UNIVERSITY^{OF} BIRMINGHAM

Research at Birmingham

Receptor modelling of fine particles in Southern England using CMB including comparison with AMS-PMF factors

Yin, Jianxin; Cumberland, Susan; Harrison, Roy; Allan, J.; Young, D. E.; Williams, P. I.; Coe, H

DOI:

10.5194/acpd-14-24523-2014

License:

Creative Commons: Attribution (CC BY)

Document Version

Publisher's PDF, also known as Version of record

Citation for published version (Harvard):

Yin, J, Cumberland, SA, Harrison, RM, Allan, J, Young, DE, Williams, PI & Coe, H 2014, 'Receptor modelling of fine particles in Southern England using CMB including comparison with AMS-PMF factors', Atmospheric Chemistry and Physics Discussions, vol. 14, no. 17, pp. 24523-24572. https://doi.org/10.5194/acpd-14-24523-2014

Link to publication on Research at Birmingham portal

Publisher Rights Statement:

Eligibility for repository : checked 18/11/2014

General rights

Unless a licence is specified above, all rights (including copyright and moral rights) in this document are retained by the authors and/or the copyright holders. The express permission of the copyright holder must be obtained for any use of this material other than for purposes permitted by law.

- Users may freely distribute the URL that is used to identify this publication.
- Users may download and/or print one copy of the publication from the University of Birmingham research portal for the purpose of private study or non-commercial research.
- User may use extracts from the document in line with the concept of 'fair dealing' under the Copyright, Designs and Patents Act 1988 (?)
- Users may not further distribute the material nor use it for the purposes of commercial gain.

Where a licence is displayed above, please note the terms and conditions of the licence govern your use of this document.

When citing, please reference the published version.

Take down policy

While the University of Birmingham exercises care and attention in making items available there are rare occasions when an item has been uploaded in error or has been deemed to be commercially or otherwise sensitive.

If you believe that this is the case for this document, please contact UBIRA@lists.bham.ac.uk providing details and we will remove access to the work immediately and investigate.

Download date: 01. Feb. 2019



This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Receptor modelling of fine particles in Southern England using CMB including comparison with AMS-PMF factors

J. Yin^1 , S. A. Cumberland¹, R. M. Harrison^{1,4}, J. Allan^{2,3}, D. E. Young², P. I. Williams^{2,3}, and H. Coe²

Received: 21 August 2014 - Accepted: 8 September 2014 - Published: 24 September 2014

Correspondence to: R. M. Harrison (r.m.harrison@bham.ac.uk)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Discussion

Discussion Paper

Discussion Paper

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page **Abstract** Introduction Conclusions References **Tables Figures** Back Close

Printer-friendly Version

Full Screen / Esc



¹Division of Environmental Health and Risk Management, School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B152TT, UK ²School of Earth, Atmospheric and Environmental Sciences, The University of Manchester, Oxford Road, Manchester M13 9PL, UK

³National Centre for Atmospheric Science, The University of Manchester, Oxford Road, Manchester M139PL, UK

⁴Department of Environmental Sciences/Center of Excellence in Environmental Studies. King Abdulaziz University, P.O. Box 80203, Jeddah, 21589, Saudi Arabia

PM_{2.5} was collected during a winter campaign at two southern England sites, urban background North Kensington (NK) and rural Harwell (HAR), in January-February 2012. Multiple organic and inorganic source tracers were analysed and used in a Chemical Mass Balance (CMB) model, which apportioned seven separate primary sources, that explained on average 53 % (NK) and 56 % (HAR) of the organic carbon (OC), including traffic, woodsmoke, food cooking, coal combustion, vegetative detritus, natural gas and dust/soil. With the addition of source tracers for secondary biogenic aerosol at the NK site, 79 % of organic carbon was accounted for. Secondary biogenic sources were represented by oxidation products of α -pinene and isoprene, but only the former made a substantial contribution to OC. Particle source contribution estimates for PM_{2.5} mass were obtained by the conversion of the OC estimates and combining with inorganic components ammonium nitrate, ammonium sulphate and sea salt. Good mass closure was achieved with 81 % (92 % with the addition of the secondary biogenic source) and 83% of the PM_{2.5} mass explained at NK and HAR respectively, with the remainder being secondary organic matter. While the most important sources of OC are vehicle exhaust (21 and 16%) and woodsmoke (15% and 28%) at NK and HAR respectively, food cooking emissions are also significant, particularly at the urban NK site (11% of OC), in addition to the secondary biogenic source, only measured at NK, which represented about 26%. In comparison, the major source components for PM_{2.5} at NK and HAR are inorganic ammonium salts (51 and 56%), vehicle exhaust emissions (8 and 6%), secondary biogenic (10% measured at NK only), woodsmoke (4 and 7%) and sea salt (7 and 8%), whereas food cooking (4% and 1%) showed relatively smaller contributions to PM_{2.5}. Results from the CMB model were compared with source contribution estimates derived from the AMS-PMF method. The overall mass of organic matter accounted for is rather similar for the two methods. However, appreciably different concentrations were calculated for the individual primary organic matter contributions, although for most source categories the CMB and AMS-PMF reACPD

Paper

Discussion Paper

Discussion Paper

Discussion Paper

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

I

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



sults were highly correlated ($r^2 = 0.69-0.91$). In comparison with the CMB model, the AMS appears to over-estimate the biomass burning/coal and food cooking sources by a factor of around 1.5 to 2 while estimates of the traffic source are rather similar for each model. The largest divergence is in the primary/secondary organic matter split, with the AMS estimating an appreciably smaller secondary component. Possible reasons for these discrepancies are discussed, but despite these substantial divergences, the strong correlation of the two methods gives some confidence in their application.

