

LINKING LONG-TERM AEROSOL CHEMICAL COMPOSITION AND OPTICAL PROPERTIES MEASURED AT THE ATOLL PLATFORM IN LILLE, NORTHERN FRANCE

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INTRODUCTION

Accurate knowledge of the physical, chemical and optical properties of aerosol particles is essential to reduce current uncertainties associated to their radiative impacts on Earth's climate. However, the wide range of particle sources and complex (trans)formation processes they undergo lead to strong spatial and temporal variability of these. The north of France is affected by many anthropogenic sources (e.g. road traffic, residential sector, agriculture and industries), maritime transport and oceanic emissions, as well as a high frequency of North-Easterly winds leading to aerosol pollution episodes (Favez et al., 2021; Potier et al., 2019; Roig Rodelas et al., 2019). Here, we investigate for the first time in situ aerosol optical properties and their relation to fine (PM₁) particle chemical composition in Northern France, based on a multi-annual in situ dataset.

METHODS

Since October 2014 near real-time in situ measurements within the fine fraction of aerosols (Table 1) are performed routinely on the ATOLL (ATMospheric Observations in LiLle) platform located on the rooftop of a University of Lille building (50.6111 N, 3.1404 E, 70 m a.s.l.), 6 km south east of Lille downtown area. The ATOLL site is part of the CARA program and of the French ACTRIS (Aerosols, Clouds, and Traces gases Research InfraStructure) National Facilities providing high-quality long-term atmospheric data.

Instrument	Measured Parameter	Time resolution
ACSM	Org, NH ₄ , SO ₄ , NO ₃ , Cl (NR-PM ₁)	~30 min
Aethalometer (7λ)	σ _{abs} & eBC	1 min
Nephelometer (3λ)	σ _{scat}	1 min
SMPS	Number size distribution	5 min

Table 1. List of in situ instrumentation and measured parameters in the PM₁ fraction at the ATOLL site.

In this study, aerosol optical properties and chemical composition coincident in situ measurements in the smallest fraction are combined to assess the particulate pollution specificities at the ATOLL platform via the IMPROVE algorithm. This method has been used for estimating the extinction coefficient (σ_{ext}) over remote and urban areas (e.g. USA, China) using fixed Mass Extinction Efficiency (MEE) values for six major chemical species of PM. Yet, MEE values vary according to aerosol types and properties, thus depending on site, season but also on derivation method (e.g. theoretical, laboratory chambers, and field

experiments) (Hand and Malm, 2007). Therefore, the application of such coefficients at different sites with no local tailoring can lead to significant uncertainties. To derive specific MEEs for ATOLL over 3 years of measurements, we applied a multiple linear regression (MLR) model using the measured optical properties (σ_{abs} , σ_{scat}) and the aerosols mass concentrations of the various chemical species (Table 1).

RESULTS

Using literature-based MEE reported for suburban and urban sites over 3 years of measurements at ATOLL, we retrieved the σ_{ext} with a wide range of biases depending on the chosen literature (Figure 1). Applying the values reported by the Interagency Monitoring of Protected Visual Environments (IMPROVE) program (Pitchford et al., 2007), the reconstructed extinction is retrieved with a slope of 0.77 and a moderate correlation (0.72) whereas, employing MLR improves the accuracy of σ_{ext} retrievals in comparison with the observations (r^2 :0.80, slope:0.85, Figure 1).

