MULTI-ANNUAL CHARACTERISATION OF PM_1 AEROSOL OPTICAL PROPERTIES AND SIZE DISTRIBUTION AT THE URBAN ATMOSPHERIC SITE ATOLL IN LILLE

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INTRODUCTION

Long-term studies of aerosol properties are essential for assessing air quality, public health impacts, and climate change mitigation efforts.
ATOLL data set offers a unique chance to retrieve variability regarding **aerosol optical** (climate change) and **physical** (climate change and health impact) **properties in urban environments** and their implications for urban residents and policymakers alike. The added value of our work is multiannual measurement and characterization of **optical properties in PM_1 fraction.**

RESULTS

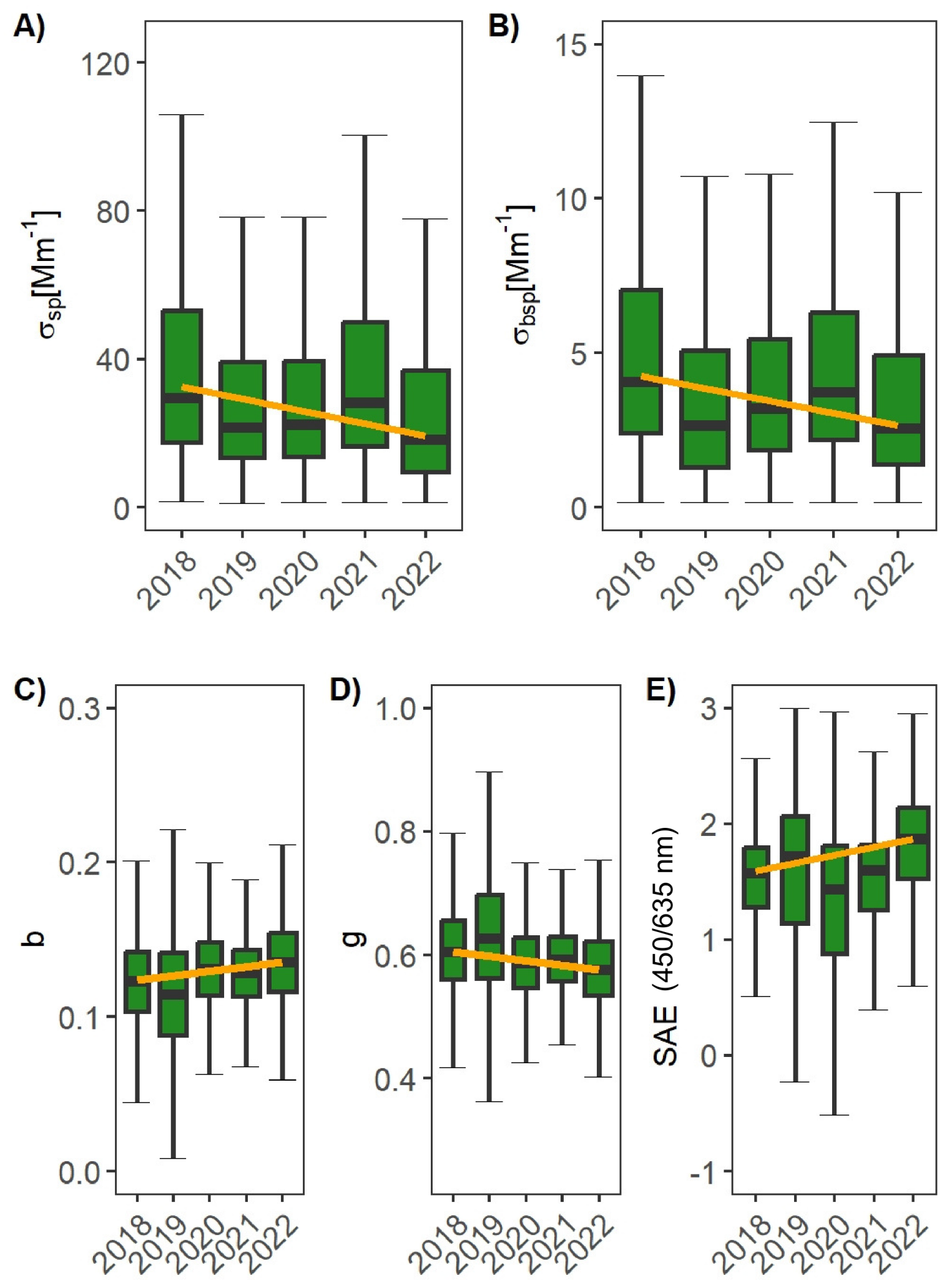


Figure 1. Multiannual median variations (525 nm) of scattering (σ_{sp}, **A**) and backscattering (σ_{bsp} , **B**) coefficients, backscattering ratio (*b*, **C**), asymmetry factor (*g* , **D**), scattering Ångström Exponent (*SAE*, **E**). The colored parts represent the 1st and 3rd quartiles. The whiskers extend to the minimum and maximum values within a range defined by 1.5 times the interquartile range. The orange lines show the trends. The proportional width of the boxplots shows the availability of data.