1 Introduction

Reduction of the airborne concentrations of particulate matter remains a high priority. The main drivers are European Union (EU) Limit Values and the health benefits to be gained from lower concentrations. In particular, the exposure reduction targets of the EU for fine particle $PM_{2.5}$ (a 15% reduction to be achieved by the UK by 2020 from 2009–2011 average concentrations) provide tough challenges for abatement measures. Cost-effective abatement depends upon a clear knowledge of the contributions of individual sources and source sectors to airborne concentrations. Currently in the UK, components of $PM_{2.5}$ for which data are particularly weak are woodsmoke (or solid fuel burning smoke more generally) (Harrison et al., 2012), cooking aerosol (especially in city centres with a high concentration of restaurants) (Allan et al., 2010), abrasion particles from road vehicles (Thorpe and Harrison, 2008; Pant and Harrison, 2013) and secondary organic fractions, which need additional research in order to fully understand their source contributions.

It has been established that significant amounts of the fine particulate matter ($PM_{2.5}$) are comprised of organic matter at sites within and outside Europe, representing around 25–31% in the UK West Midlands (Harrison et al., 2004), 21–33% in Ireland (Yin et al., 2005), 27–47% in Australia (Chan et al., 1997), 38–47% in France (Bressi et al., 2013) and 50% in Michigan, USA (Pancras et al., 2013). Organic matter is derived from both primary sources from which it is directly emitted to the atmosphere,

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

■ Back Close

Full Screen / Esc

Printer-friendly Version



and secondary production through oxidation of volatile organic compounds (VOCs) in the atmosphere. While numerous studies have been carried out upon the primary organic compounds in terms of their speciation and sources (e.g. Schauer et al., 1996; Stone et al., 2008; Yin et al., 2010; El Haddad et al., 2011a; Hasheminassab et al., ₅ 2013), the contribution of secondary organic aerosol to the total organic carbon and particle mass remains less clear due to its complex origins, composition and formation mechanisms in the atmosphere (Turpin et al., 2000; Hallquist et al., 2009). A number of studies have been carried out over mainland Europe on secondary organic aerosols formed through oxidation of biogenic volatile organic compounds (BVOCs) (Böge et al., 2006; Plewka et al., 2006; Wagener et al., 2012a, b), since their global emissions have been estimated to be 10 times higher than those of anthropogenic VOCs (Guenther et al., 1995). The major molecular markers for biogenic secondary organic aerosol constituents identified/used include (a) pinic and pinonic acid (the major oxidation products of α -pinene) (Presto et al., 2005), and (b) 2-methyltetrols (i.e., 2-methylthreitol and 2methylerythritol: oxidation products of isoprene) (Claevs et al., 2004; Kourtchev et al., 2005; Clements and Seinfeld, 2007; Stone et al., 2009).

Receptor modelling methods have been used for quantitative source apportionment of both primary and secondary particulate matter using chemically discriminated composition to provide source attribution. The widely used receptor models include Principal Component Analysis with Multiple Linear Regression (PCA-MLR), Positive Matrix Factorization (PMF), UNMIX and Chemical Mass Balance (CMB). The molecular marker-based CMB model requires aerosol chemical composition data from both the pollution sources and the receptor site, and has proved able to distinguish different primary sources of carbonaceous aerosols (Schauer et al., 1996; Zheng et al., 2002; Fraser et al., 2003; Stone et al., 2008; Yin et al., 2010; El Haddad et al., 2011a).

The contribution of secondary organic aerosol (SOA) has been estimated based simply upon laboratory-derived ratios of secondary organic carbon (SOC) mass to individual secondary organic marker compounds from the precursors isoprene, α -pinene, β -caryophyllene and toluene (Kleindienst et al., 2007; Lewandowski et al.,

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

▶ I

Full Screen / Esc

Close

Back

Printer-friendly Version

Interactive Discussion



Discussion Paper

Back **Printer-friendly Version**

Interactive Discussion



2008; Kourtchev et al., 2009; El Haddad et al., 2011b), although this method is subject to considerable uncertainties due to the simplification of replacing the complex atmospheric chemical reactions responsible for SOA formation with a laboratory-derived single-value mass fraction. The CMB model has also been used to apportion both primary and secondary sources (e.g. Stone et al., 2009) by the addition of specific secondary organic molecular markers derived from isoprene, α -pinene, β -caryophyllene and toluene, with the highest ambient concentrations observed for derivative species of isoprene and α -pinene (Lewandowski et al., 2008; Stone et al., 2009).

