

Composition and Properties of the Natural Aerosol over the Boreal and Tropical Forests, A Review Comparing the Tropical and the Boreal Forest.

Lead authors HC Hansson¹, Paulo Artaxo², , Meinrat O. Andreae³, Markku Kulmala³

¹ Department of Environmental Science, Stockholm University, Stockholm, Sweden

² Institute of Physics, University of Sao Paulo, Sao Paulo, Brazil

³ Max Planck Institute for Chemistry, Mainz, Germany

⁴ Institute for Atmospheric and Earth System Research INAR, University of Helsinki

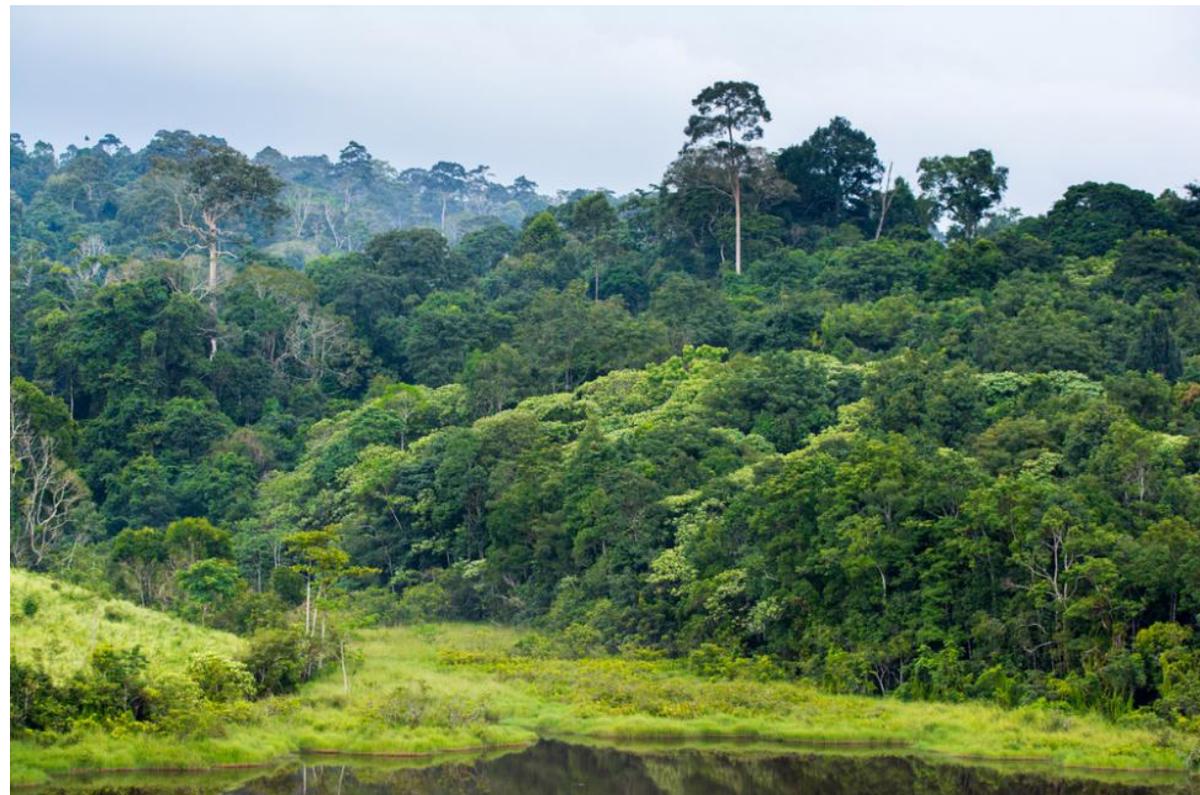
To be submitted to Tellus B

Based on a review with the following authors

Paulo Artaxo¹, Hans-Christen Hansson², Meinrat O. Andreae^{3, 27}, Jaana Bäck⁴, Eliane Gomes Alves⁵, Henrique M. J. Barbosa¹, Frida Bender⁶, Efstratios Bourtsoukidis⁷, Samara Carbone⁸, Jinshu Chi⁹, Stefano Decesari²⁸, Viviane R. Després¹⁰, Florian Ditas¹¹, Ekaterina Ezhova¹², Sandro Fuzzi²⁸, Niles J. Hasselquist⁹, Jost Heintzenberg¹³, Bruna A. Holanda¹¹, Alex Guenther¹⁴, Hannele Hakola²², Liine Heikkinen¹², Veli-Matti Kerminen¹², Jenni Kontkanen¹², Radovan Krejci², Markku Kulmala¹², Jost Lavric⁵, Gerrit de Leeuw²¹, Katrianne Lehtipalo^{12, 21}, Luiz Augusto T. Machado¹⁵, Gordon McFiggans¹⁶, Marco A. M. Franco¹, Claudia Mohr², William Morgan¹⁶, Mats B. Nilsson⁹, Matthias Peichl⁹, Tuukka Petäjä¹², Maria Praß¹¹, Christopher Pöhlker¹¹, Mira L. Pöhlker¹¹, Ulrich Pöschl¹¹, Celso Von Randow¹⁵, Ilona Riipinen², Janne Rinne¹⁷, Luciana V. Rizzo¹⁸, Daniel Rosenfeld¹⁹, Maria A. F. Silva Dias²⁰, Larisa Sogacheva²¹, Philip Stier²², Erik Swietlicki²³, Matthias Sörgel³, Peter Tunved², Aki Virkkula^{12, 21}, Jian Wang²⁴, Bettina Weber^{11, 30}, Ana Maria Yáñez-Serrano^{25, 26}, Paul Zieger², Eugene Mikhailov^{29, 3}, James Smith¹⁴, Jürgen **Kesselmeier**¹¹.

¹ Institute of Physics, University of Sao Paulo, Sao Paulo, Brazil, ² Dep of Environmental Science & Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden, ³ Max Planck Institute for Chemistry, Mainz, Germany, ⁴ Institute for Atmospheric and Earth System Research INAR / Forest sciences, Faculty of Agriculture and Forestry, University of Helsinki, Finland, ⁵ Department of Biogeochemical Processes, Max Planck Institute for Biogeochemistry, Jena, Germany, ⁶ Department of Meteorology and Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden, ⁷ Department of Atmospheric Chemistry, Max Planck Institute of Chemistry, Mainz, Germany, ⁸ Universidade de Uberlândia, Uberlândia, Brazil, ⁹ Department of Forest Ecology and Management, Swedish University of Agricultural Sciences, Umeå, Sweden, ¹⁰ Institute of Molecular Physiology, Johannes Gutenberg University, Mainz, Germany, ¹¹ Department of Multiphase Chemistry, Max Planck Institute for Chemistry, Mainz, Germany, ¹² Institute for Atmospheric and Earth System Research/Physics, Faculty of Science, University of Helsinki, Helsinki, Finland, ¹³ Leibniz-Institute for Tropospheric Research, Leipzig, Germany, ¹⁴ University of California, Irvine, USA, ¹⁵ Instituto Nacional de Pesquisas Espaciais, INPE, São José dos Campos, Brazil, ¹⁶ Atmospheric and Environmental Sciences, University of Manchester, Manchester, UK, ¹⁷ Department of Physical Geography and Ecosystem Sciences, Lund University, Sweden, ¹⁸ Federal University of São Paulo, Brazil, ¹⁹ Institute of Earth Sciences, The Hebrew University of Jerusalem, Israel, ²⁰ Department of Atmospheric Sciences, IAG-USP, University of São Paulo, Brazil., ²¹ Finnish Meteorological Institute, Climate Research Programme, Helsinki, Finland, ²² University of Oxford, United Kingdom, ²³ Department of Physics, Lund University, Sweden, ²⁴ Center for Aerosol Science and Engineering, Department of Energy, Environmental and Chemical Engineering, Washington University in St. Louis, St. Louis, USA., ²⁵ Center for Ecological Research and Forestry Applications (CREAF), Bellaterra, Spain, ²⁶ Global Ecology Unit, CREAM-CSIC-UAB, Bellaterra, Spain, ²⁷ Scripps Institution of Oceanography, University of California San Diego, La Jolla, USA, ²⁸ Institute of Atmospheric Sciences and Climate, CNR, Bologna, Italy, ²⁹ St. Petersburg State University, 7/9 Universitetskaya nab, St. Petersburg, 199034, Russia., ³⁰ Department of Biology, University of Graz, Graz, Austria

- *Compare how two major forest types interacts with the atmosphere and climate.*



- *Explore how these forests ecosystems affect and are affected by atmospheric processes.*
- *Focus on the "natural" systems and their interaction.*

Alarming findings:

- **Climate change, temperature and precipitation changes are inducing major changes in the ecosystem and thus all interaction with the atmosphere.**
- **The tropical forest is most likely in a transition phase strongly affecting its interaction with the atmosphere. It is no longer a carbon sink and decreased evapotranspiration reduce precipitation down wind.**
- **The continental boreal forest experience increasing occurrence of fires due to dryer and hotter summers.**

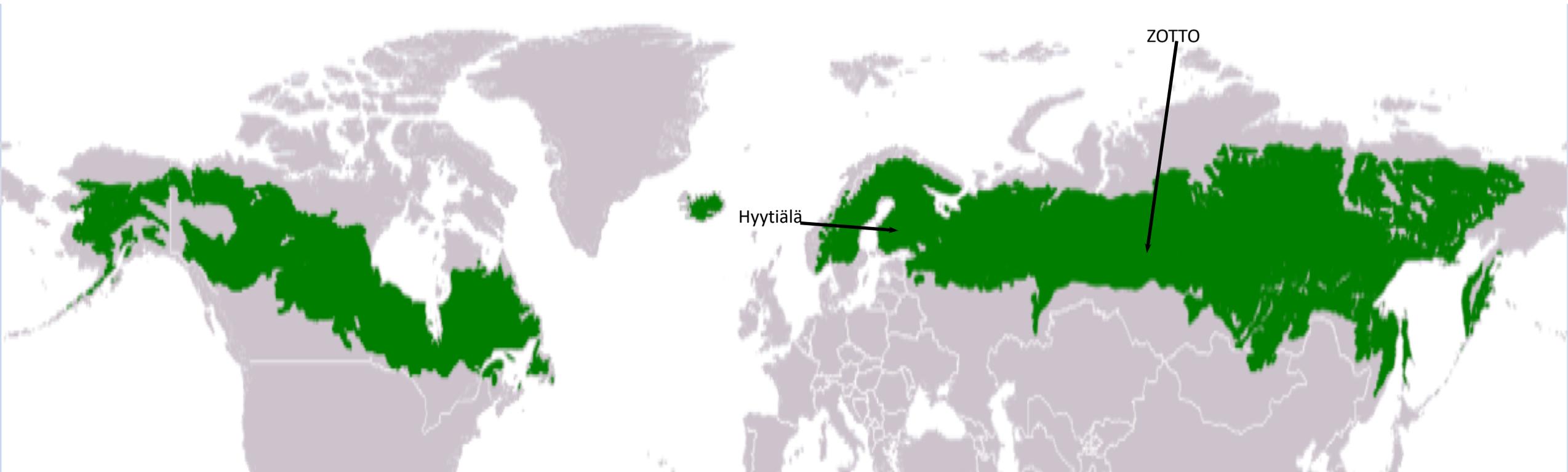
Key findings on similarities and differences on aerosols

- Continental boreal and Amazon tropical forest similar on
 - Particle chemistry, organic dominates
 - Size distributions, nucleation in boundary layer very rare
 - Biomass burning is the major source during warm and dry periods
 - Hygroscopic and optical properties during clean conditions
- Continental boreal and Amazon tropical forest differ
 - ZOTTO 2-3 higher SO₂ and BC background concentrations
 - ZOTTO monoterpene and ATTO isoprene driven SOA formation
- Finnish boreal site Hyytiälä show higher background concentrations, chemistry and size, much more frequent nucleation than the continental boreal site ZOTTO which also has more undefined NPF.
- All sites depend on meteorology and climate. More data needed!!!!