Table 1. Mann Kendall trend test results (tau estimating the strength of trend change, p-value showing the significance, α=0.05) for σ_{sp} and σ_{bsp} at 525 nm, *SAE (450/635 nm)* and σ_{FFap}. The trends marked in red are statistically significant.

Mann Kendall test tau (p value)	Yearly	Winter	Spring	Summer	Fall
σ_{sp}	-0.18 (0.049)	-0.03 (0.869)	-0.12 (0.520)	-0.42 (0.025)	-0.16 (0.412)
σ_{bsp}	-0.15 (0.098)	-0.09 (0.622)	-0.12 (0.520)	-0.31 (0.103)	-0.06 (0.784)
SAE (450/635 nm)	0.08 (0.364)	-0.08 (0.702)	0.20 (0.299)	0.08 (0.702)	0.38 (0.055)
σ_{FFap}	-0.20 (0.030)	-0.30 (0.143)	-0.03 (0.890)	-0.53 (0.007)	-0.25 (0.182)

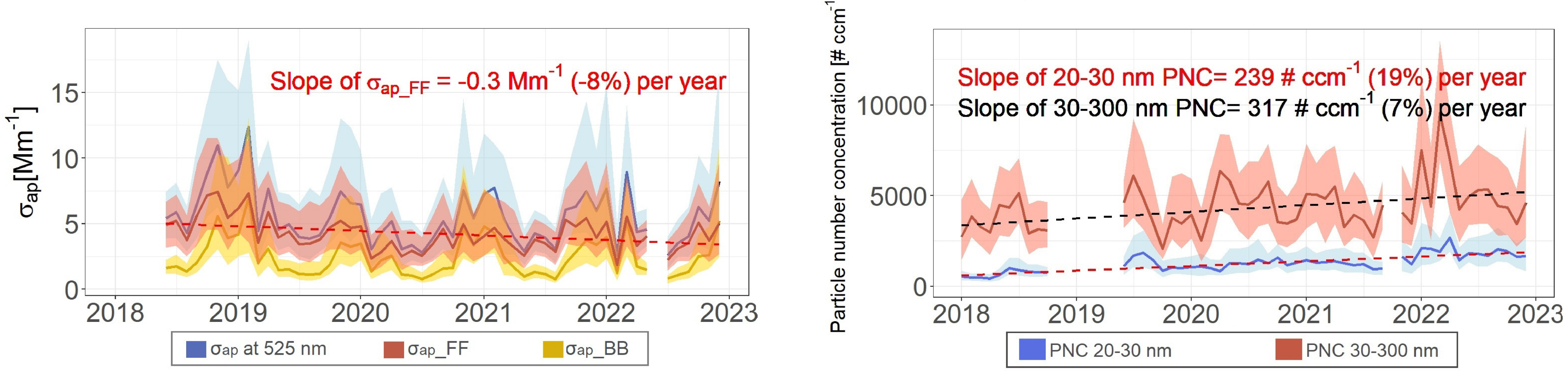


Figure 2. Monthly-based median variations with slopes of left: absorption properties right: particle number concentration of given modes observed at the ATOLL site from 2018 to 2022. The solid line shows the median; shadowed areas show the 25th and 75th percentile;dashed line shows the significant trendline change.