Our previous study at two sites in the West Midlands area of the UK (Yin et al., 2010) identified eight primary sources that contribute about 56-85 % on average to fine particulate organic carbon, including vehicular emissions (diesel engines, gasoline engines, smoking engines), wood smoke, vegetative detritus, natural gas combustion, coal combustion and road dust/soil. Vehicle exhaust emissions from all engines contributed up to 57% of the fine OC, with a relatively smaller amount up to 14% from other known sources, whilst a large amount (up to 34%) of the OC remained unexplained (termed as Other-OC), and was inferred to be mostly associated with secondary organic compounds.

A further study, reported here, has been carried out in southeast England at urban background and rural sites in order to obtain updated and extended information. Ambient aerosol samples have been analysed for multiple organic and inorganic source tracers, specifically including a number of additional markers for food cooking and secondary biogenic aerosols, in addition to the primary molecular markers previously analysed in the earlier study (Yin et al., 2010). This new dataset, particularly the food cooking and biogenic secondary markers, has allowed further evaluation of the concentration and sources of those components and the possibility for the first time to estimate, with the CMB method, the contributions of the main groups of biogenic VOC to secondary organic aerosol in the UK atmosphere. The CMB results have been compared with source contribution estimates derived from an Aerosol Mass Spectrometer (AMS), with an emphasis on sources of food cooking and secondary particles.

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page **Abstract** Introduction Conclusions References **Tables Figures** Close Full Screen / Esc

2.1 Site location and aerosol sampling

Sampling was carried out during the winter ClearfLo campaign in January–February 2012 in southeast England, UK at two sites, an urban background site, North Kensington, (NK) London and a rural site, Harwell, (HAR), Oxfordshire. The NK site (51°31″ N, 0°12″ W) is situated in the grounds of a school in a residential area, 7 km to the west of central London and is widely accepted as representative of air quality across a large part of London. The air pollution climate at the NK site has been previously analysed in detail by Bigi and Harrison (2010). The HAR monitoring station (51°34″ N, 1°20″ W) is situated to the west of London. The nearest road is a minor road located approximately 140 m from the station and the surrounding area is generally open with agricultural fields, with the nearest trees at a distance of approximately 25 m.

There were two collocated instruments at NK and HAR sites, a dichotomous Partisol 2025 sampler and a Digitel DHA-80 sampler for the purpose of both chemical and physical analyses. The Partisol sampler was used to collect 24 h fine (PM_{2.5}) particles onto 47 mm PTFE filters used for gravimetric and metal analyses. The Digitel was used for collecting also 24 h fine particles on 150 mm diameter quartz fibre filters, which were analysed for organic molecular markers, total organic carbon (OC), elemental carbon (EC), anions and cations. In addition to the samplers at NK, an Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer was deployed. The sampling record, instrument/filter media used for ambient sampling, chemical and physical properties analysed and the methodologies/instrumentation used in the laboratory are summarised in Table 1. Most of the detailed procedures have been outlined in previous studies (Harrison and Yin, 2010; Yin et al., 2010) and are briefly described here along with the new procedures which are described in greater detail.

14

Discussion

Paper

Discussion

Paper

Discussion Paper

Discussion Paper

14, 24523-24572, 2014

ACPD

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

I

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.2.1 PM_{2.5} mass and metals

The Partisol PTFE filters collected at NK and HAR were conditioned and weighed in a controlled environment room ($20\pm2\,^{\circ}$ C and $35-45\,^{\circ}$ R.H.) before and after exposure to obtain the gravimetric mass of PM_{2.5}. After gravimetric analysis, those samples were analysed for elements Fe, Si and AI using a Bruker S8 Tiger WD-XRF (X-ray Fluorescence Spectrometer) instrument, and then for metals Ti, Mn, Ni, Cu, Zn and Ba by Agilent 7500ec ICP-MS, after extraction using an aqua regia acid solution (Harper et al., 1983; Allen et al., 2001; Birmili et al., 2006).

2.2.2 lons, OC, EC and organic markers

The Digitel PM_{2.5} samples on quartz filters at NK and HAR were analysed for ions SO₄²⁻, NO₂⁻, CI⁻, Na⁺, K⁺, Ca²⁺, Mg²⁺ and NH₄⁺ using a Dionex ion chromatograph, OC and EC by Sunset Laboratory thermal-optical OC/EC analyser and organic markers (Table S1) by GC-MS, including 12 n-alkanes C₂₄-C₃₅, 9 hopanes, 10 PAHs, 2 sterols (cholesterol and levoglucosan), 6 fatty acids and 4 secondary biogenic molecular markers (at NK only), i.e. oxidation products of α -pinene (pinonic acid, pinic acid) and isoprene (2-methyltetrols: 2-methylthreitol and 2-methylerythritol). Similar methods to those reported by Yue and Fraser (2004) and Yin et al. (2010) have been applied for the sample extraction and analysis procedures, but a modified derivation/quantification method from Wagener et al. (2012a) was used for the secondary biogenic markers. In brief, one quarter of the Digitel sample was spiked with internal standards octacosane-d₅₈, aaa-20R-cholestane-d₄, dibenz(ah)anthracene-d₁₄, cholesterol-2,2,3,4,4,6-d₆, methyl-beta-D-xylopyranoside, heptadecanoic acid-d₃₃ and meso-erythritol and extracted with DCM and methanol (2:1) by undergoing mild ultrasonic treatment at room temperature. The combined extract was reduced in volume to approximately 5 mL using a turbo evaporator, then filtered/dried and further concen-