General findings:

- **The understanding of land-ecosystem-atmosphere has increased considerably lately, but still** on a global scale the knowledge is only in many parts **rudimentary concerning emissions, atmospheric chemistry, particle formation and effects on clouds and climate**. The feedback processes are mostly hypothetical where many interfering processes are not well known.
- **The really remote continental boreal forest seems at the moment least studied** and where major ecosystem changes are expected to be large due to climate change.
- **Climate change, temperature and precipitation will induce major changes** in the ecosystem and with that in the interaction with the atmosphere. **The forest carbon sink might be gone before we know. Knowledge and measurements needed!!!**
- **More supersites are desperately needed!!!** I wish for a CaTTO, The Canadian Tall Tower Observatory, placed as remote as ever possible in the Canadian Boreal Forest.

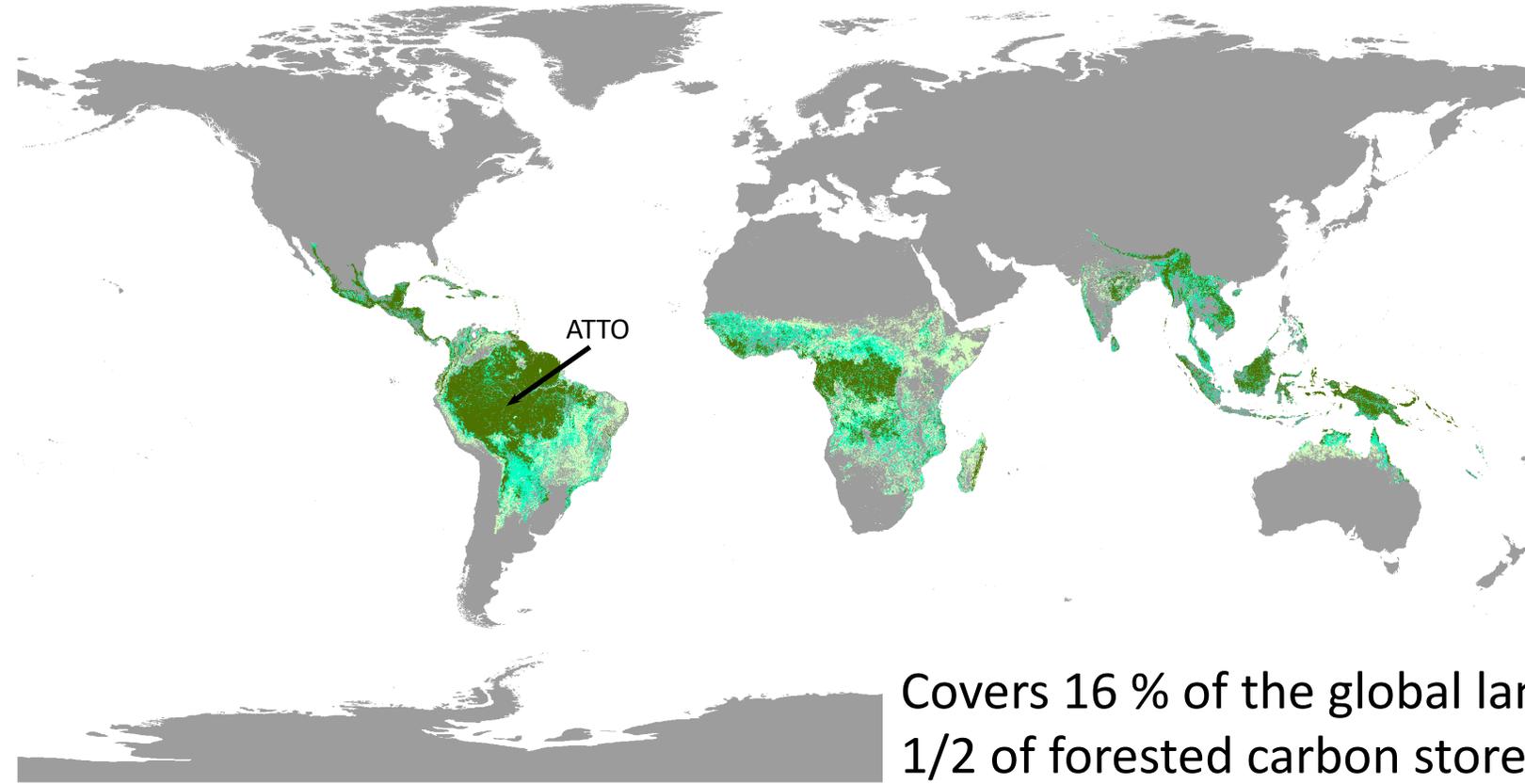
The boreal forest and important observation sites



Covers 10 % of the global land area, i.e. 30% of forested area, 1/3 of forested carbon store.

Winter and summer, large temp differences, about 0.5 (0.2-1) m annual precipitation with a maximum during summer

The tropical forest and the observation site

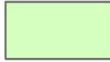


Tropical forest land based on UN FAO FRA 2000 dataset. Mark Marathon, 2017. Licensed under the [Creative Commons Attribution-Share Alike 4.0 International](https://creativecommons.org/licenses/by-sa/4.0/) license.

Covers 16 % of the global land area, i.e. 50 % of forested area, 1/2 of forested carbon store.

Dry and wet season, small temp difference, generally >2 m annual precipitation with a maximum during Nov – March (i.e. summer)

Tropical Forest Types

-  Tropical other wooded land
-  Tropical open/fragmented forest
-  Tropical closed forest

The tropical and boreal forests, $>\frac{3}{4}$ of all forested areas
The forested area is 40 milj km², 31% of global land.

ATTO, Amazonas



Hyytiälä (SMEAR2), Finland



ZOTTO, Krasnoyarsk

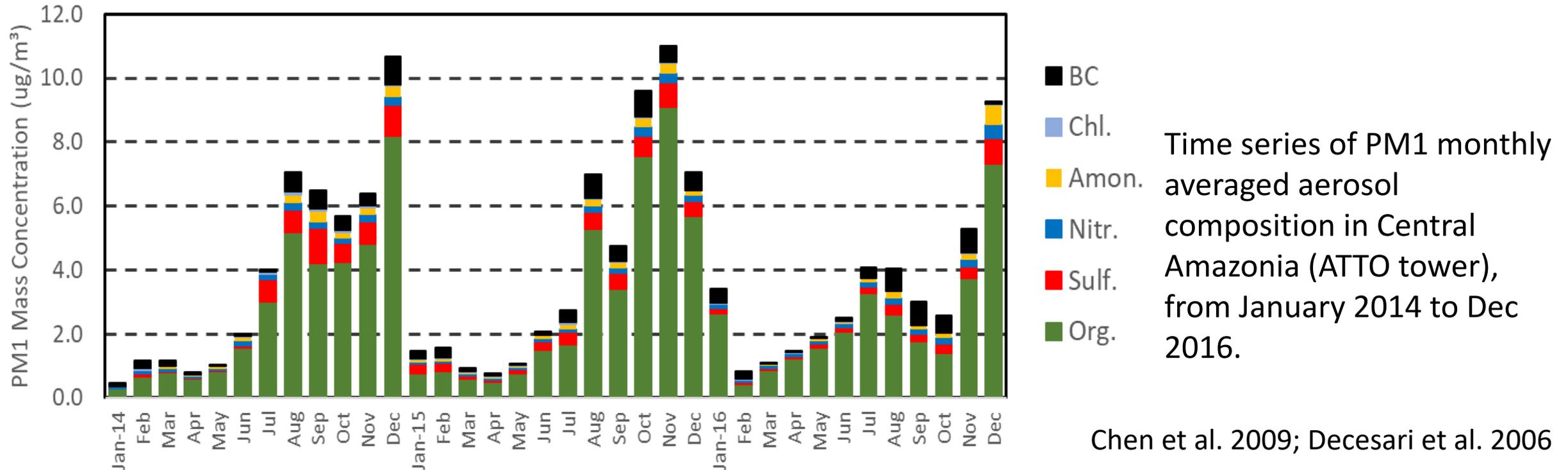


+ many large field campaigns

Observations; Atmospheric Chemistry

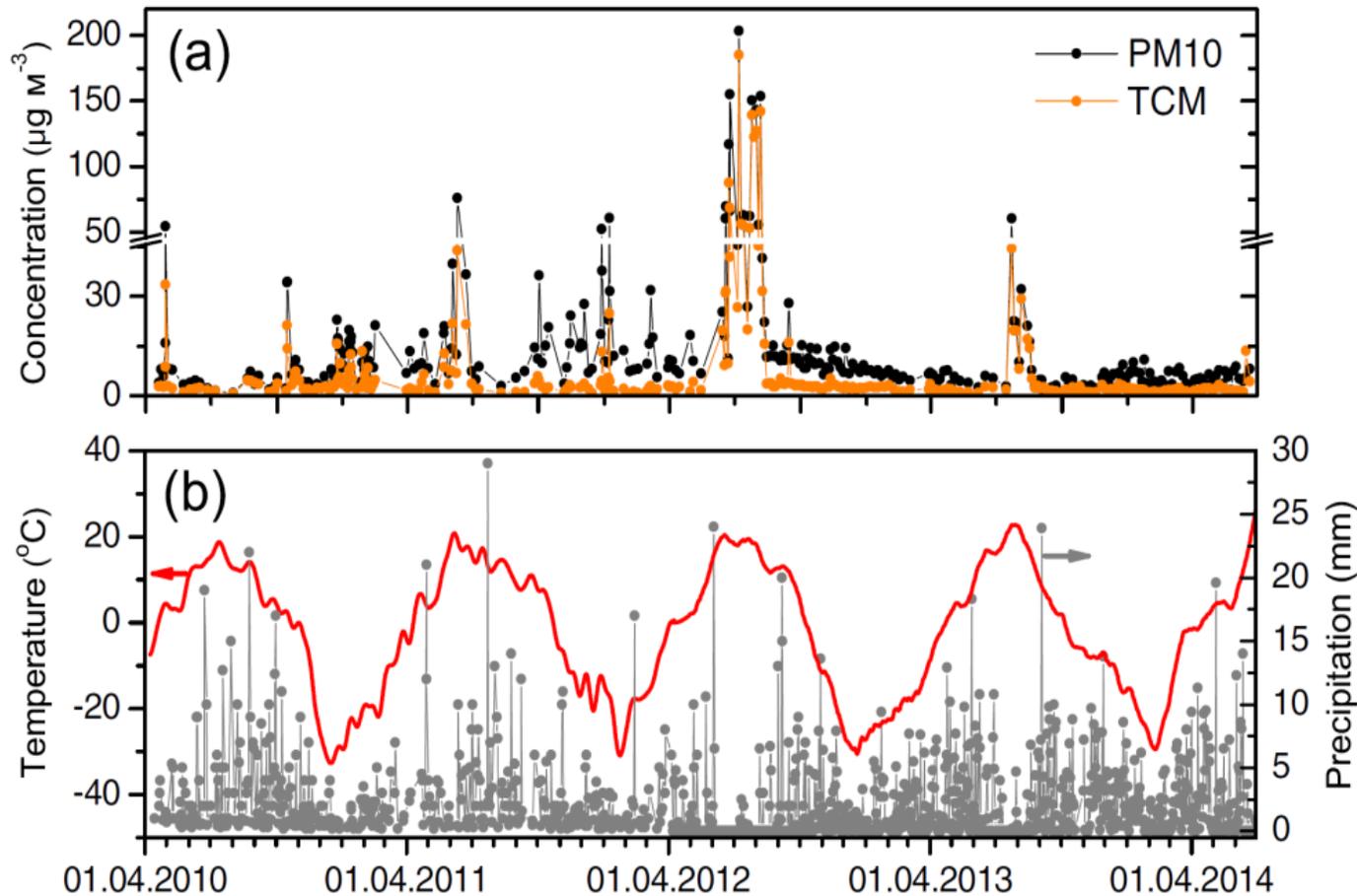
Organics dominate the aerosol in both the boreal and tropical forest

PM1 Aerosol Composition at ATTO ACSM 2014-2016



Chen et al. 2009; Decesari et al. 2006

Meteorology in both the boreal and tropical forest strongly affect the aerosol, concentrations and properties.