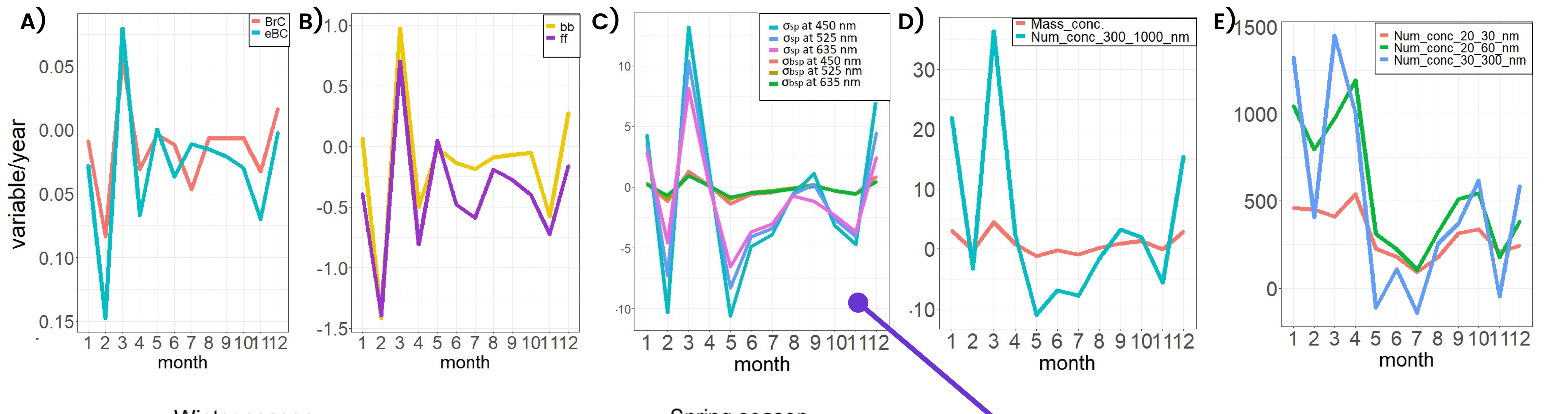


Figure 3. Slopes of separate monthly variations per year(2018-2022)of BrC and eBC (μg/m^3,A), contribution of biomass burning anf fossil fuels to σ_{ap}(Mm^{-1},B) σ_{sp} and σ_{bsp} at 450, 525 and 635 nm(Mm^{-1} ,C), number conc. of 300-1000 nm particle mode and mass conc. (#/ccm and μg/m^3, D), number conc. of 20-30, 20-60 and 30-300 nm particle modes (#/ccm, E).