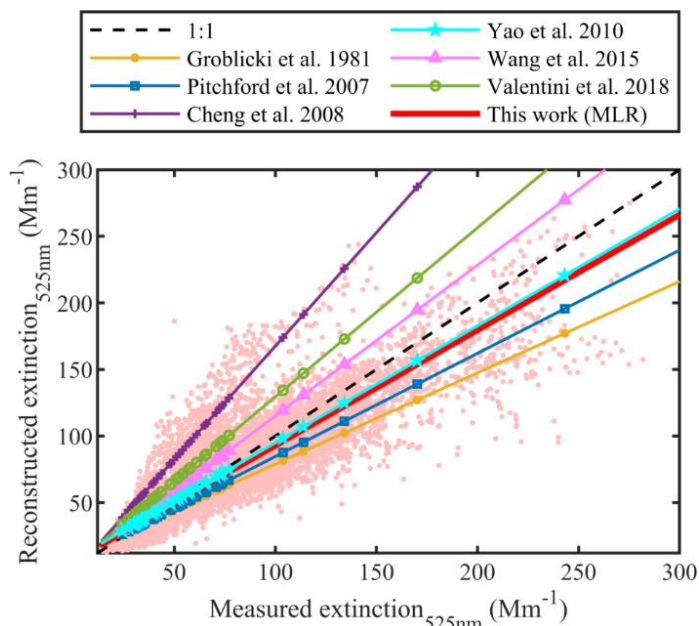


Figure 1. Scatter plot of reconstructed versus measured aerosol extinction coefficient at 525 nm for ~3-year measurements at the ATOLL platform. Solid lines represent the linear least-square fit through the data where y-values have been calculated using fixed MEE taken from the literature (Cheng et al., 2008; Groblicki et al., 1981; Pitchford et al., 2007; Valentini et al., 2018; Wang et al., 2015; Yao et al., 2010) combined with ATOLL PM_{10} species mass concentrations. Results of the Multiple Linear Regression with ATOLL specific MEE values are represented by the pink dots and the solid line in red.

This multi-annual analysis of in situ dataset at the ATOLL site highlights that ammonium nitrate contributes about 26% of the total mass, thus being the predominant species regarding light extinction at 525 nm (35%), followed by the carbonaceous aerosols (Org 26%, eBC 25%) and ammonium sulfate (14%). Regarding the absorption at 370 nm, organics are responsible of 11% of light absorption, showing significant contribution of Brown Carbon (BrC).

Figure 2 represents the monthly light extinction broken down according to species contribution, a product of mass efficiency and changes in the mass concentrations. As expected, light extinction is generally higher during the cold months. These months are strongly impacted by aerosol emissions related to solid fuel combustion from residential heating (carbonaceous aerosols), and agricultural activities leading to strong ammonium nitrate formation during spring.

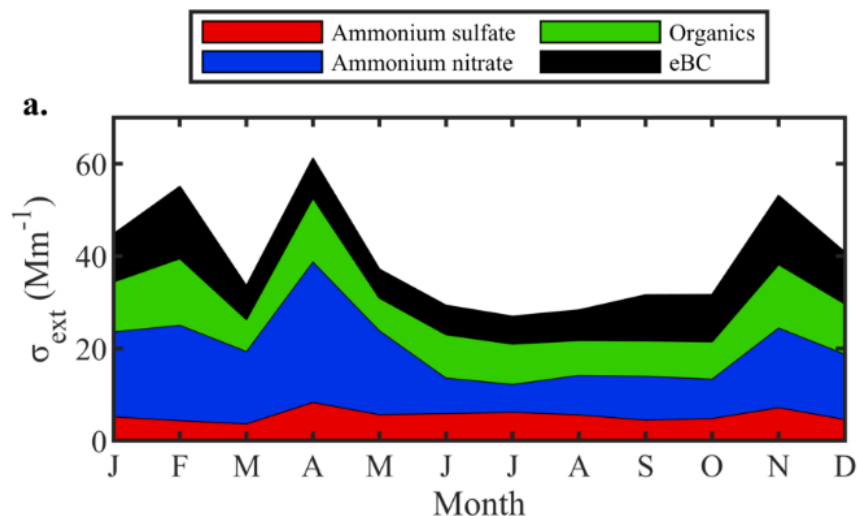


Figure 2. Monthly-averaged variation of the light extinction coefficient at 525 nm (in Mm⁻¹) at ATOLL over the period July 2017 – December 2019 for each aerosol species.

CONCLUSIONS

This study shows that not considering aerosol heterogeneity among different sampling sites and seasons, leads to significant biases in estimates of their extinction. Our analysis of the ATOLL data set in Lille highlights the ability of retrieving robust aerosol optical properties from chemical composition measurements, constrained by coincident absorption and scattering measurements. Our results support the use of site-specific parameters (that reflect various influences, as mixing state, size distribution of aerosol components) along with long-term field observations, to derive key aerosol optical properties relevant for climate modelling and air quality assessment.

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