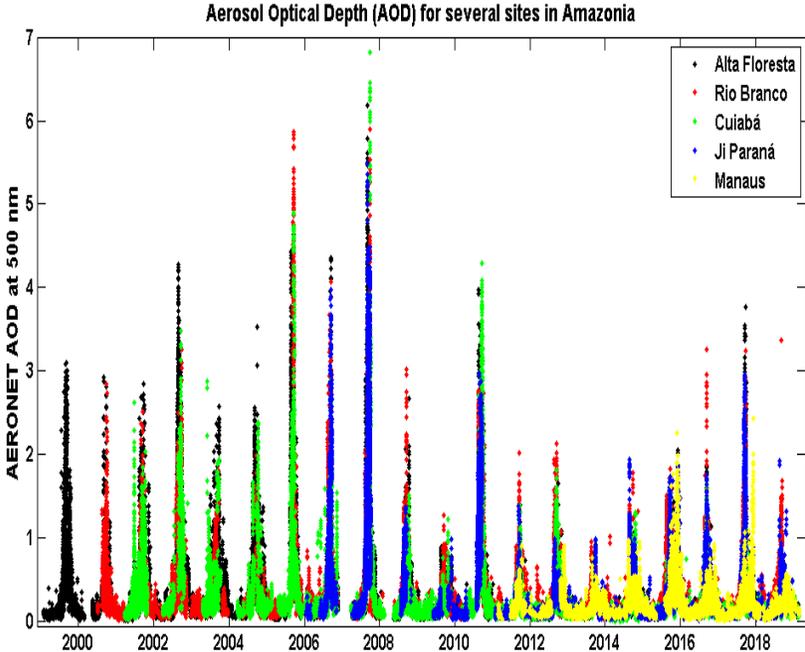


Seasonal and interannual variations at ZOTTO in the aerosol particulate matter (PM10) and TCM concentrations (a) and of daily averaged meteorological parameters (b): temperature – red line; precipitations – grey line and symbols.

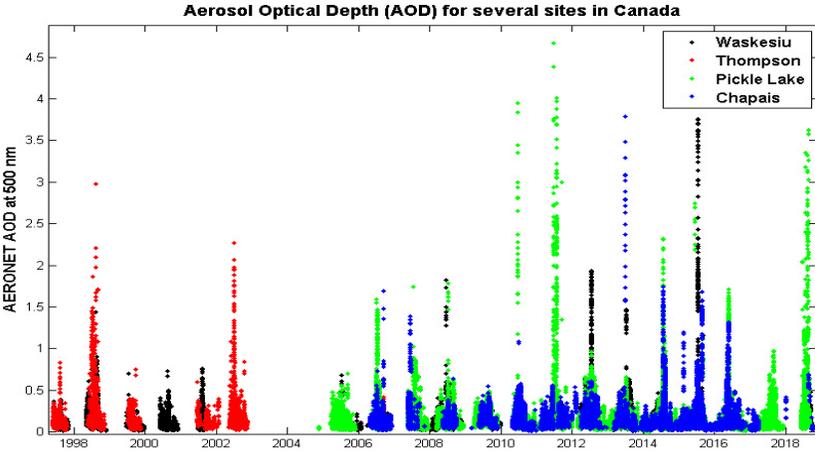
Mikhailov et al., 2017

Biomass burning are the dominating source in both the boreal and tropical forest during the summer (May – September) and dry periods.

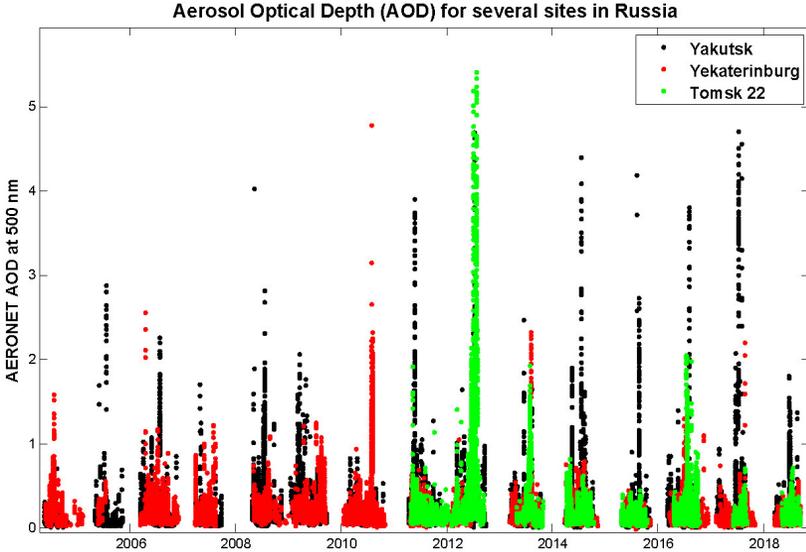
Amazonas



Canada

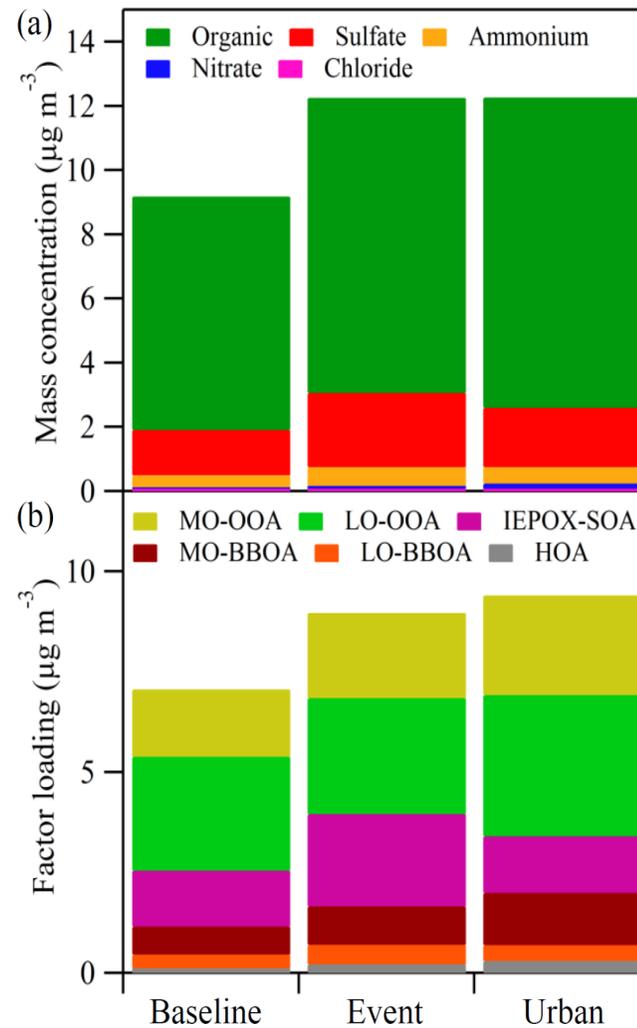
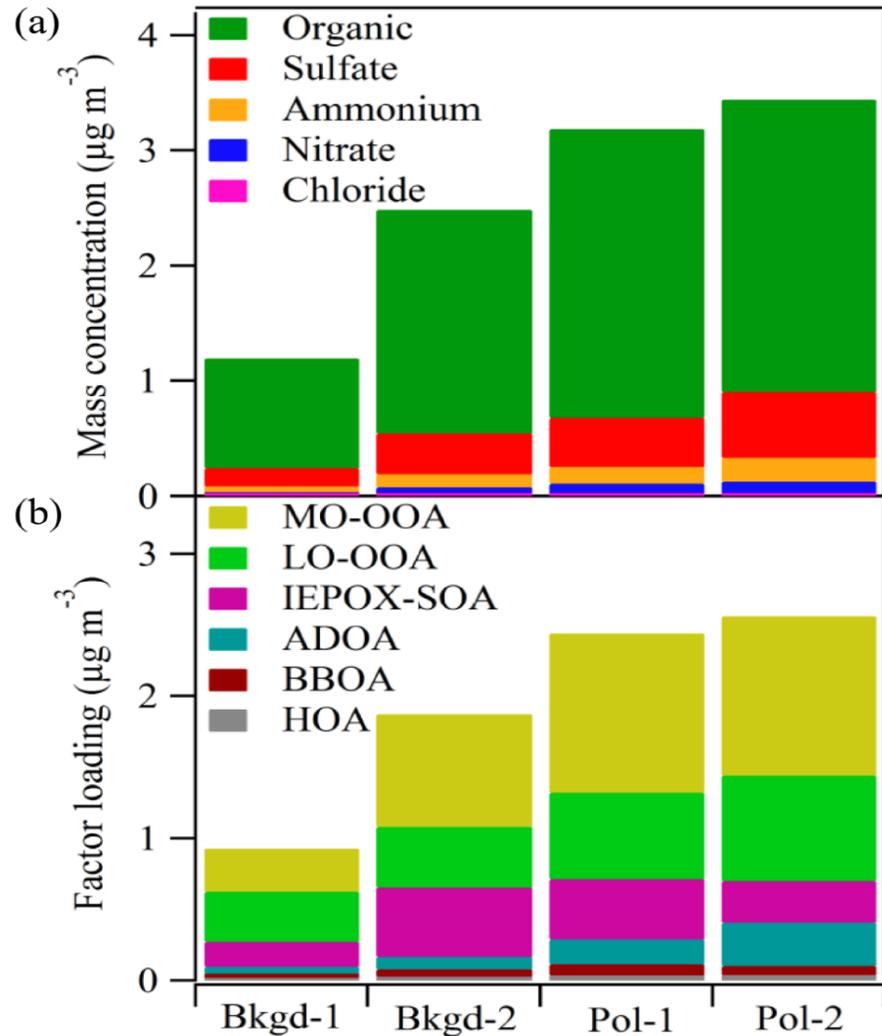


