Investigating the Significance of Organic Water Towards Atmospheric Aerosol Liquid Water Content in Florida

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Background

- Aerosol liquid water content (ALWC) is ubiquitous in the atmosphere, i.e., it is an important component of the atmosphere as it is widely present.[1]
- Atmospheric liquid water is affected by relative humidity (RH), as well as inorganic and organic aerosol concentrations and composition, both inorganic and organic matter.[2]
- ALWC can affect the quantity and chemical composition of organic aerosols.[3]
- Although the organic contribution to ALWC concentrations has been neglected in some studies, evidence of the significant contribution of organic to ALWC has been reported.[4]
- At present, the contribution of organic versus inorganic aerosols towards the ALWC is uncertain; reports can vary between 8% and 52% organic contribution.[4]
- The contribution of water associated with organic and inorganic aerosol can differ due to geographic location, concentrations of pollutants and their sources, in addition to the time of year (season)[5].

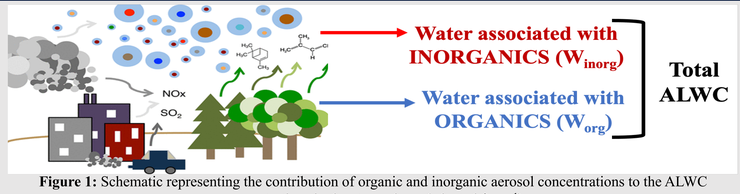

Figure 1: Schematic representing the contribution of organic and inorganic aerosol concentrations to the ALWC concentrations in the atmosphere. (adapted from [1])

Objective

The goal of this study is to:

- Determine the contribution of organic aerosol to the concentration of ALWC.
- Analyze organic and inorganic contributions to ALWC across multiple seasons and under contrasting environmental conditions in Florida.

Motivation

- Provide data to be used to improve the accuracy of ALWC modeling.
- Determine differences in organic ALWC contribution based on changes in location, population density, and other factors.
- Determine the temporal resolution of organic and inorganic ALWC.

Study Location

Three sites governed by the IMPROVE network were investigated. These sites represent the Northern, Central, and Southern regions of Florida (Fig. 2).

1. Tallahassee Community College (Tallahassee)
- Pop. Density: 1809.3 persons/mile^2
- Urban/suburban

2. Sydney (Dover)
- 1255.67 persons/mile^2
- Suburban/rural
- Near Tampa

3. Daniela BANU NCORE (Davie)
- Pop. Density: 3028.9 persons/mile^2
- Suburban
- Suburb of Ft Lauderdale

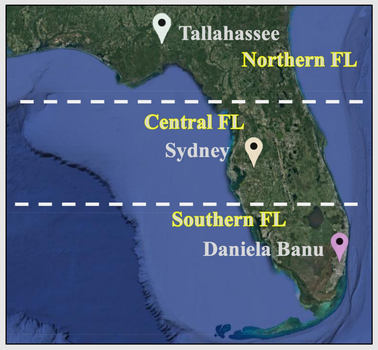

Figure 2: A map of the sites in Florida used in this study.

Data

- Speciated organic and inorganic $PM_{2.5}$ concentrations were acquired from the Florida Department of Environmental Protection[8].
- Gaseous ammonia and nitric acid concentrations were provided by EPA's CASTNET network.
- Specific meteorological data (Temperature and Relative Humidity) were acquired from Weather Underground[9].

Methodology

Inorganic water (W_{inorg}): ISORROPIAv2.1 aerosol thermodynamic equilibrium model[6].
$W_{inorg} = f$(inorganic concentrations, temperature, and relative humidity)

Organic water (W_{org}): Based on assumptions based on the kappa-Kohler theory[7].

$$W_{org} = \left\{\frac{m_{org}}{\rho_{w^*}\,\rho_{org}}\right\} * \left\{\frac{k_{org}}{(1/RH)-1}\right\}$$

where m_{org} is the measured organic mass concentration, ρ_w is water density, ρ_{org} is the organic density, k_{org} is the average kappa value of organics

Total ALWC: The sum of organic and inorganic concentrations.

$$\text{Total ALWC} = W_{inorg} + W_{org}$$

Contributions of W_{org} toward ALWC

- **Tallahassee**
  - ALWC concentrations did not vary over time.
  - Comparable W_{inorg} and W_{org} contributions towards the total ALWC concentrations.
- **Sydney**
  - ALWC concentrations increased by about 2 µg m^{-3} over the 5 years perhaps due to its proximity to the increasingly populated urban city of Tampa.
  - W_{org} contributions towards ALWC concentrations increased from 30% to more than 40% in 5 years.
- **Daniela Banu**
  - No clear temporal variation in ALWC concentrations.
  - W_{org} contributions towards ALWC concentrations were generally less than 20% except for 2021.
- **Overall**
  - ALWC concentrations were the highest in Sydney.
  - The organic contribution was 46.5%, 36.3%, and 17.5% in Tallahassee, Sydney, and Daniela Banu, respectively.