Discussion Paper

Discussion Paper

Discussion Pa

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

Back Close

Full Screen / Esc

Printer-friendly Version



trated down to 300 µL. One aliquot of the extract was analysed directly using an Agilent GC-MS system for non-polar compounds, n-alkanes, hopanes and PAHs, whilst the polar organics needed to be derivatised before the GC-MS analysis. For fatty acids, one aliquot of the extract was evaporated to near dryness and derivatised by addition of methanol and 2.0 M trimethylsilyldiazomethane (TMS-DM) in diethyl ether. For sterols and biogenic markers, another aliquot of the extract was concentrated down to near dryness and derivatised by addition of N,O-bis(trimethylsilyl)trifluoroacetamide plus 1 % trimethylchlorosilane (BSTFA + 1 % TMCS) and pyridine at 70 °C for 1 h, and finally cooled in a desiccator before being run on the GCMS. The analytical precision and detection limit for individual compounds calculated using repeated measurement of the lowest standard are listed in Table S2. Blank values higher than the DL were subtracted from the sample results.

2.2.3 AMS data analyses

The chemical composition of non-refractory PM_1 species was measured by an Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS, hereafter AMS), which operated in the standard configuration at NK, taking both MS and PToF data. A detailed description of the instrument can be found elsewhere (DeCarlo et al., 2006; Canagaratna et al., 2007). As the AMS sampled in an alternating sequence with other black carbon and aerosol volatility measurements, 5 min averaged ambient samples in "V mode" were only obtained every 30 min. Full details of the measurements are given in Young et al. (2014).

Data were analysed within Igor Pro (Wave metrics) using the standard analysis software packages, SQUIRREL v1.52J and PIKA v1.11J. A time and composition dependent collection efficiency (CE) was applied to the data based on the algorithm by Middlebrook et al. (2012) and was validated by comparing the volume concentration with that of the DMPS measurements. The AMS was calibrated using 350 nm monodisperse ammonium nitrate particles.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

Back Close

Full Screen / Esc

Printer-friendly Version



Discussion Paper

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page

Abstract Introduction Conclusions References

Tables Figures

Close

Back

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Positive matrix factorization (PMF) (Paatero, 1997) was performed on the organic data matrix from the "V-mode" data, permitting analysis of peaks according to elemental composition (Sun et al., 2011). While the "W-mode" data could in theory provide a more detailed analysis, too low a fraction of peaks were consistently fit by PIKA (due to the lower signal-to-noise) to permit a meaningful PMF analysis. A front-end for using the related ME-2 algorithm (Lanz et al., 2008; Paatero, 1999) is currently available (Canonaco et al., 2013), which in some circumstances can produce more accurate data. However, the benefits of this approach are most significant when applied to unit mass resolution (UMR) data (from the Q-AMS, C-TOF-AMS and ACSM), where key peaks (such as C₃H₇⁺ and C₂OH₃⁺) cannot be explicitly separated and therefore contribute to rotational ambiguity under normal PMF analysis. As this is not an issue with the HR-TOF-AMS data presented here, it was decided that it would be most appropriate to use PMF, so the results would not be influenced by a priori assumptions regarding the aerosol's behaviour.

The data were pre-processed in the recommended method of practice as described by Ulbrich et al. (2009). Isotopes were not included in the organic matrix and nitrogencontaining peaks were not deemed to have been successfully retrieved using PIKA. Five factors were identified: oxygenated OA (OOA), cooking OA (COA), hydrocarbonlike OA (HOA) and two solid fuel OA (SFOA 1 and SFOA 2), which had the appearance of "split" factors. While the 4-factor solution (which contained only one SFOA factor) seemed to be valid, the 5-factor solution gave improvements to diagnostics (e.g. Q) and correlations with ancillary data (e.g. NO_x, BC and CO), so it was deemed that the 5-factor solution with the split SFOA factors was the most appropriate. The 6-factor solution was discarded due to its significant dependency on initialisation seed (unlike the solutions with 5 or fewer factors) and as well as the production of a factor that did not appear physically meaningful. Further details are presented in Young et al. (2014).

Ambiguity due to rotational freedom within the solutions is a problem inherent to PMF, in common with many multivariate analyses; subtle changes in the mass spectral profiles can alter the mass concentrations of the factors, while still producing mathematically viable solutions (Paatero et al., 2002). This ambiguity was explored through varying the "fpeak" parameter and it was found that values between -0.6 and 1.0 produced solutions that could be considered valid (see Table S3). Outside of this range, solutions produced nonphysical factors or failed to converge properly. It was found that between these values, the concentrations of HOA and COA showed some variation, however the ambiguity was not a direct rotation between the two factors as would be expected for factors derived from UMR data. Because the high-resolution data is good at distinquishing HOA (which is mainly hydrocarbons) from COA (which contains oxygenated peaks), the HOA profile was consistent between all values of fpeak. Instead, the exchange of signals between profiles seemed to involve the COA and two SFOA factors. with HOA variance accounting for changes in the hydrocarbon peaks in the SFOA.