Russia



Groundbased AOD measurements from three major forested areas

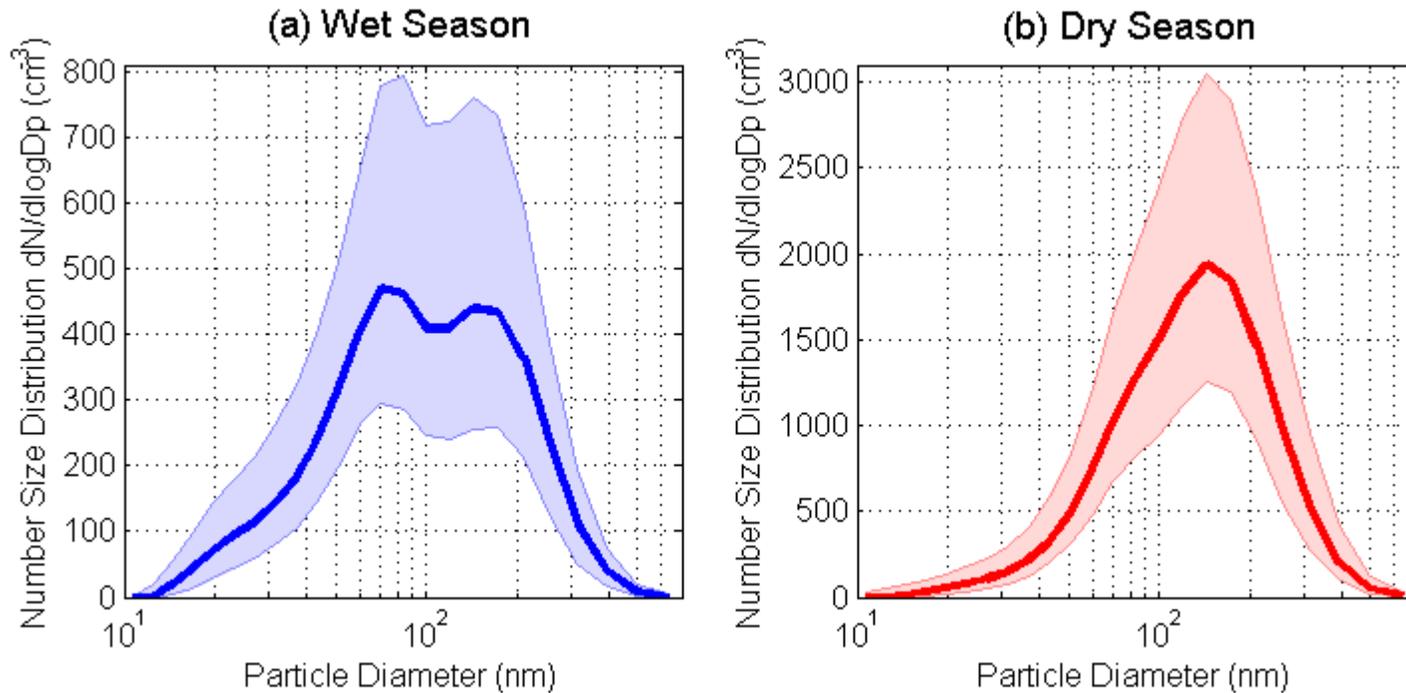
Urban emissions boost natural atmospheric chemistry over the Amazonas



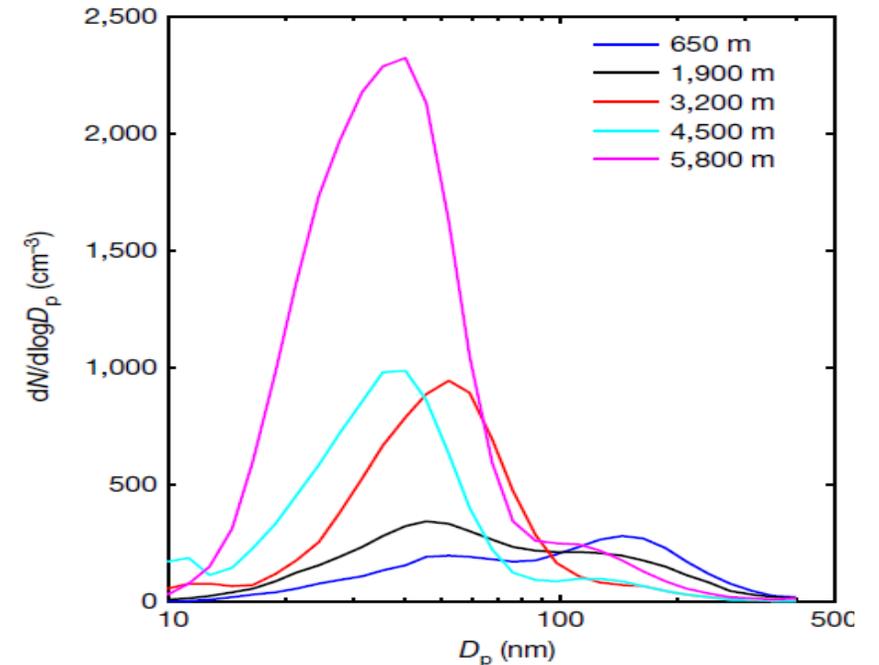
Increase in SOA formation from background to polluted conditions for the wet (left) and dry (right) seasons in Central Amazonia. In the wet season (left) polluted conditions increase aerosol mass from 1.2 to 3.3 $\mu\text{g}/\text{m}^3$. In the dry season, aerosol concentrations increased from 9 to 12 $\mu\text{g}/\text{m}^3$.

(Sá et al., 2018, 2019)

Tropical Aerosol Size Distribution shows seasonal and altitude dependence

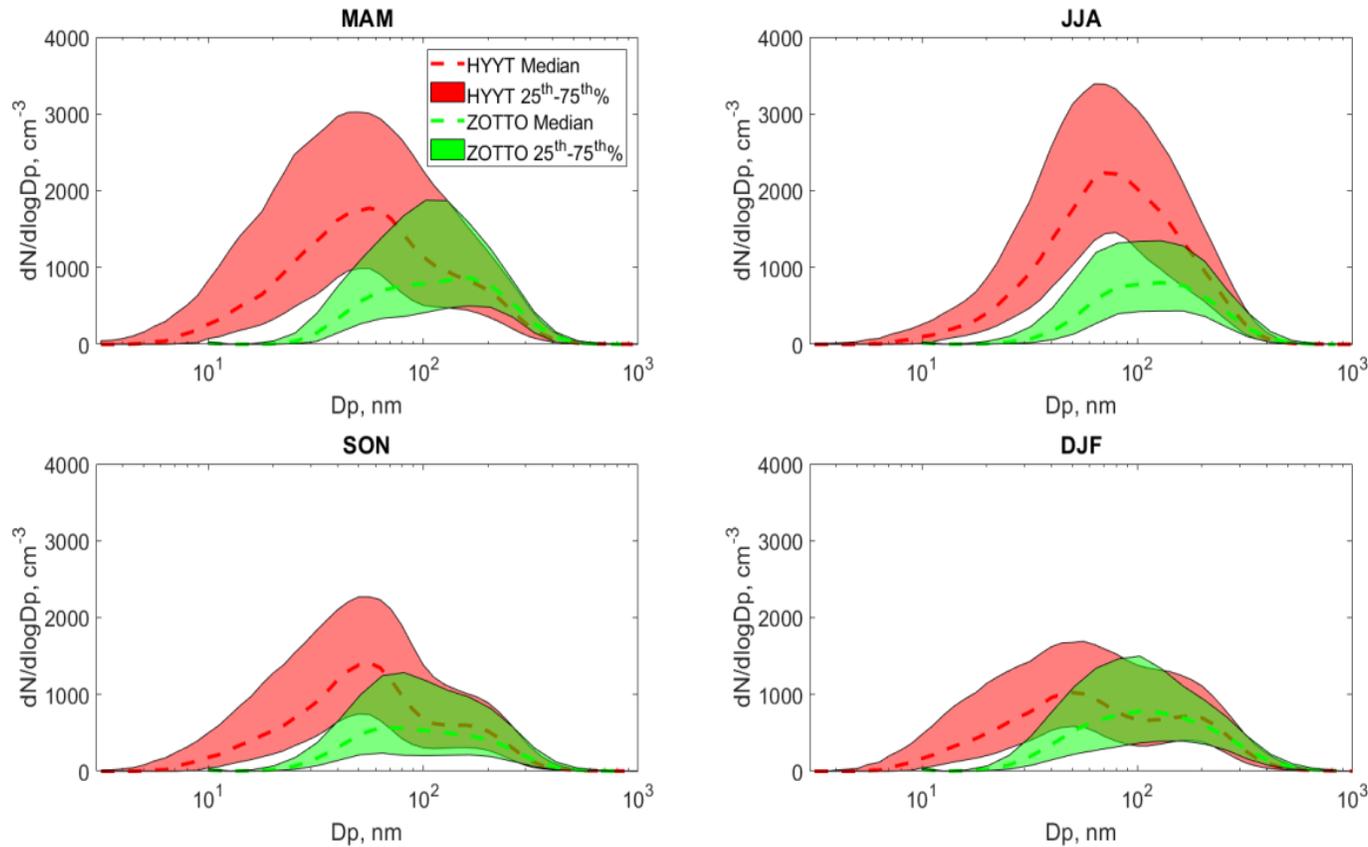


Median particle number size distributions in Central Amazonia between 2008 and 2014 during the (a) wet and (b) dry season. Low RH conditions (30-40%) and about 10 m above the canopy. Shadows represent the 25-75th percentile range (Rizzo et al., 2018).



Airborne observations of submicrometer particle size distributions above a forest area in Amazonia on 7 March 2014 (Wang et al., 2016).

Boreal Aerosol Size Distributions reveal large differences between Siberia and Fennoscandia.



New Particle Formation (NPF) is common at Hyytiälä but very rare at ZOTTO.

NPF at Hyytiälä is detected in clean air masses from the Arctic Sea or very North Atlantic. The aerosol is sampled in the forest canopy

Zotto is located 1500 km from the Siberian Coast in the central Siberia and the sampling at ZOTTO is at 320 meter altitude.

Seasonal median and 25th-75th percentile ranges of particle number size distributions observed at Hyytiälä and ZOTTO 2006-2011. MAM: March-May; JJA: June-August; SON: September-October; DJF: December-February.

Out of 752 days with valid measurements, only 11 were new particle formation events, 83 were undefined and 658 were non-event days.

