Combining online and offline measurements for organic aerosol source apportionment

Combinaison de mesures automatiques et manuelles pour l'étude des sources de l'aérosol organique

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ABSTRACT

The present study proposes a novel approach to combine online and offline measurements in statistical source-receptor model such as Positive matrix factorization (PMF). Offline measurements include here POA (primary organic aerosol) and SOA (secondary organic aerosol) molecular markers. Their combination with data obtained using an Aerosol Chemical Speciation Monitor (ACSM) notably allowed for a better deconvolution of SOA fractions - at short time resolution (30min) - of SOA fractions than usually not obtained with online measurements only.

RESUME

Ces travaux proposent une nouvelle approche permettant de combiner des mesures automatiques et manuelles de la composition chimique des aérosols à l'aide d'un modèle statistique de type "Positive Matrix Factorization" (PMF). Les mesures manuelles incluent des marqueurs moléculaires spécifiques des aérosols organiques primaires et secondaires (AOP et AOS). La combinaison de ces mesures avec les données issues d'un Aerosol Chemical Speciation Monitor (ACSM) a notamment permis d'obtenir une meilleure discrimination des différentes fractions de l'AOS - sur un pas de temps court (30min) – que ce qu'il est généralement réalisé à l'aide des seules mesures automatiques.

KEYWORDS: Source apportionment, PMF, Organic aerosol, Online/Offline measurements/ **MOTS-CLES :** Modèle source-récepteur, PMF, aérosol organique, mesures automatiques et manuelles

1. INTRODUCTION

Understanding the sources and processes responsible of atmospheric Particulate matter (PM) composition and concentration is required to implement effective PM control strategies. During the last decade, the use of online non-refractory submicron aerosol mass spectrometer (AMS) and aerosol chemical speciation monitor (ACSM) measurements have successfully allowed for real time measurements of organic fraction. However, to better understand the sources and formation processes of organic aerosol, a comprehensive source apportionment analysis is still needed. The combination of different datasets from several measurements to refine the apportionment of OA (organic aerosol) sources, and notably secondary ones, is probably one of the best way to achieve this goal (Crippa et al. 2013; Huang et al. 2014; McGuire et al. 2014; Petit et al. 2014; Sun et al. 2012). In the present study, we propose a novel approach for combining online and offline measurements in statistical source-receptor model such as Positive matrix factorization (PMF).

2. METHODOLOGY

Measurements were conducted at SIRTA (Site Instrumental de Recherche par Télédétection Atmosphérique, 2.15° E; 48.71° N; 150 ma.s.l; about 25 km SW from Paris city center ; http://sirta.ipsl.fr). This ACTRIS monitoring station is representative of the suburban background air quality of the Paris area, mainly surrounded by forests, agricultural fields and urban areas. PM_{10} samples were collected every 4 hours over a period of intensive PM pollution events (PM > 50 µg m^{-3} over several days) on March 6-24, 2015, concomitantly with online measurements, carried out using ACSM, 7λ Aethalometer, TEOM-FDMS, NOx and O_3 analyzers.

An extended chemical characterization was performed on PM_{10} filters for the quantification of species such as Organic and Elemental Carbon (OC/EC), anions (Cl$^-$, NO$_3^-$, SO$_4^{2-}$), cations (NH$_4^+$, Ca$^{2+,}$ Na$^+$), methane sulphonic acid (MSA), oxalate (C$_2$O$_4^{2-}$), metals, levoglucosan, PAHs, oxy-PAHs and nitro-PAHs, and SOA markers.