This range of variation can been seen as indicative of the amount of rotational ambiguity present in the solutions (Allan et al., 2010). However, the solution for fpeak = 0 is used for further analysis, as this is most likely to be physically meaningful according to the recommendations of Paatero et al. (2002), which does not advocate the use of nonzero values of fpeak for environmental data.

The CMB model 2.2.5

The US EPA CMB8.2 software was used for CMB modelling, with mostly similar source profiles to our earlier work, including vegetative detritus (Rogge et al., 1993a), natural gas combustion (Rogge et al., 1993b), wood smoke/biomass burning (Fine et al., 2004), dust/soil (Schauer, 1998) and coal combustion (Zhang et al., 2008). For traffic, the split of source profiles for gasoline, diesel and smoking engines may be incorrect as it is based on old engine source profiles from Los Angeles in 2001 (Lough

Paper

Discussion Paper

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page

Introduction

References

Figures

Close

Abstract

Conclusions

Tables

Back

Discussion Paper





Discussion Paper

Conclusions References **Tables**

Abstract

Figures

ACPD

14, 24523–24572, 2014

Receptor modelling

of fine particles in

Southern England

J. Yin et al.

Title Page

Close

Introduction

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



et al., 2007) and tends to overestimate the emissions from the UK traffic fleet (Yin et al., 2010; Pant et al., 2014). Therefore they were not applied here, and instead a single traffic source profile was generated from a twin site measurement from London (roadside site - background site) (Pant et al., 2014). Additional source profiles ₅ used were food cooking (Zhao et al., 2007b) and secondary biogenic emissions, which was generated from ambient measurements in Germany (Wagener et al., 2012a, b). Selected fitting species used in the model are elemental carbon, silicon, aluminium, levoglucosan, C_{29} – C_{35} n-alkanes, 17a(H)-22,29,30-trisnorhopane, 17a(H)-21b(H)hopane, 17b(H)-21a(H)-30-norhopane, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene, indeno(1,2,3-cd)pyrene, benzo(ghi)perylene, picene, n-hexadecanoic acid, n-octadecanoic acid, 9-octadecenoic acid, 9,12-octadecadienoic acid, pinonic acid, pinic acid, 2-methylthreitol and 2-methylerythritol. Detailed model performance measures can be found in Yin et al. (2010).

Results and discussion

Measured ambient concentration levels at NK and HAR

Average concentration statistics are shown in Table 2 for the measured components that are used in the CMB modelling. The mean concentrations of PM_{2.5} and its chemical components were mostly higher at the NK urban site than at the HAR rural site except for woodsmoke (levoglucosan) and vegetative detritus (n-alkanes) marker compounds which showed the opposite, whilst similar concentrations were observed for chloride, nitrate and sulphate.

3.1.1 Secondary organic marker components at NK

Quantifiable concentration levels of the secondary biogenic compounds were detected (0.19-1.3 ng m⁻³) (Table 2) at the London NK urban background site, but these levels are lower than those measured from other European sites (Table 3). Wagener









Discussion Paper

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page

ACPD

14, 24523–24572, 2014

Abstract Introduction Conclusions References **Tables Figures**

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



et al. (2012a) conducted measurement at three sites (HV – high vegetation influenced site, LV - low vegetation influenced site and regV - regional vegetation influenced site) in Berlin, Germany, and reported higher levels of those molecular markers for PM_{10} (0.8–13.2 ng m⁻³) and PM_{10} (0.6–11.9 ng m⁻³) at HV, PM_{10} (0.6–8.4 ng m⁻³) and $PM_{1.0}$ (0.3–6.9 ng m⁻³) at LV and PM_{10} (0.8–15.3 ng m⁻³) at regV. Regardless of the different particle size fraction measured, the concentration levels in Berlin are roughly 2-10 times higher than those at the London site, presumably due to influences from the surrounding forest area at the Berlin sites. Higher levels were also found at a background station in southern Sweden, showing average concentrations of 3.02 and 3.03 ng m⁻³ for pinonic and pinic acids in the PM₁₀ fraction, with higher levels in summer than in winter (Hyder et al., 2012), and at a rural background site at Hyytiälä, Finland, with mean PM_{1.0} concentrations for 2-methylthreitol and 2-methylerythritol at 5.1 and 21.2 ng m⁻³ in summer 2004 (Kourtchev et al., 2005). It is interesting to note that similar concentrations have been observed at the rural background site, Hyytiälä, Finland in autumn 2004 for the two isoprene-oxidation products (0.18 and 0.29 ng m⁻³), to those at the UK NK site, although the former is surrounded by forests. Clearly meteorological/seasonal effects as well as source proximity can influence the levels of the biogenic secondary organic markers, which explain the low concentrations detected at our site in the winter months. In particular, the isoprene-derived compounds show much higher levels in summer than in winter, whilst similar concentrations were observed for α -pinene derived products (Wagener et al., 2012a), which may explain the higher concentrations of pinic and pinonic acids than 2-methyltetrols at our NK site. Another study in summer 2002, in a coniferous forest in Germany (Plewka et al., 2006) indicated very different day and night concentrations, with higher night-time levels for pinic acid, but higher daytime levels for pinonic acid, 2-methylthreitol and 2-methylerythritol.