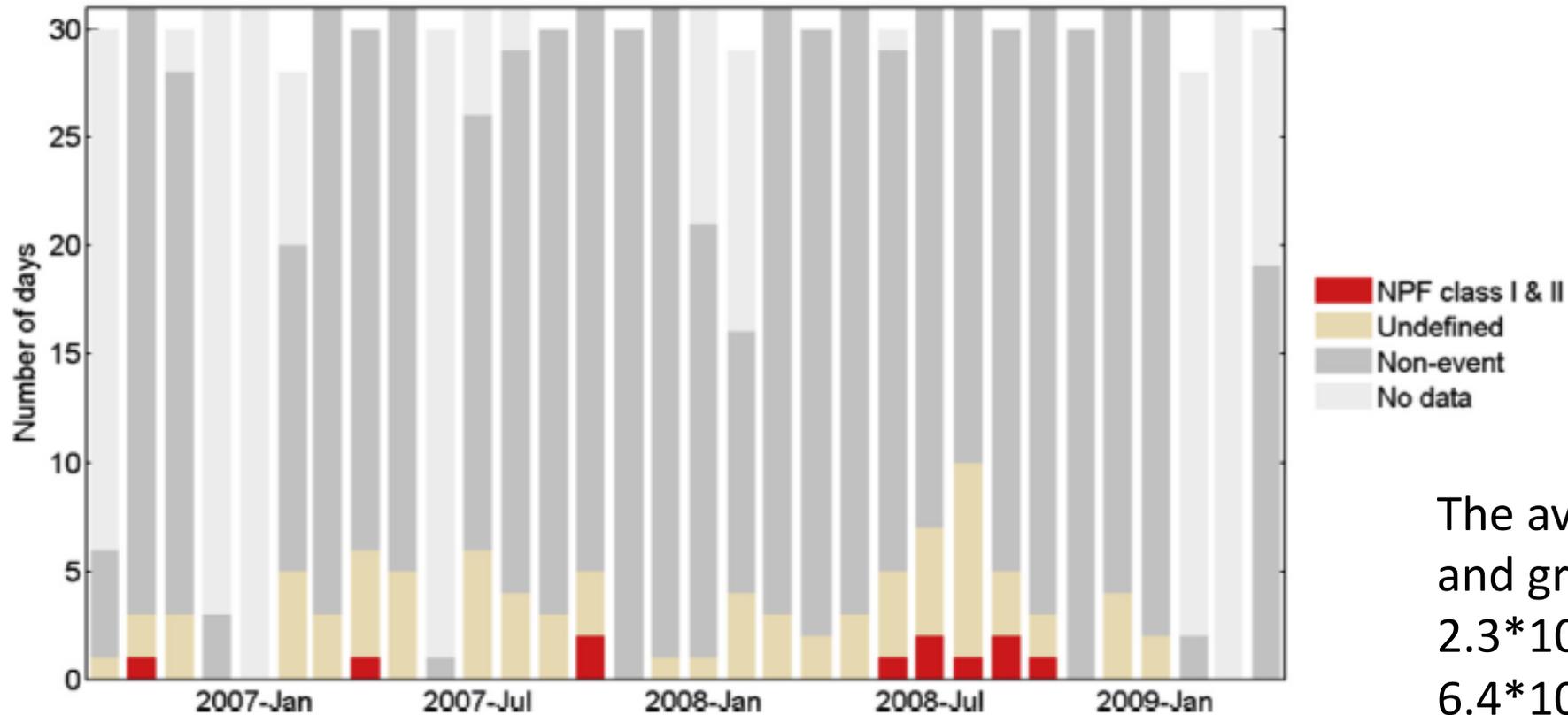
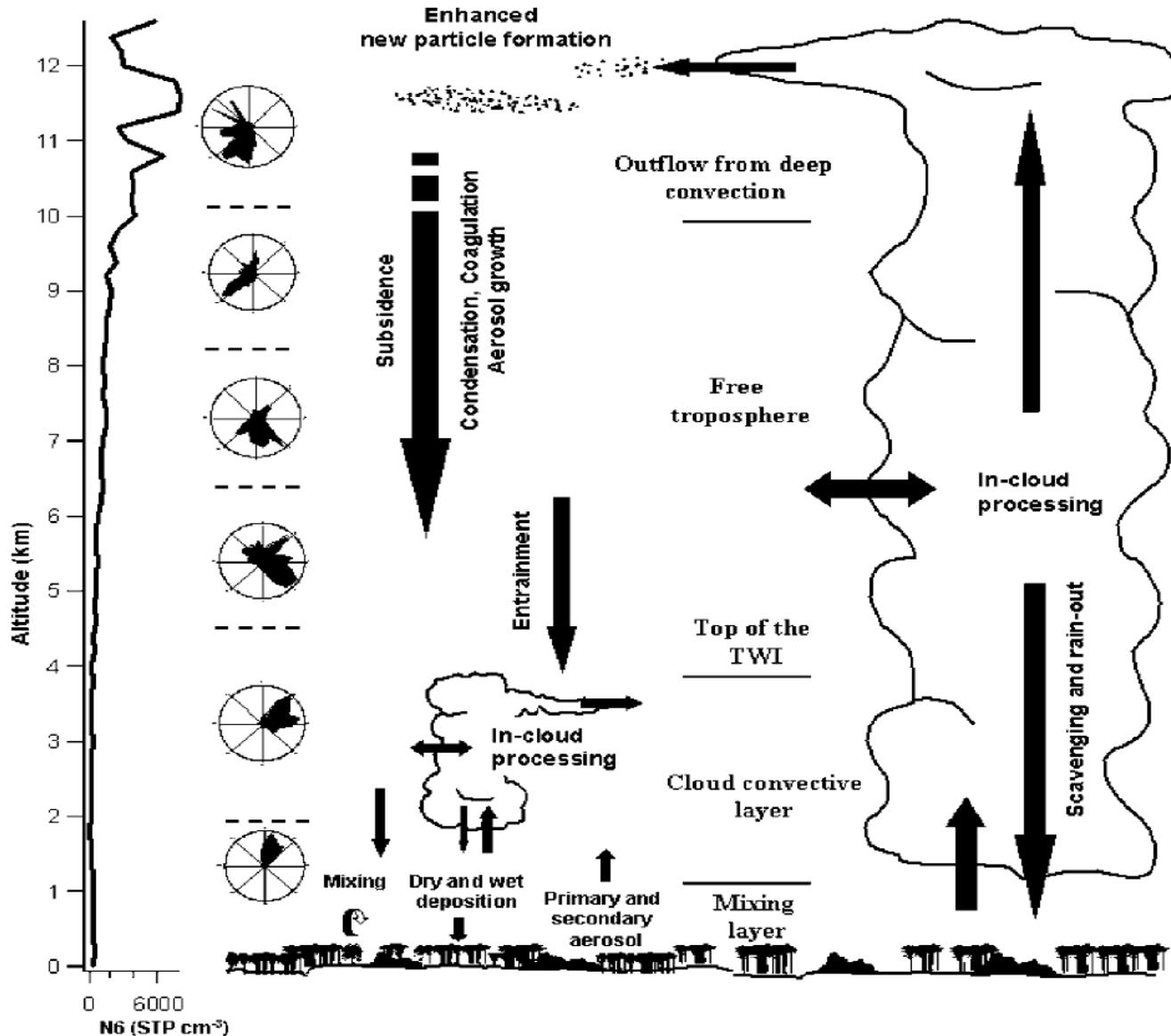


Fig. 1. Statistics of new particle formation (NPF) events in each month during the experiment period.

The average formation rate (J15) and growth rate at clear events are $2.3 \cdot 10^{-2} \text{ cm}^{-3} \text{ s}^{-1}$ ($5.2 \cdot 10^{-3} - 6.4 \cdot 10^{-2}$) and 2.4 nmh^{-1} (0.8 -4.6), respectively.

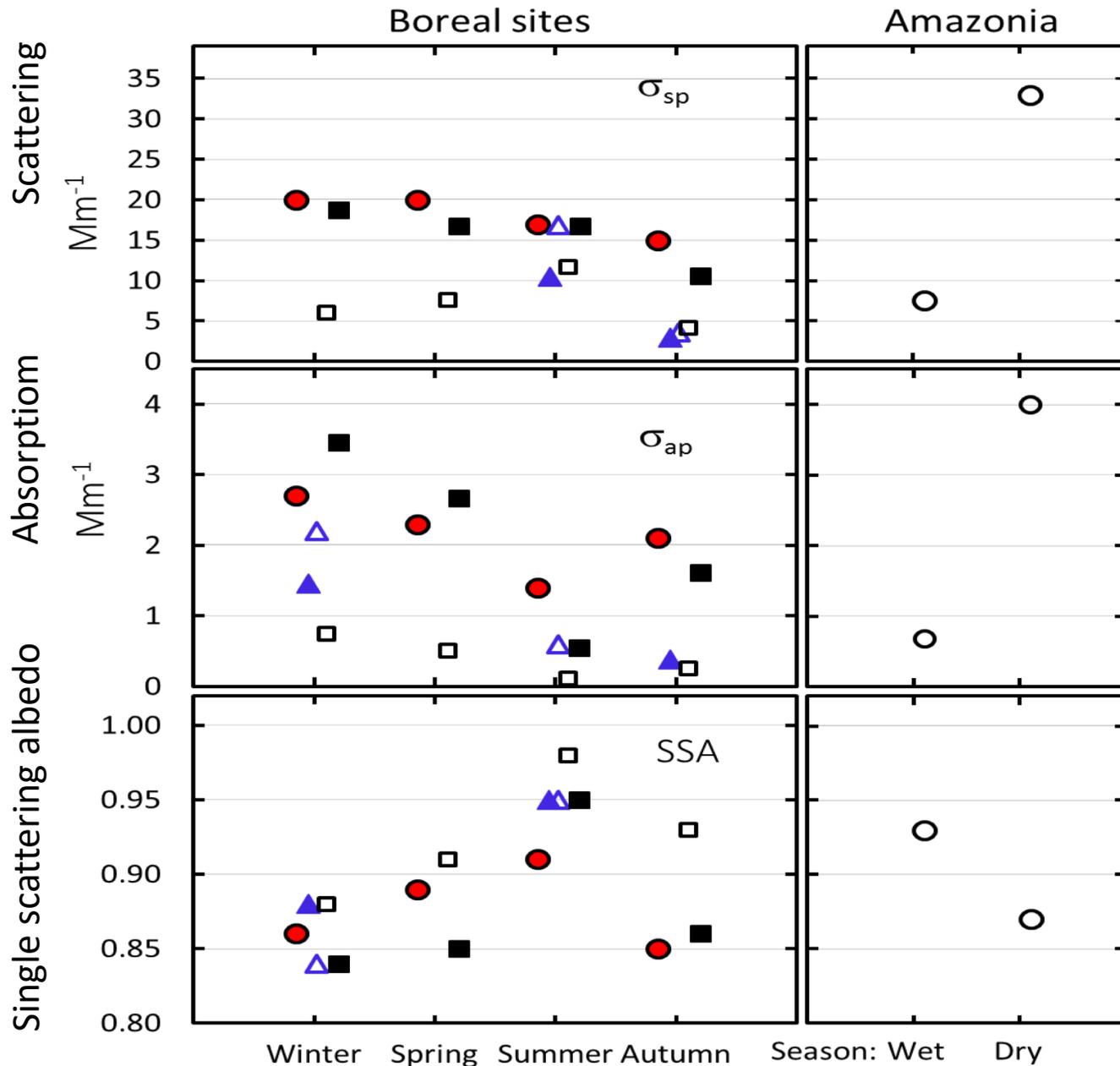
New Particle formation



Fine particles with enhanced ultrafine number concentrations in convective cloud outflow above Suriname in Northern Amazonin suggesting nucleation ([Krejci et al., 2003](#)).

Martin et al, 2010, Reviews of Geophysics “The original intent was to describe observations over Suriname, but the processes depicted are applicable to the wider Amazon Basin.

Aerosol Optical Properties



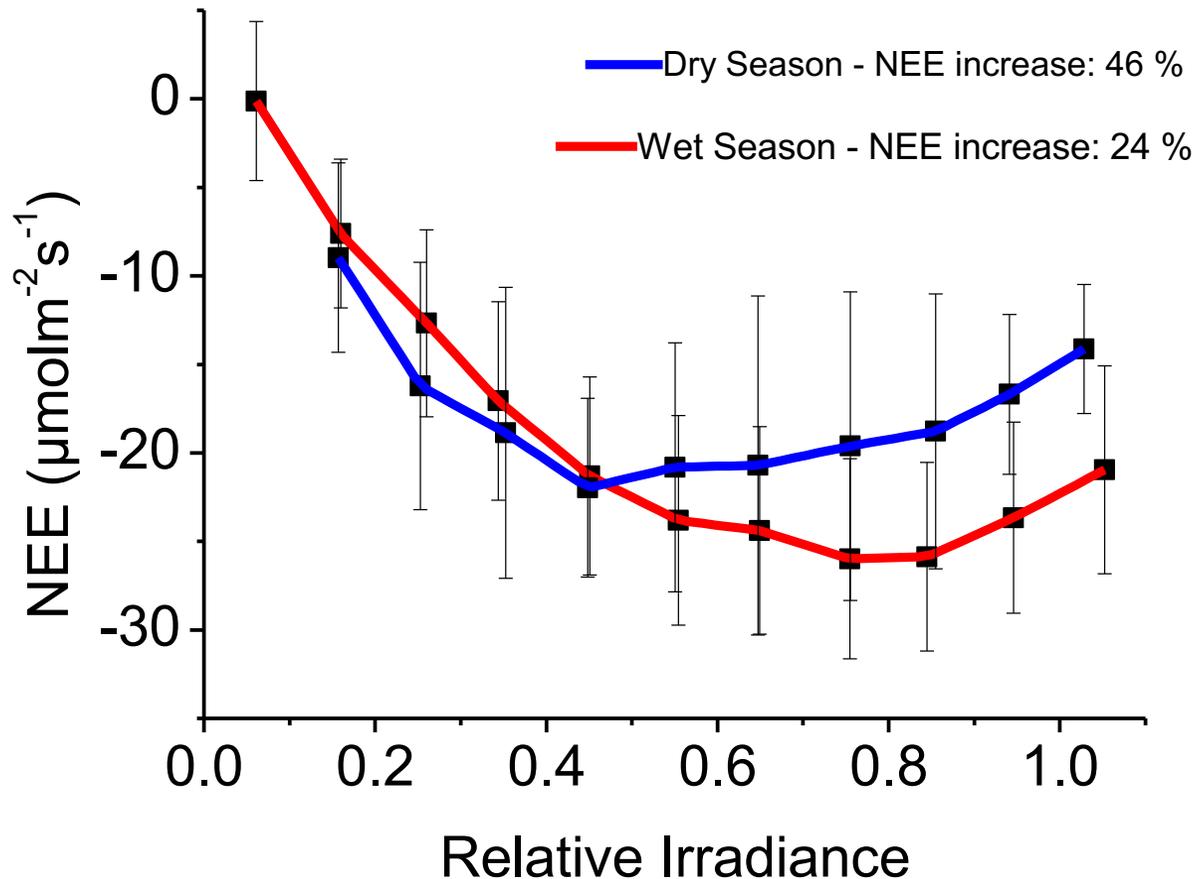
Scattering and absorption show
 ATTOwet = ZOTTO clean
 ATTOdry >> Hyytiälä = ZOTTO polluted

Data from Virkkula et al. (2011), Lihavainen et al. (2015), Chi et al. (2013), Saturno et al. (2018).

