Field observations of aerosol physical and chemical properties in the Netherlands

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Summary

The primary aim of this PhD thesis is to investigate the aerosol physical and chemical characteristics in the Netherlands, including their chemical composition, size distribution, and optical properties. The research in this thesis began with a field experiment, namely the Ruisdael Intensive Trace-gas and Aerosol (RITA) campaign in 2021. The goal was to evaluate a range of aerosol instruments to ensure the reliability of observational data and to accurately assess local aerosol composition and concentration. The research then progressed to examining the vertical distribution of aerosol optical properties by combining calculated profiles with lidar retrieved profiles. The calculated profiles were based on those well-evaluated ground-based data from the RITA-2021 campaign. Lastly, the research extended to analyzing long-term observational data to provide a comprehensive understanding of the aerosol characteristics, particularly the particle size distribution in the Netherlands.

In Chapter 2, we applied various online and offline aerosol measurement instruments, with a particular focus on evaluating the time-of-flight aerosol chemical species monitor (TOF-ACSM) equipped with a $PM_{2.5}$ aerodynamic lens and a capture vaporizer. Offline filter analysis demonstrated excellent agreement with online ACSM measurements, with R^2 values ranging from 0.87 to 0.99 for NH_4^+, NO_3^-, and SO_4^{2-}. The bias in secondary inorganic aerosol concentrations was less than 10%, which is significantly better than the nominal ± 30% accuracy, reported from ACSM previously. Therefore, this advanced ACSM configuration is recommended for routine monitoring of the $PM_{2.5}$ chemical constituents. In addition, the outstanding performance allows further quantification of the local non-refractory (NR)-$PM_{1.0}$ and NR-$PM_{2.5}$. The average mass concentrations were 4.11 ± 3.32 μg m^{-3} for NR-$PM_{1.0}$ and 5.27 ± 3.98 μg m^{-3} for NR-$PM_{2.5}$ during the RITA-2021 campaign. On average, NR-$PM_{1.0}$ constituted 70%–80% of NR-$PM_{2.5}$, with organics (>33%) being the dominant component, followed by nitrate (>27%), sulfate (~18%), and ammonium (~17%). However, the chemical composition is size-dependent, with a higher mass fraction of organic carbon but a lower fraction of nitrate in $PM_{1.0}$ compared to $PM_{2.5}$. Besides, strong correlations ($R^2 > 0.9$) were observed among various online measurements, although the bias varied with relative humidity. Higher relative humidity resulted in increased hygroscopic growth and led to a smaller dry size cut-off of particles that differed slightly between different inlets (e.g., impactors vs. cyclones). To alleviate this issue, we recommend employing uniform size cut-off methods for different instruments to ensure the comparability of measurement and the consistency of long-term observations.

In Chapter 3, we investigated the vertical distribution of aerosol optical properties with the objective of predicting these properties by combining ground-based in situ measurements (chemical composition and particle size distribution) with ECMWF meteorological data, and

then comparing them to lidar retrievals. The optical properties included the scattering coefficient, backscatter coefficient, extinction coefficient, and lidar ratio. The predicted results were first validated against a TSI integrating nephelometer observations under dry conditions. Good agreements were achieved, with slopes for the scattering coefficient between 0.84 and 0.96 (R^2 >= 0.90) and for the backscatter coefficient between 1.01 and 1.18 (R^2 >= 0.67) at different wavelengths. Subsequently, we compared the predicted results with 26 aerosol optical profiles from Raman lidar measurements, finding that predicted and retrieved profiles (backscatter and extinction coefficients) matched approximately 35% of the time. However, the predicted lidar ratio profiles often aligned well with retrieved profiles, despite the considerable differences in the backscatter or extinction profiles. The differences between predicted and retrieved lidar ratios were generally within ± 30% for all the cases in our study. This approach, utilizing routine aerosol measurement data, not only validates lidar measurements but also bridges the gap between in situ measurements and lidar remote sensing. It aids in extending extinction profiles to lower altitudes and has the potential to support aerosol optical depth calculations. This method also has the potential to be applied to more aerosol measurement stations, for the prediction and validation of aerosol optical properties over longer periods in different environments.

In Chapter 4, we analyzed two years of aerosol data collected from two atmospheric observatories in the Netherlands, including a rural site Cabauw, and a more remote coastal site (Lutjewad). This research focused on the particle size distributions and incorporated chemical composition of aerosols and precursor gases, as well as meteorological data to understand the processes governing aerosol number concentrations in the Netherlands. Results showed a good correlation (R^2 = 0.62) between the concentrations of large aerosol particles (100-800 nm) at Lutjewad and Cabauw. This indicated that long-range transport or regional formation mechanisms predominantly contributed to the particle concentrations in this size range. However, the total average particle number concentration in Cabauw was approximately twice that of Lutjewad. This difference was mainly attributed to ultrafine particles (UFPs), as UFPs constituted over 80% of the total particle number concentration at both sites. In particular, particles in the nucleation size range (< 25 nm) accounted for an average of 35% of UFPs at Lutjewad and 46% of UFPs at Cabauw. High concentrations of UFPs in Cabauw were frequently linked with winds originating from specific directions, such as Schiphol airport, Rotterdam port, and the city of Utrecht nearby. Notably, a strong increase of particles in the nucleation size range during the morning rush hour on weekdays indicated traffic emissions as a primary or secondary source of particles with diameters < 25 nm. Cluster analysis of the particle size distribution showed a corresponding nucleation mode cluster with a high frequency of occurrence in the morning hours. Overall, new particle formation (NPF) analysis indicated that solar radiation is the key factor in NPF at both sites. However, NPF at Lutjewad was also influenced significantly by the availability of SO_2, while this influence was not pronounced at Cabauw. Overall, the study underscores the necessity

for further research to investigate the new particle formation mechanisms associated with traffic emissions, as well as their implications for public health and climate effects.

In summary, this thesis focused on the aerosol physical and chemical properties observed through field measurements in the Netherlands and discussed the aerosol characteristics from multiple aspects such as measurement and model prediction. The insights provided are particularly relevant to aerosol characteristics in the region, but some conclusions are also more broadly applicable.