Apart from the European data, sampling has also been reported from four sites in a heavily wooded region in the south-eastern US in June 2004, and also showed higher average concentrations of 2-methylthreitol and 2-methylerythritol of 4.8 and 11.9 ng m⁻³ at the inland sites, and 1.6 and 4.9 ng m⁻³ at the coastal site (Clements

Discussion Pape

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page **Abstract** Introduction Conclusions References **Tables Figures** Back Close Full Screen / Esc

Printer-friendly Version

Interactive Discussion



and Seinfeld, 2007). It is noticeable that the concentrations of 2-methylerythritol are always higher than those of 2-methylthreitol at both our NK site and in the published work, whilst higher levels of pinonic acid than pinic acid were observed at NK and in Berlin, Germany (Wagener et al., 2012a) but not at the background station in Southern Sweden (Hyder et al., 2012) where similar mean concentrations were observed.

3.1.2 Primary organic components at NK and HAR

Concentrations of both biogenic and anthropogenic primary molecular marker compounds were mostly similar or higher in comparison with those of secondary marker compounds at the UK NK site, where anthropogenic sources such as traffic emissions, wood smoke and food cooking markers play an important role. Higher levels were found for levoglucosan (Levo) $(73.9 \text{ ng m}^{-3} \text{ and } 94.5 \text{ ng m}^{-3})$, hopanes $(0.25-0.50 \text{ ng m}^{-3} \text{ and } 94.5 \text{ ng m}^{-3})$ $0.079-0.36 \,\mathrm{ng}\,\mathrm{m}^{-3}$) and PAHs (0.10-0.67 $\mathrm{ng}\,\mathrm{m}^{-3}$ and 0.044-0.51 $\mathrm{ng}\,\mathrm{m}^{-3}$) at the current southeast England sites NK and HAR in winter 2012 than that measured at the UK West Midlands urban background monitoring site, EROS, (Levo: 9.2 ng m⁻³; hopanes: $0.08-0.18 \, \text{ng m}^{-3}$; PAHs: $0.06-0.27 \, \text{ng m}^{-3}$) and rural site CPSS (Levo: $7.7 \, \text{ng m}^{-3}$; hopanes: $0.07-0.15 \text{ ng m}^{-3}$; PAHs: $0.05-0.21 \text{ ng m}^{-3}$) in 2007–2008 (Harrison and Yin, 2010), but lower levels were observed for n-alkanes from the current study (0.58- $2.1 \, \text{ng m}^{-3}$ and $1.2 - 3.7 \, \text{ng m}^{-3}$ for NK and HAR), presumably due to a seasonal effect, since earlier results (1.0-5.2 ng m⁻³ and 1.8-4.7 ng m⁻³ for EROS and CPSS) cover both summer and winter periods. Similar or higher n-alkane concentration levels can be found from the current study if compared with winter periods only for EROS (0.73- $1.9 \,\mathrm{ng}\,\mathrm{m}^{-3}$) and CPSS (0.47–1.7 $\mathrm{ng}\,\mathrm{m}^{-3}$).

CMB model results

Source contributions to fine particulate OC and PM_{2.5} were calculated with the CMB model for the averaged samples for the whole sampling periods and for the individual daily samples for NK and HAR sites (Table 4 and Figs. 1-4).

Seven primary pollution sources were apportioned initially using the average concentration data (Table 4 and Fig. S1) that contribute on average about 53% at NK and 56% at HAR of the particulate organic carbon including traffic, wood smoke/biomass burning, food cooking, vegetative detritus, coal combustion, natural gas combustion and road dust/soil. The most significant sources identified are vehicle exhaust and woodsmoke emissions contributing about 21 % and 15 % of organic carbon (OC) at the London urban background NK site, and 16% and 28% at the rural HAR site. Other sources together contribute a relatively smaller amount of about 18 % for NK and 12 % for HAR respectively, including a significant amount of food cooking particularly at NK (NK: 11 % and HAR: 3 %), coal combustion (NK: 2 % and HAR: 2 %), vegetative detritus (NK: 2 % and HAR: 5 %), natural gas combustion (NK: 1 % and HAR: 2 %) and dust/soil (NK: 1 % and HAR: 1 %). As expected, most of the source contribution estimates, such as traffic, food cooking, coal combustion, dust/soil were larger at the urban site NK than that at the rural site HAR, where dust/soil (in bold figures) was not statistically significant over the winter period. The unidentified sources, referred to as "Other-OC", calculated as the difference between the measured total organic carbon and the sum of all source contribution estimates has been considered as being mostly secondary organic aerosol (SOA) (Yin et al., 2010) and any unidentified primary sources that are not accounted for in the CMB modelling. These represent about 47 % at NK and 44 % at HAR of the measured particulate OC over the whole sampling period. Daily source contributions fluctuate at both sites with a tendency that higher percentage mass explained by the model mostly occurred when ambient OC levels were low, and on the other hand, a large un-apportioned Other-OC component was often associated with high OC levels, indicative that secondary sources played an important role in these samples.