Seasonal variations of σ_{sp} , σ_{ap} and SSA at the boreal forest sites Hyytiälä, Pallas and ZOTTO and ATTO.

All σ_{sp} are at $\lambda=550$ nm, at Hyytiälä and Pallas also σ_{ap} and SSA are at $\lambda=550$ nm. At Zotto σ_{ap} and SSA are at $\lambda=574$ nm and at ATTO σ_{ap} and SSA are at $\lambda=637$ nm

Increased scattered light increase C uptake!

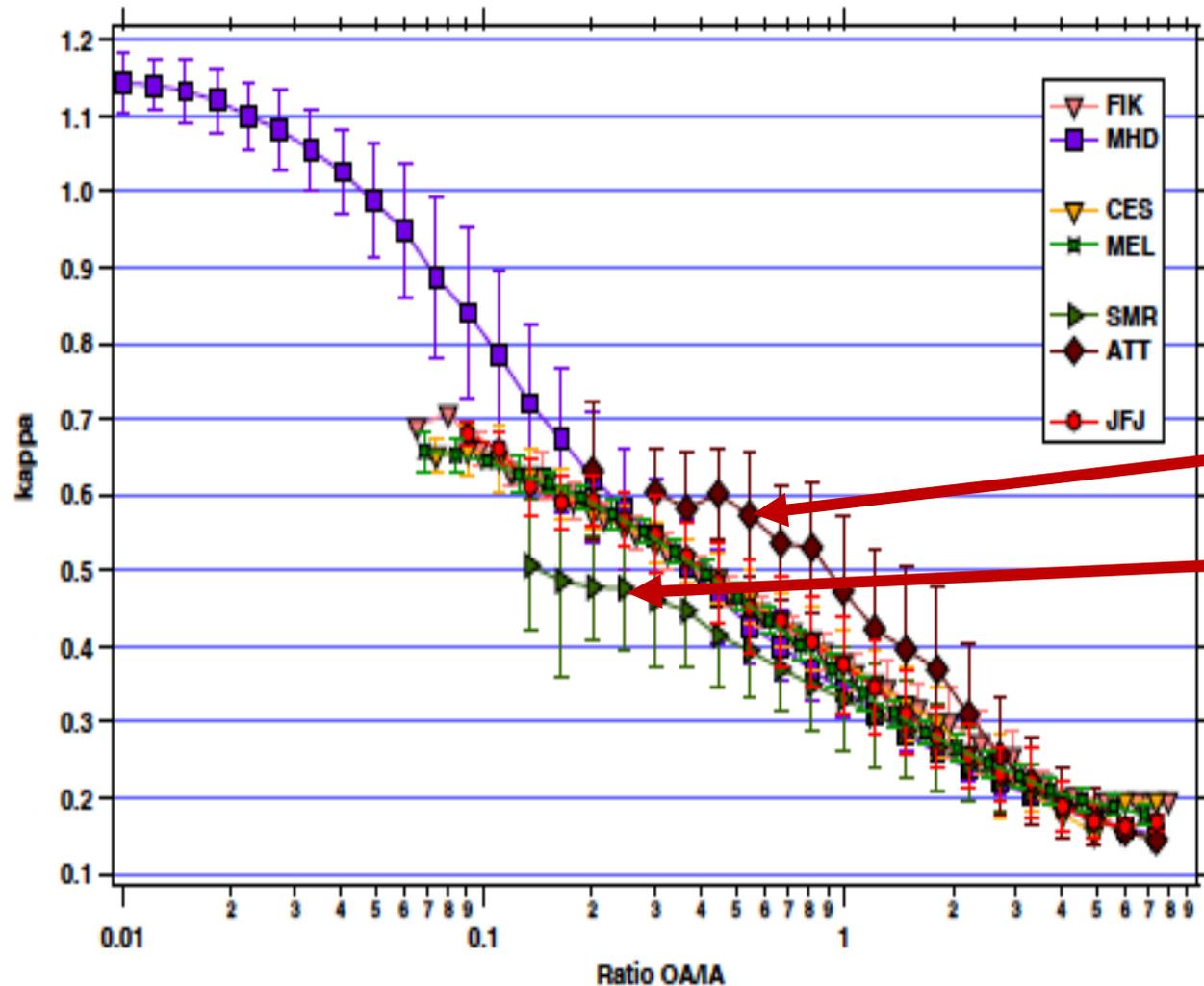


Effects of aerosols on carbon uptake expressed as Net Ecosystem Exchange (NEE) for dry and wet seasons in a LBA tower in Rondonia (Rebio Jaru), Amazonas.

Negative NEE = uptake of C

(Cirino et al., 2014)

Inorganic fraction determines hygroscopicity but ATTO more sensitive!



Relationship of the composition-derived hygroscopicity parameter κ to the binned and averaged ratio of organic (OA) to inorganic (IA) aerosol components for:

FIK = Finokalia, Crete;

MHD = Mace Head, Ireland;

ATT = ATTO tower, Amazon;

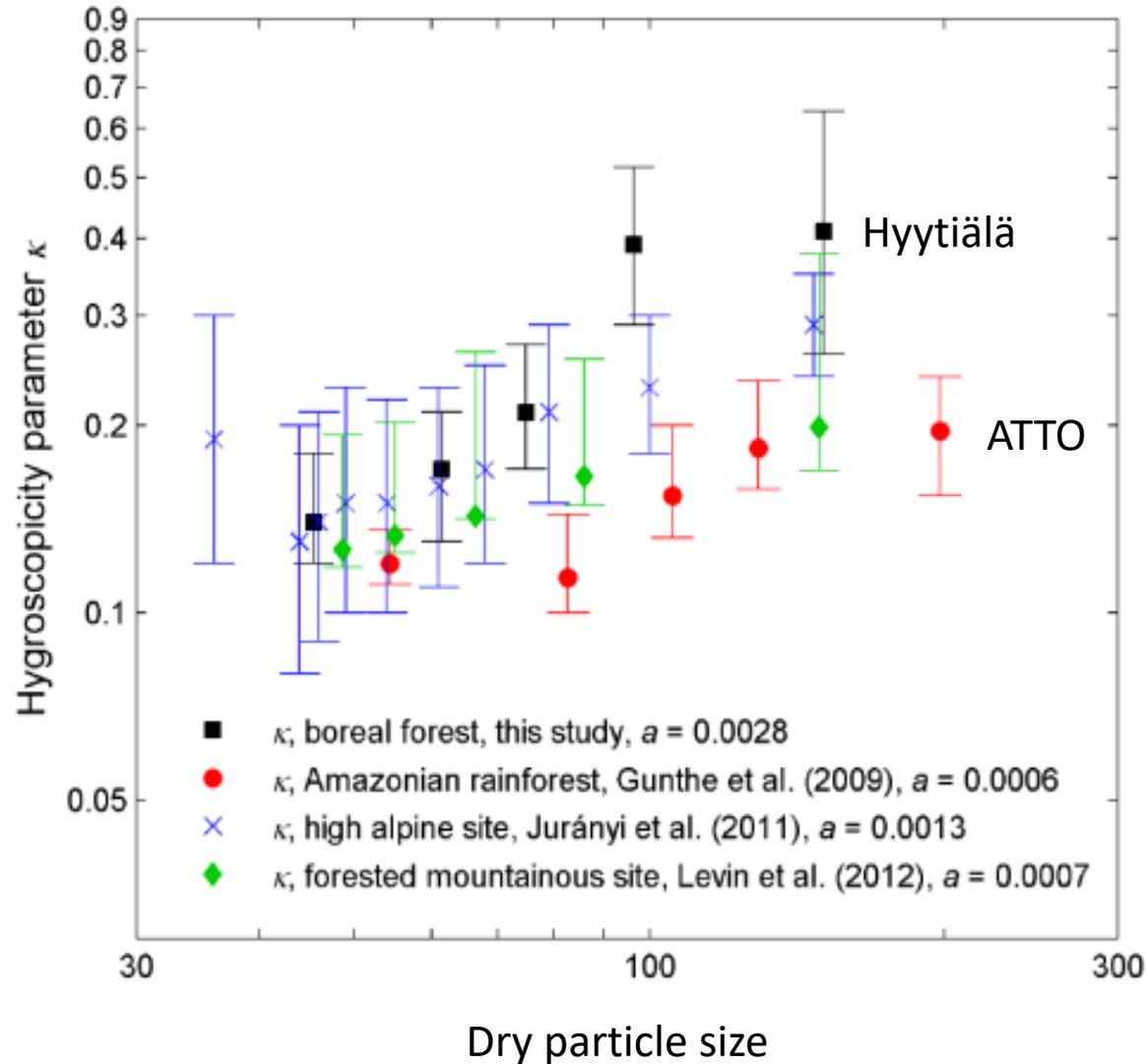
MEL = Melpitz, Germany;

SMR = Hyytiälä, Finland;

JFJ = Jungfrauoch, Switzerland.

Note that the asymptotic-like approach of the curves towards 0.1 is due to the assumption of $\kappa_{OA} = 0.1$.

Larger particles less hygroscopic in the tropical forest



Relationship between particle dry diameter and κ for boreal and tropical forests.

The median values are shown with error bars being 25th and 75th percentiles. Legend entries also indicate the slope of the linear regression $y = ax + b$ fit.

Figure from Paramonov et al., 2013.

