st comparison in situ to lidar, case April 4, 2009



Extinction in photometer(s) smaller than in lidar, cause:? Optical systems insensitive below 80nm So, accum. mode: basically log-normal distribution

Summary

Lidar active remote sensing, give height resolved information on many parameters. Aerosol: backscatter, extinction & depolarisation

 \rightarrow allows (sometimes) estimation of size distribution & refractive index in accumulation mode

Is handicapped in Aitken mode can easily see giant mode, clouds

Should be compared to ground-based in-situ data: Is the aerosol in the column / free troposphere the same as on ground?

Has typical resolution of minutes / 10m

Arctic Haze: spring-time "air pollution"



Arctic Haze: Sulfates, soot, ... Small particels, but they can scatter light

Max. in spring but large year to year variability Sandra is analysing it:



Motivation:

Warum interessieren wir uns für das Klima der Polargebiete? **Telekonnektion** der Atmosphäre! Korrelation der Abweichung meteor. Größen auf Skalen bis 10000km



Eine wärmere, eisarme Arktis könnte die Wahrscheinlichkeit kalter Winter in gemäßigten Breiten erhöhen



"Arktische Oszillation"

warum? Rückkopplungen "Feedbacks" Ursache \rightarrow Effekt (der) \rightarrow Ursache verstärkt \rightarrow (stärkerer) Effekt ...



Eis-Albedo Rückkopplung

2014: Pithan, Mauritsen (MPI-Hamburg)

Arktische Verstärkung nur durch Temperatureffekte

(generell) Strahlungsbilanz:



Absorbierte Energie > 0 → Erwärmung

Aerosole:

Alle festen oder flüssigen Teilchen in Atmosphäre als Trägergas:

Wüstenstaub (Sahara, Gobi)



Waldbrände





anthropogen

weiterhin: Seesalz, Vulknstaub,

"sekundärem Aerosol", aus reaktiven Gasen gebildet

Durchmesser: $10nm - 10\mu m$ (Sedimentationsge-schwindigkeit ~ r^2)

Ganz unterschiedliche Form, Größe und Chemie

Ohne Aerosole kein Niederschlag!

Betrachten kleine Wassertröpfchen:

 Haben hohe Oberflächenspannung
 → haben höheren Sättigungsdampfdruck als große Tropfen
 Tröpfchen verdunsten – Tropfen wachsen "Kelvineffekt" "Oberflächenspannug aufrecht zu erhalten, kostet Energie, Natur will das vermeiden "



Radius → ∞

Aerosole beeinflussen Strahlungsbilanz:

direkt (Streuung, Absorption – abhängig von Albedo) oder indirekt (Wolkenbildung, deren Lebensdauer, Helligkeit) "forcing"





Aerosole ändern ständig Durchmesser und chem. Zusammensetzung Verschwinden aus Atmosphäre durch Niederschlag, Sedimentation

Consider an aerosol cloud:



b) Internal mixing:



Chemically external mixture: most particles have one chemical comosition

Internal mixing: most particles have heterogene composition

Aging: Due to Brownian motion coagulation, more internal mixture But also new particle formation due to SO_2 , O_3 und UV radiation

Problems with climate ...

Deficiencies in physics:

Aerosol: scattering properties (shape, size, index of refraction!), interaction with clouds Clouds: size, altitude, phase (IN), life-time, precipitation, brightness "nothing known" Turbulence (origin of) long-scale variations, circulation pattern

Dependence on external forcing: sun, cosmic rays, sea ice, ocean

Deficiencies in description:

Non-sufficient horizontal and vertical resolution: parameterization Reifen & Toumi, GRL 2009: "non-stationarity of climate feedbacks" ... \rightarrow models which are good in one period might be inferior at other times

Principal challenges:

Collins: Climate Dynamics 2002: critical dependence on initial conditions, is chaotic system

 \rightarrow still no predictability beyond seasonal scale

Introduction III where does the aerosol come from?





Method: take all AOD data Ny Alesund from 1998 – 2008 where air trajectories came from same origin in 850hPa, 700hPa, 500hPa (ECMWF) Remove all aerosol events of known origin (mainly forest fire / agricul. flaming)

Cluster 1 Eastern Arctic/ Siberia 2 Beaufortsea 3 Western Russia 4 North-East Canada Cluster 5 Europe 6 central Arctic 7 local 8 Greenland

Aerosol origin



Cluster 1 Eastern Arctic/ Siberia 2 Beaufortsea 3 Western Russia 4 North-East Canada 5 Europe 6 central Arctic 7 local 8 Greenland

Most aerosol from Beaufortsea, Eastern or central Arctic, least from Europe, Greenland Spring: annual max and max of variations between clusters Europe: rapid / (direct) transport assoziated with cloud formation (no photometer observation and/or wet scavening)

Where does aerosol come from? Measurements in central and Russian Arctic required! Follow pollution plumes with aircraft/satellites over several days

Radiative Forcing Components



Downwind of emission comparable to Greenhouse gases

Assumptions the similar? \rightarrow results similar

The headache caused by aerosol:



Challenges: Various types Non-uniform distribution Properties change with meteorology (hysteresis) RF also dependent on ground Chemically mixture: intern or extern

Remote sensing: Measures optical parameters, then estimates (overall) size distribution, (overall) shape and effective RI In-situ measurements: Get size distribution, chemical composition (almost) directly, then estimates scattering properties



In situ & remote sensing must not agree



Overview over aerosol effects



Twomey effect: smaller cloud particles have less absorption, more scattering (polluted clouds are whiter)

higher albedo, larger life-time




Why aerosol in the Arctic?

Arctic relatively clean : AOD(550nm): 0.05 – 0.1 But climatological sensitive environment ("polar amplification") Many different aerosol processes:

- in atmosphere (scattering, absorption: "dimming")
- on ground: decrease of albedo "darkening"



Polar amplification: (from IPCC) $\Delta T (2071 - 2100) -$ (1961 - 1990) Reason: feedbacks (e.g. ice - albedo)

Challenges: Seasonality: albedo, solar incident angle Aerosol: (interannual) variability

LIDAR (LIght Detection and Ranging)

Active remote sensing: information on altitude by time delay and c