In order to apportion the Other-OC component, a source profile of the secondary biogenic component was generated using the mean ambient measurement data from

Paper

Discussion Paper

Discussion Paper

Discussion Paper

14, 24523–24572, 2014

ACPD

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page **Abstract** Introduction Conclusions References **Tables Figures** Close

Full Screen / Esc

Back

Printer-friendly Version

Interactive Discussion



Discussion

Berlin, Germany (Wagener et al., 2012a). Both PM₁₀ and PM₁₀ fractions were available, but the PM_{1.0} was used since previous data obtained by Wagener et al. (2012a) suggested that those biogenic markers were present mostly in the fine rather than the coarse fraction. The newly measured data on secondary biogenic molecular markers ₅ at NK, the 2-methyltetrols and the α -pinene-oxidation products, pinonic and pinic acid was combined with those source markers used earlier, and the CMB calculation was repeated to estimate an OC source contribution from secondary biogenic sources. Table 4 and Fig. S1 show the mean results with (NK2012b) and without (NK2012a) the addition of these new molecular markers.

On average, organic carbon was much better accounted at NK (79%) with the addition of the secondary biogenic components than without (54%). In addition to the major primary source components, traffic $(0.73 \,\mu\mathrm{g}\,\mathrm{m}^{-3})$, wood smoke $(0.54 \,\mu\mathrm{g}\,\mathrm{m}^{-3})$ and food cooking (0.39 µg m⁻³), the secondary biogenic concentration was estimated at 0.90 µg m⁻³, representing about 26 % of the total organic carbon mass for North Kensington in winter 2012. Similar concentration estimates were obtained using a PMF model in Berlin with ranges for PM_{10} of 0.34–0.84 μ g m⁻³ and PM_{10} of 0.43– 1.03 μ g m⁻³ in the colder months, and as 0.9–1.5 μ g m⁻³ for PM₁₀ and 1.1–1.2 μ g m⁻³ for PM_{1.0} in the warmer months (Wagener et al., 2012b). Relative source contributions to OC in Berlin were mostly similar to the UK site for the colder months (6.3-32.2%), but higher values were obtained for the warmer period (20.0-54.5%) in Berlin. Daily CMB results (Figs. 1 and 2) showed, as expected, fluctuations for the source contribution, with a few days when more OC was apportioned by the model than was measured (Fig. 2). Over 30 % of the OC was attributed to Other-OC on the days starting 13, 17, 30-31 January and 3-5 February when higher pollution levels of OC occurred, which is likely due to other biogenic and anthropogenic primary or secondary sources that are not accounted for on those days. Air mass back trajectories (Fig. S2) over those periods indicated that the high OC levels were strongly influenced by pollutants transported from mainland Europe on 17, 30 and 31 January, whilst during 13 January local or regional sources within the UK were dominant as the air mass travelled across the

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page **Abstract** Introduction Conclusions References **Tables Figures**

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Back

Atlantic Ocean and passed through southern England before reaching the site. On 3–5 February, both sources from mainland Europe and UK regional/local may have contributed.

3.2.2 Source contributions to PM_{2.5}

Source contribution estimates were calculated by multiplication of the fine OC source apportionment concentrations by the ratios of $PM_{2.5}$ mass to fine OC obtained from the same source profiles used for the primary OC apportionment (Pant et al., 2014; Rogge et al., 1993a, b; Fine et al., 2004; Zhang et al., 2008; Schauer, 1998; Zhao et al., 2007b). Whilst the aerosol mass to OC ratio is not available for the secondary biogenic sources, a ratio of 1.8 was used to obtain this source contribution estimate for the NK site. In addition to the seven/eight source components calculated from OC source contribution estimates, other organic matter (Other-OM) was estimated by multiplying the "Other-OC" by a factor of 1.8 (Utembe et al., 2009), sea salt calculated as $1.65 \times Cl^-$, ammonium sulphate as $1.38 \times SO_4^{2-}$ and ammonium nitrate as $1.29 \times NO_3^-$ were added into the $PM_{2.5}$ source apportionment (Harrison et al., 2003).

The output of the CMB model is critically dependent upon the source profiles used. The starting point was those used by Yin et al. (2010) which were mostly derived from earlier work in North America. A sensitivity study was conducted in which three ways were used of estimating the profile for road traffic following the work of Pant et al. (2014). The first two methods used dynamometer data, one using profiles of gasoline and diesel exhaust from dynamometer tests reported by Schauer et al. (1999, 2002). Secondly, more recent dynamometer data, reported by Lough et al. (2007), were utilised. Thirdly, a profile for road traffic created from measurements in a heavily-trafficked street canyon in London after subtraction of the local urban background as reported in Pant et al. (2014) was tested. This profile seems more likely to be reflective of the current vehicle fleet in London as the data are relatively recently collected. For each of these three traffic profiles, CMB was run with two different woodsmoke profiles, both taken from the USEPA SPECIATE database, one for USEPA Region 4 and