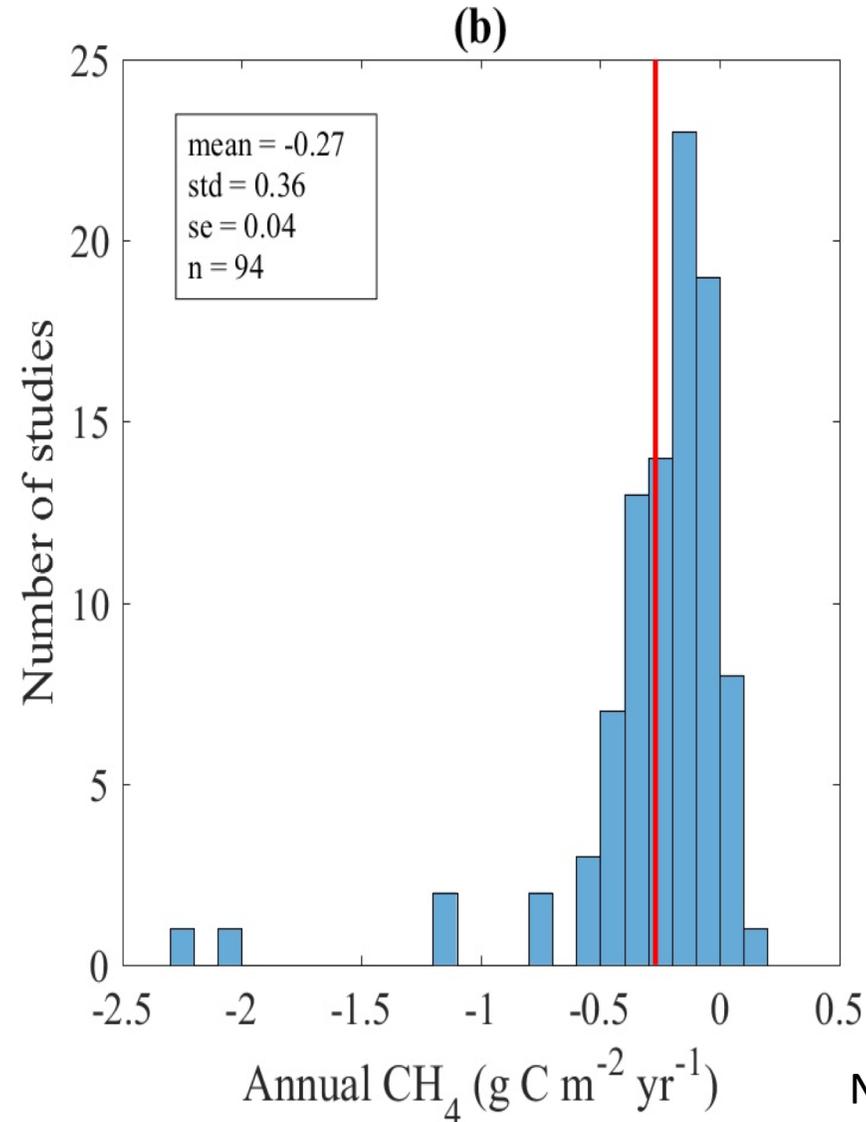
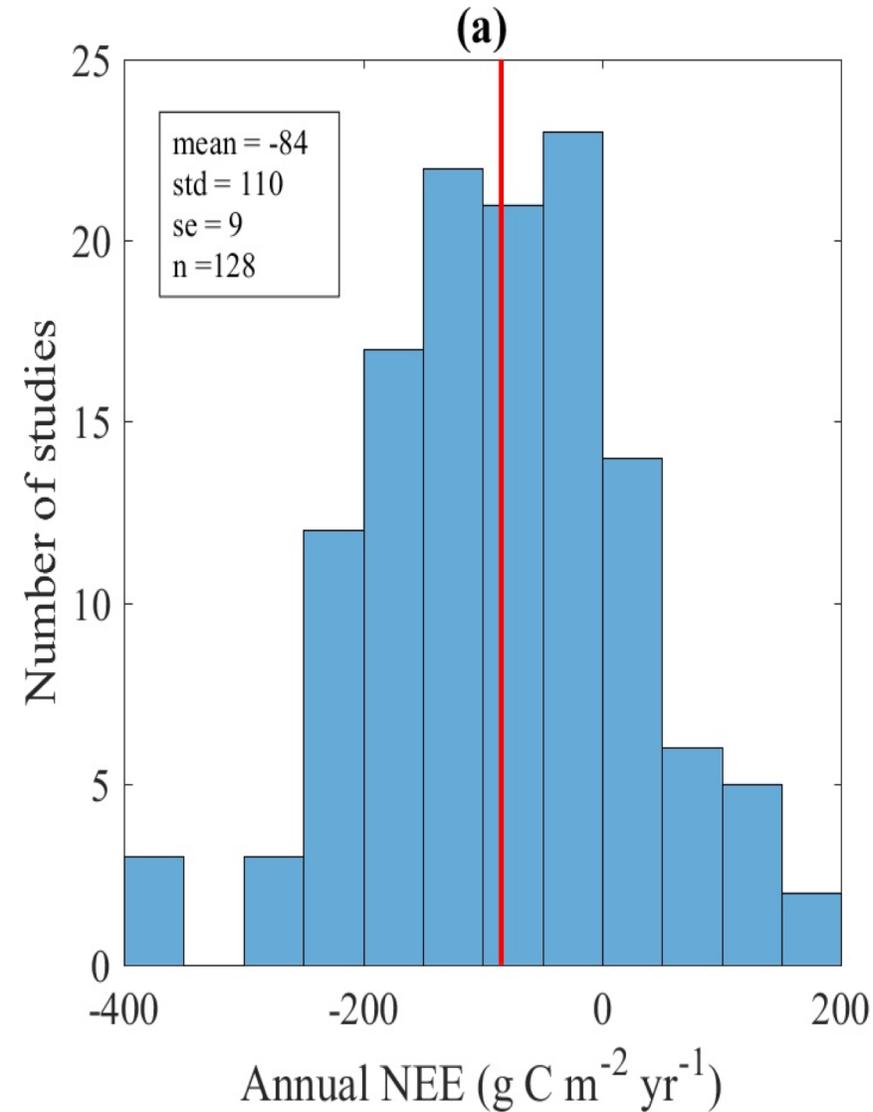
BVOC emissions strongly dependent on ecosystem type

- Major and clear difference is isoprene is the primary SOA precursor in the Amazonas and monoterpenes in the boreal forest.
- The BVOC emissions are highly sensitive to landuse change, climate change, and other disturbances but our limited understanding of the processes controlling the specific responses makes it difficult to quantitatively compare and contrast tropical and boreal ecosystems.

Similarities and differences

- ZOTTO and ATTO similar on
 - Particle chemistry, organic dominates
 - Size distributions, nucleation in boundary layer very rare
 - Biomass burning is the major source during warm and dry periods
 - Hygroscopic and optical properties during clean conditions
- ZOTTO and ATTO differ
 - ZOTTO 2-3 higher SO₂ and BC background concentrations
 - ZOTTO monoterpene and ATTO isoprene driven SOA formation
- Hyytiälä show higher background concentrations, chemistry and size, much more frequent nucleation. Zotto has more undefined NPF. Time perspective?
- All sites depend on meteorology and climate. More data needed!!!!

The ecosystem take up CO₂ and CH₄ in the boreal forests

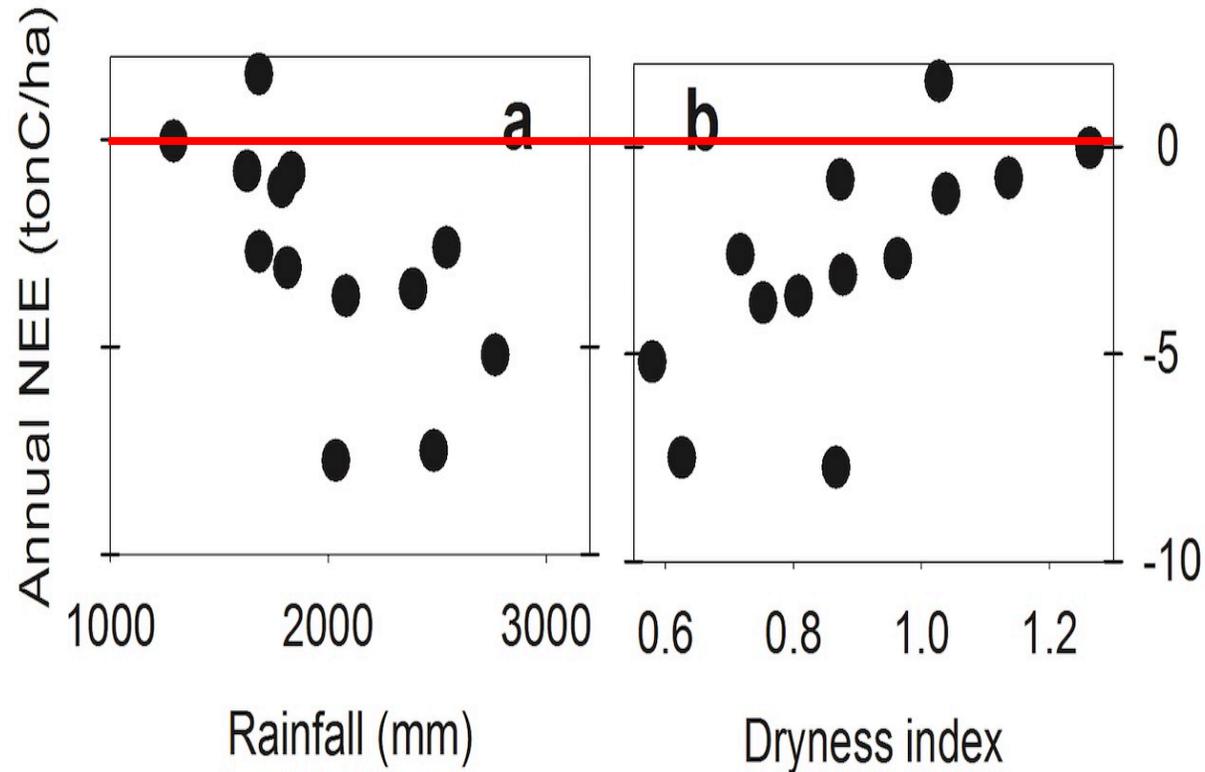


Histograms of published annual net ecosystem exchanges of CO₂ and CH₄ in boreal forests from 1990 to 2015.

Boreal forest takes up about 80 ton C / km² year

Ni and Groffman (2018).

Less rainfall and more dryness decrease C-uptake in the Amazon



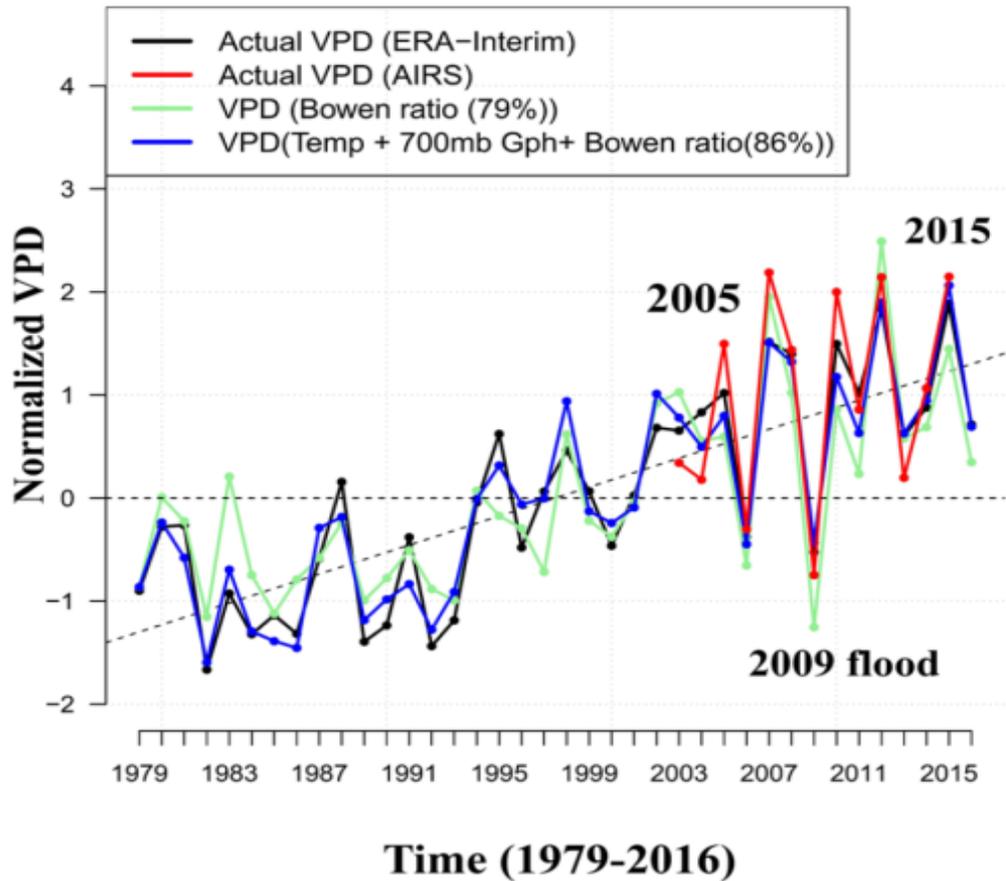
Annual Net Ecosystem Exchange of carbon measured in Amazon flux towers at pristine forests, in the period 1999-2006, compared to measured rainfall and dryness index ($D = R_n / \lambda P$; R=radiation, λ =latent heat of vaporization, P=precipitation). Adapted from Von Randow et al., 2013.