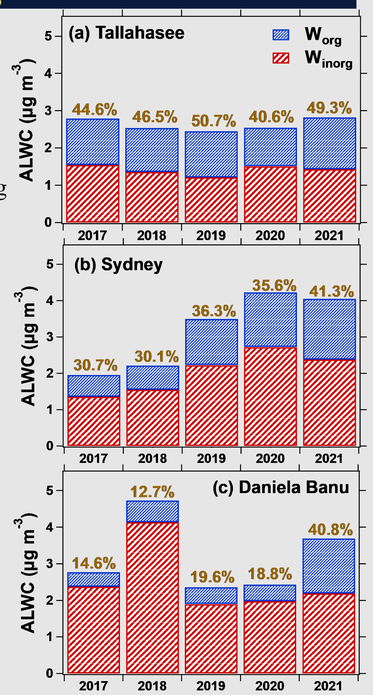

Figure 3: Seasonal median concentrations of W_{org} and W_{inorg} at (a) Tallahassee, (b) Sydney, and (c) Daniela Banu. Percentages represent W_{org} contributions.

Seasonal Variations in ALWC

- ALWC concentrations are the highest in the wintertime in Florida, opposite to trends observed in Baltimore[3].
- Spring has the lowest ALWC concentrations in Florida.
- Daniela Banu is coastal and rural in nature compared to the other sites, hence the lower W_{org} contributions to ALWC concentrations.

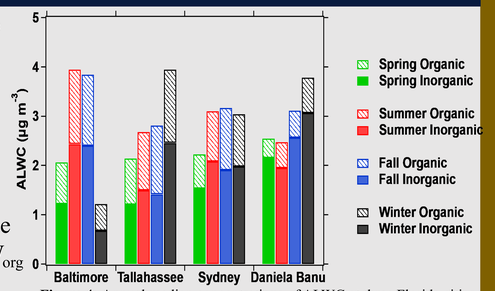

Figure 4: Annual median concentrations of ALWC at three Florida cities compared to Baltimore [5].

Conclusions

- Organic water is a significant contributor to ALWC concentrations in Florida.
- The highest contribution of W_{org} is in north Florida followed by the central and southern regions of the state.
- The contribution of organic water does not change as a function of season in Florida.

Future Work

- This study will be extended to include other states/locations with implications for the accurate representation of ALWC in models.
- Sensitivity analyses will be conducted to test the sensitivity of each of these concentrations to the sum of the aerosol and gaseous concentrations.

References

1. Nguyen, T. K. V., et al.: Liquid Water: Ubiquitous Contributor to Aerosol Mass, *Environ. Sci. Technol. Lett.*, 3, 257-263, **2016**.
2. Guo, H., Xu, L., et al.: Fine-particle water and pH in the southeastern United States, Atmos. Chem. Phys., 15, 5211–5228, 2015.
3. Carlton, A. G., and Turpin, B. J.: Particle partitioning potential of organic compounds is highest in the Eastern US and driven by anthropogenic water, *Atmos. Chem. Phys.*, 13, 10203-10214, **2013**.
4. El-Sayed M.M.H.: Formation of Secondary Organic Aerosols in Atmospheric Liquid Water, **2018**.
5. Jin, X., et al.: Significant contribution of organics to aerosol liquid water content in winter in Beijing, China, Atmos. Chem. Phys., 20, 901–914, **2020**.
6. Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+–Ca^{2+}–Mg^{2+}–NH_4^+–Na^+–SO_4^{2-}–NO_3^-–Cl^-–H_2O aerosols, Atmos. Chem. Phys., 7, 4639–4659, **2007**.
7. Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, **2007**.
8. Environmental Protection Agency. (n.d.). Pre-generated Data Files. Air Data. https://aqs.epa.gov/aqsweb/airdata/download_files.html **2023**.
9. IBM. (n.d.). *History*. Weather Underground. https://www.wunderground.com/, **2023**.

Acknowledgments

- Thanks to CASTNET for providing the gaseous ammonia and nitric acid data.

Tallahassee
Sydney
Daniela Banu