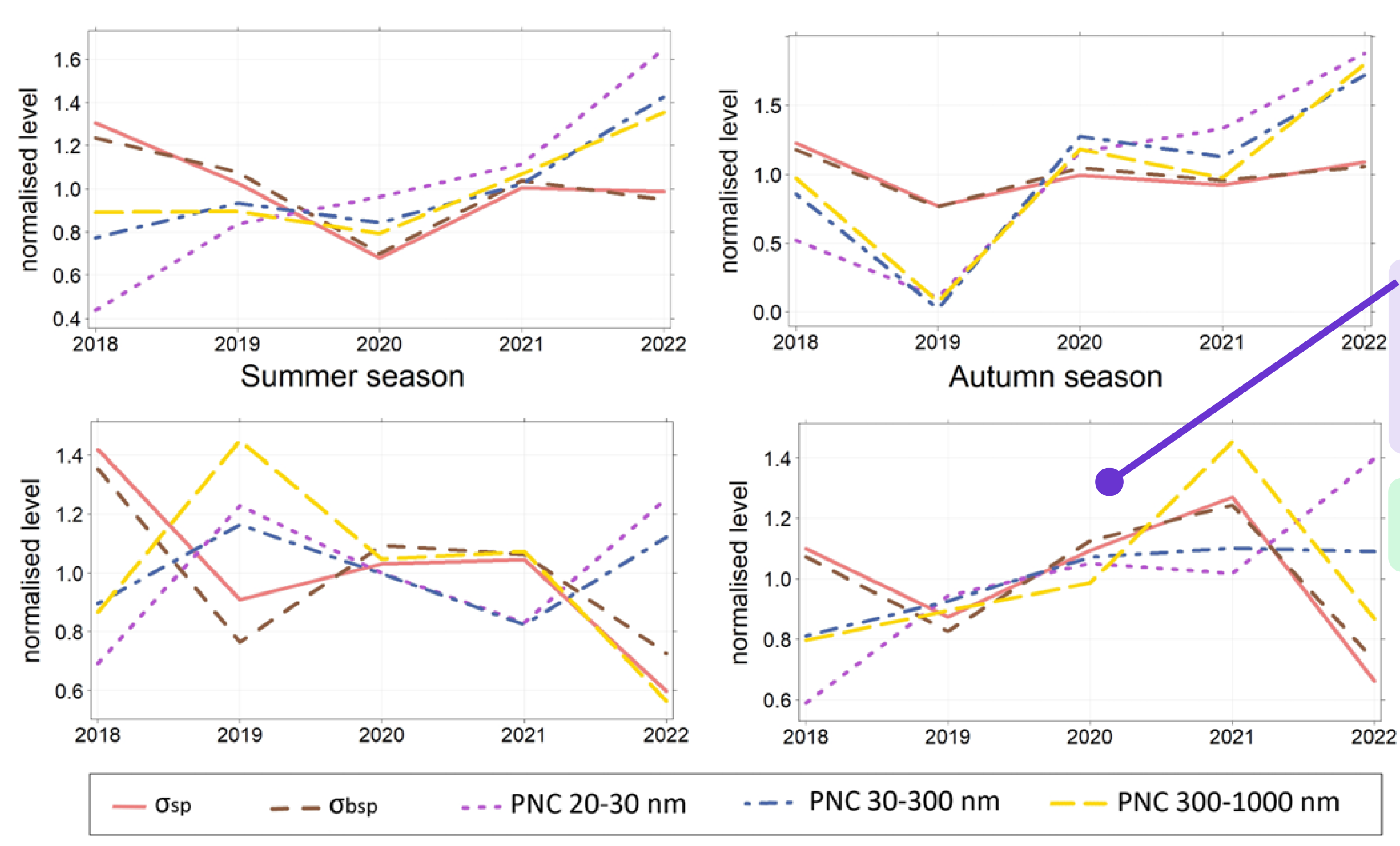
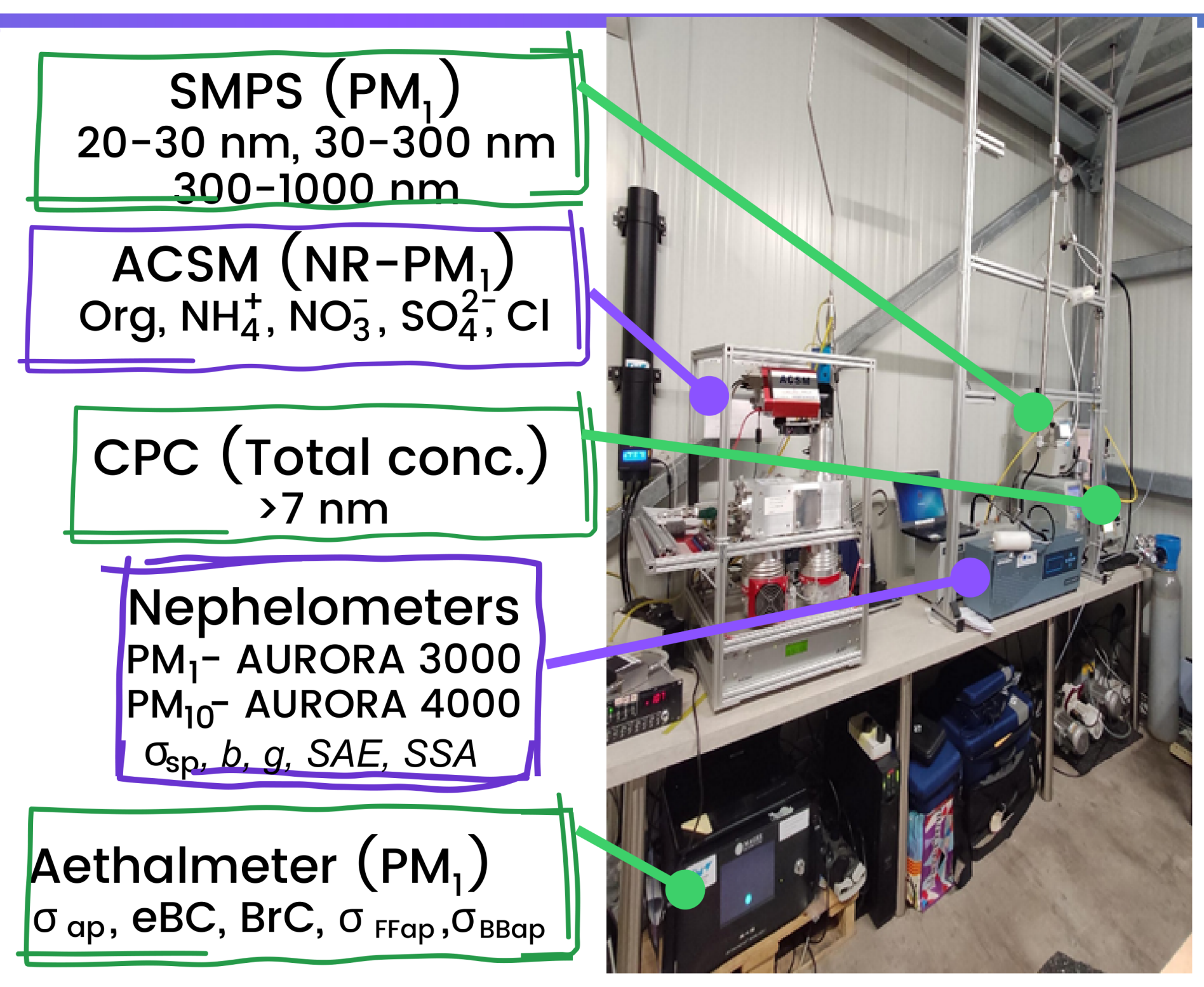