3. RESULTS & DISCUSSION

Regular PMF was first performed on organic matrix obtained from offline measurements using the specific primary (i.e. levoglucosan (biomass burning), methane sulphonic acid (MSA) (marine), 1-nitropyrene (traffic)) and secondary organic molecular markers (i.e. 6H-dibenzo[b,d]pyran-6-one (PAHs), 2-nitrofluoranthene (PAHs), 3-methyl,5-nitrocatechol (biomass burning), α-methyl glyceric acid (isoprene)). PMF was also performed on ACSM OA matrix and has been deconvolved into four factors including HOA (Hydrocarbon OA), BBOA (biomass burning OA), MO-OOA (more oxidized oxygenated OA) and LO-OOA (low oxidized oxygenated OA).These results show that PMF performed on individual dataset is not truly viable to procure complete information about the different atmospheric processes. Lack of high-resolution time to understand rapid atmospheric processes in case of filter measurements, and lack of specific markers to validate oxygenated factors while using online measurements, have emerged out as a critical limitation of regular PMF.

Here, the synergic approach proposes to combine traditional off-line filter data including specific primary and secondary organic markers with OA matrix from ACSM measurements. The unified matrix was again deconvolved with PMF in order to retrieve factors such as HOA, BBOA, OOA, and to explore additional information about OA sources. Preliminary results applied on SIRTA (Paris) March 2015 observations allowed to resolve seven factor solution, notably decompose SOA in 5 explicit origins: 2 biogenic, 1 from nighttime chemistry, 1 related to oxy-PAHs chemistry and 1 related to aged traffic emissions (Figure 1).
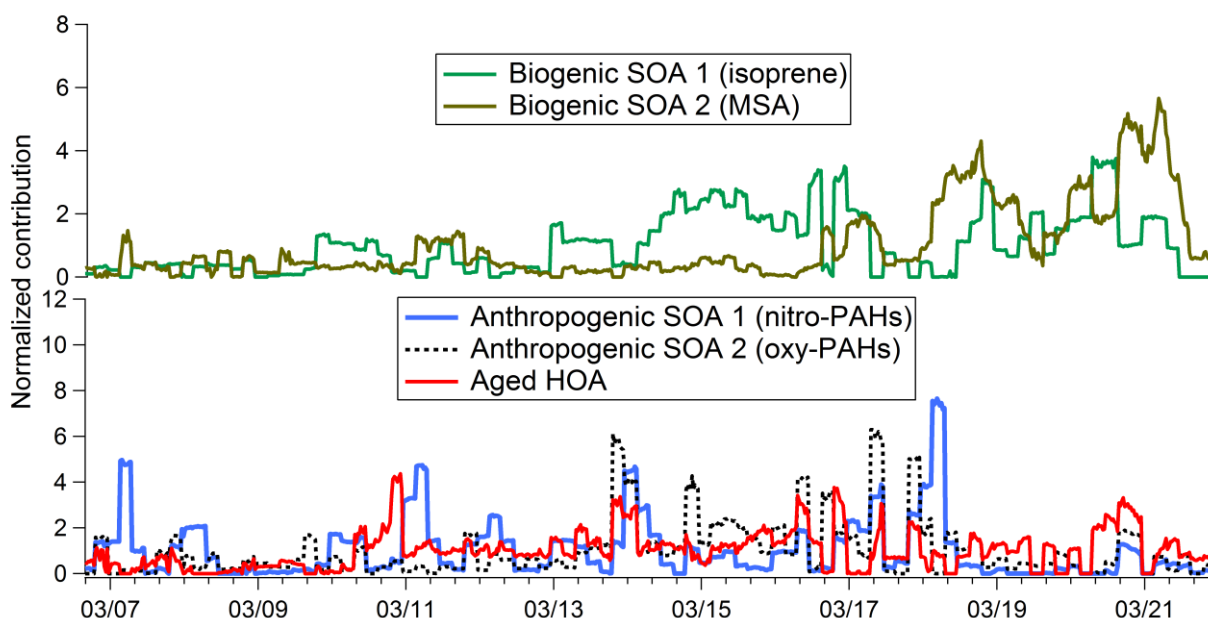

Figure 1. Secondary factor obtained using PMF performed on combined dataset at SIRTA (France).

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