Measurements as ATTO show decrease from about 50 ton C uptake per km² and year **to zero uptake during the last years !!**

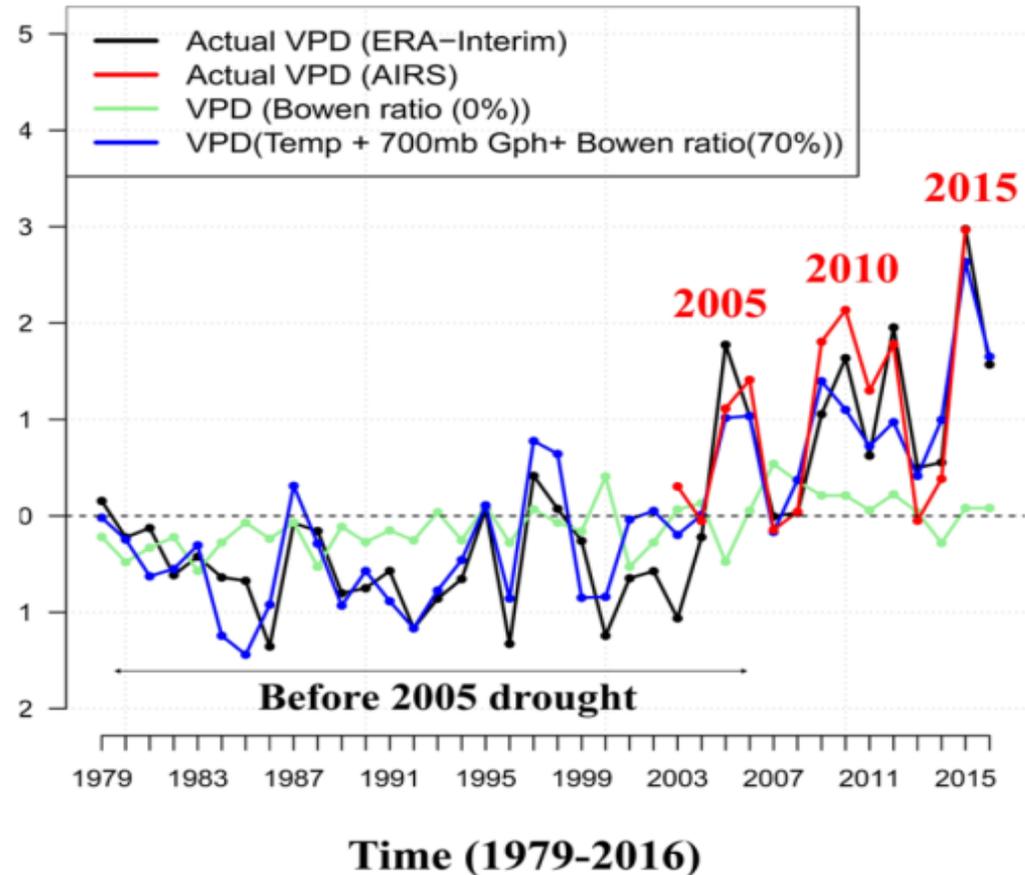
Major change in the hydrological cycle, The forest return less of the precipitation to the atmosphere!!

Figure show Increase in vapor pressure deficit (VPD) in Southeast and Northwest Amazonia from 1979 to 2016. From Barkhordarian et al., 2019.

d) Southeast Amazon (Green box)



e) Northwest Amazon (Blue box)



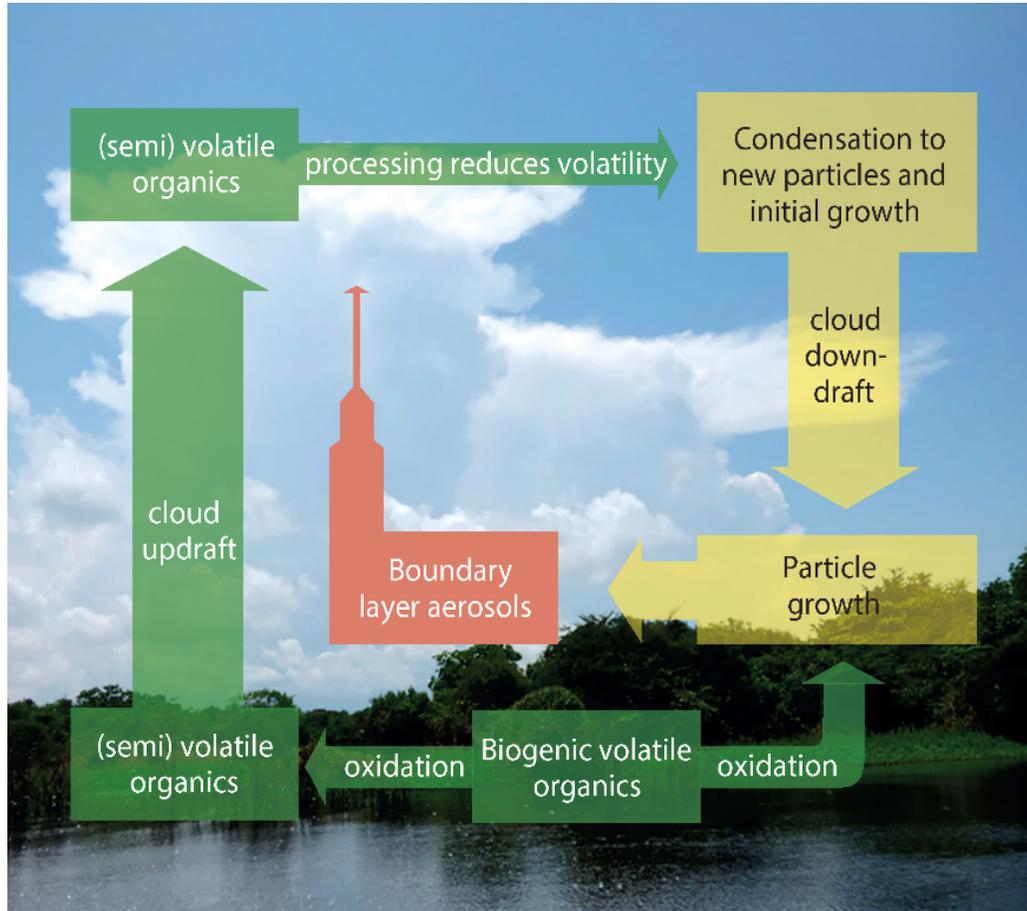
Land atmosphere exchange of CO₂, CH₄ and other trace gases are affected by climate change.

- According to Global Carbon Project the global forests take up 29% of anthropogenic CO₂ emissions, i.e. about 3.2 Gton C /y = 80 ton C /y and km²
- Forest Net Ecosystem Exchange is regulated by rainfall, and any changes in the hydrological cycle affects CO₂ exchange. Recent increase in extreme events are reducing the net uptake of CO₂ and evapotranspiration of H₂O in Amazonia.
- Tropical forests are the major source of N₂O, due to high nitrogen turnover. N₂O and CH₄ are emitted by tropical soils, and increase in global temperature can change these fluxes.
- Tropical flooded areas are a major source of CH₄, as well as boreal regions with permafrost.

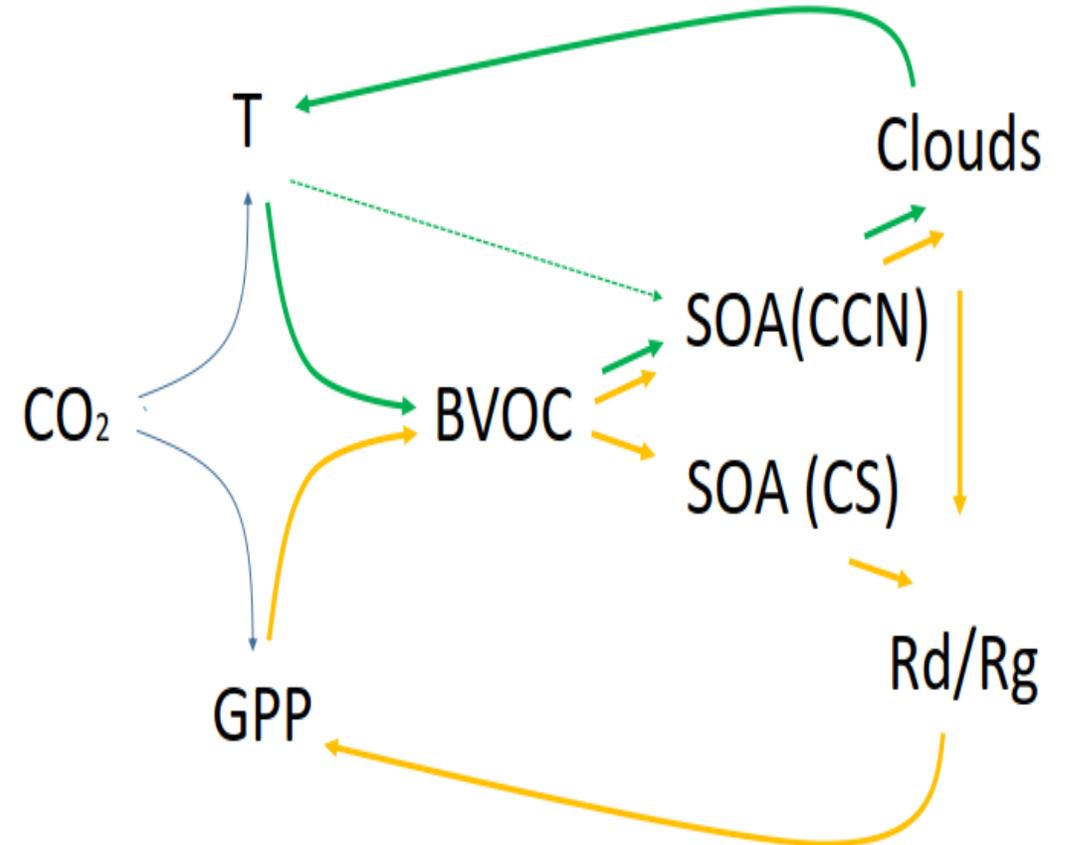
Land atmosphere exchange of CO₂, CH₄ and other trace gases are affected by climate change.

- CO and O₃ precursors from biomass burning are key to regulate oxidative processes in the atmosphere.
- Ozone damage to plants can reduce significantly the CO₂ uptake for tropical forests. OH reactivity is controlled by VOC emissions and reactions, and is key to atmospheric lifetime of key gases.
- NO_x regulates the pathway of VOCs oxidation.
- SO₂ emissions are important to NPF even at very low concentrations.
- **The effect on atmospheric composition, and thus feedback on climate, due to climate change is not known!**

Ecosystem feedback processes



Aerosol life cycle,
close interaction ecosystem – clouds
(Andreae et al, 2018) .



Ecosystem emission affect clouds and
radiation affecting the ecosystem (Kulmala
et al., 2013) .

AT LAST final conclusions

- **The understanding of land-ecosystem-atmosphere has increased considerably lately, but still** on a global scale the knowledge is only in many parts **rudimentary concerning emissions, atmospheric chemistry, particle formation and effects on clouds and climate**. The feedback processes are mostly hypothetical where many interfering processes are not well known.
- **The really remote continental boreal forest seems at the moment least studied** and where major ecosystem changes are expected to be large due to climate change.
- **Climate change, temperature and precipitation will induce major changes** in the ecosystem and with that in the interaction with the atmosphere. **The forest carbon sink might be gone before we know. Knowledge and measurements needed!!!**
- **More supersites are desperately needed!!!** I wish for a CaTTO, The Canadian Tall Tower Observatory, placed as remote as ever possible in the Canadian Boreal Forest.