Figure 4. Interanual variations of σ_{sp} , σ_{bsp} at 525 nm and different modes or particle number concentration for every season. Each dataset is divided by its means to compare variables with diverse ranges and units; y axis is dimensionless.

Table 2. Increasing annual and seasonal slopes of particle number conc. of 20-30 and 20-60 nm mode.

Slope (#/ ccm/year)	Yearly	Winter	Spring	Summer	Fall
N_{20-30}	239	314	393	Not statistically significant	229
N_{20-60}	439	559	978		366
N_{20-300}	596	888	1584		504

METHODOLOGY



- ATmospheric Observations in LiLLE (ATOLL) ACTRIS station is located in Villeneuve d'Ascq (50.6114 N, 3.1406 E, 60 m a.s.l.).
- In-situ and remote sensing instruments (physical, chemical, optical and radiative properties of particles and clouds).
- Study period: January, 1 2018-December, 31 2022

RAW DATA → VALIDATIONS (STP,corrections , flagging, etc.) → **EBAS DATA** → Level 0 / Level 1 / **Level 2** → Climate relevant variables (b, g, SAE, SSA) , fossil fuel (**σ_{FFap}**) and biomass burning (**σ_{BBap}**) contribution to **σ_{ap}** calculation. → **Statistical analysis of variables (temporal variation)** with **corresponding slopes** R 4.1.0 + Matlab

NON-PARAMETRIC MANN KENDALL SEASONAL TREND TEST
- Used to estimate monotonic trends in the datasets over time
- The difference S_t between the later-measured value and all earlier-measured values, (y_j -y_i), where j>i,integer value is 1, 0, or –1 to positive differences, no differences, and negative differences, resp.
- Importance of data deseasonalization (S_t is sum of S for each season)
- from S: $\tau = \frac{S}{n(n-1)/2}$, where n is number of observations and τ is test statistic tau (-1 for complete decreasing trend to + 1 for complete increasing trend)

H_0: No monotonic trend in the series. H_1: A monotonic trend (positive/negative) in the seres presented

CONCLUSIONS

OPTICAL PROPERTIES

- Total significant decrease of σ_{sp} by -41.03% at 525 nm:
  - consistent with a decrease of total particle volume within the PM_1 fraction.
- *b(g)* consistently increasing (decreasing) → increased climate cooling aerosol potential.
- Scattering Ångström Exponent (*SAE*) → potential significant increase in the future (from 1.5 in 2018 to 1.9 in 2022):
  - PROVEN SIZE SHIFT TO SMALLER PARTICLE MODE.
- σ_{FFap} consecutively decreasing (25.00%) → source reductions or sinks of particular particles are more efficient?
- σ_{sp} and σ_{bsp}(525 nm) decrease for each season with a larger decrease in summer (-12.95 % and -9.20 % per year) (aerosol layer increasing by 5.10 % per year in summer).

PHYSICAL PROPERTIES

- Annual significant increase of 20-30 nm particles by 19.42%/year and 20-60 nm by 13.48%/year, (based on annual medians), significant increase in all seasones except summer
- Significant decrease of daily max. diameter by 4.7 μm/year.
- No yearly increase of NPF frequency during 2018-2022, Crumeyrolle et al., 2023 (size of backgorund aerosol shifted?).
- The month March shows unexpected behaviour (Figure 3.) as mentioned also in in CAMS reanalysis of aerosol optical depth (Flemming et al., 2017).

PERSPECTIVES

- Link to health effect (fine and ultrafine particles increase).
- Link to radiative transfer calculations.
- Comparison to others sites within the region such as Paris and Cabauw.
- Conflict in Ukraine resulted in oil crisis in Europe:
  - Increased prices of gas and oil → local change of fossil fuel sources (wood, coal, etc.)?
  - Increased SO_4^{2-} and NO_3^- conc. as well as particle conc. in accumulation mode → source apportionment .

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