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Conclusions
Tables



Introduction

References



Abstract









Printer-friendly Version



14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

ACPD

J. Yin et al.

Title Page **Abstract** Introduction Conclusions References **Tables Figures** Back Close Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the other for USEPA Region 5. In order to judge which source profiles gave the best results, the estimate for road traffic exhaust from CMB was compared with an estimate based on elemental carbon using an OC/EC ratio of 0.63 as measured in recent London data as well as an OC/EC ratio of 0.35 as reported for roadside sites in Europe by Pio et al. (2011). The derivations from elemental carbon concentration gave estimated traffic OC of 0.96 µg m⁻³ and 0.54 µg m⁻³ from the two OC/EC ratios and the estimate from the application of the traffic source profile derived from data collected in London of an OC concentration of 0.73 µg m⁻³ lay comfortably between these values and the measured London profile from Pant et al. (2014) was our final choice.

There was also some sensitivity of the model output to the choice of woodsmoke profile, with the EPA Region 5 profile giving an estimate for woodsmoke OC of $0.53 \, \mu g \, m^{-3}$ and that for EPA Region 4, giving 0.78 µg m⁻³ of organic carbon. The estimate for woodsmoke organic carbon based upon mean levoglucosan for the campaign times a factor of 7.35 (Puxbaum et al., 2007) gave an OC concentration of 0.54 µg m⁻³ which lay very much closer to the estimate using the USEPA Region 5 source profile and therefore this profile was adopted for the final runs of the model.

Once the optimal source profiles had been selected using the campaign averaged dataset, source contribution estimates to OC and PM_{2.5} were run both on the averaged dataset and on the data for each separate day of the campaign. Since the model needs to be separately optimised for each measurement day, the average of the daily model runs presented in Table S4 is slightly different from that derived from the overall campaign averaged concentration data which is reported in Table 5. Unless stated otherwise, source contribution estimates reported are derived from the overall average dataset.

Concentrations of woodsmoke PM_{2.5} were found by the CMB model to be an average of 0.64 µg m⁻³ at North Kensington in the winter 2012 campaign and 0.77 µg m⁻³ at Harwell during the same campaign (from daily data). These concentrations are slightly higher but comparable magnitude to those measured in London in the winter of 2011 (Harrison et al., 2012). As they were measured at the coldest time of the year, it can be

anticipated that annual average concentrations of woodsmoke would be appreciably lower than from those measured during the winter campaign and probably no more than 50% of these concentrations.

PM_{2.5} source apportionment results for both mean and daily samples at NK and 5 HAR sites are shown in Table 4 and Figs. 3 and 4. PM_{2.5} mass was well explained by those source components which represented about 81 %/92 % without/with the addition of secondary biogenic component at the urban NK site over the winter period. This comprised on average of 37 % ammonium nitrate, 14 % ammonium sulphate, 8 % exhaust emissions, 10% secondary biogenic, 7% sea salt and 24% of all other identified/unidentified source components (vegetative detritus, wood smoke, natural gas, coal, dust/soil, food cooking, Other-OM and the unidentified component). The rural HAR site, with 83% total explained PM_{2.5} mass, showed similar relative source contributions from ammonium salts (37% ammonium nitrate and 19% ammonium sulphate) but a smaller relative contribution from vehicle exhaust emissions (6%). In comparison, ammonium salts were also predominant in the UK West Midlands sites for both winter (urban background: 33.6 %, rural: 37.7 %) and summer (urban background: 52.5 %, rural: 44.2 %) periods (Yin et al., 2010), with much higher estimated contributions from vehicular emissions in the winter months (urban background: 29.0 %, rural: 23.7 %), due to apportionment with separate traffic source profiles for diesel, gasoline and smoking engines which lead to an overestimation (Pant et al., 2014). In addition, the source contribution estimates from food cooking (previously not apportioned) were not large but significant particularly at the urban NK site (OC: 0.39 μg m⁻³, PM_{2.5}: 0.69 μg m⁻³) representing about 11 % of the OC and 4 % of the $PM_{2.5}$ mass. As discussed for OC above, the secondary biogenic source contribution, 26% for OC and 10% for PM25 at the UK site NK, cannot be ignored, particularly as during summer months this component may be significantly larger (Wagener et al., 2012b). Heal et al. (2011) studied the carbon-14 content in PM_{2.5} samples from a UK West Midlands urban background site (EROS) in 2007/08, and found that the fraction of contemporary total carbon f_c (TC) was positively correlated to the ratio SOC/TC, which were both related to airmass oriACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Full Screen / Esc
Printer-friendly Version



Discussion Paper

Discussion Paper

Conclusions References **Tables Figures**

Abstract



Introduction

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



gin, suggesting that secondary organic aerosol is substantially associated with the oxidation of biogenic VOC emissions from terrestrial contemporary carbon sources from continental Europe. An average estimate of about 40 % of the total carbon and 9-29 % of the PM_{2.5} was attributed to biogenic SOC or biogenic SOA (Heal et al., 2011). Those contribution estimates were higher in comparison with the current CMB estimates from the NK site (26 and 10 % for OC and PM_{2.5}), which is likely due to (a) use of a different sampling season, i.e. the study at NK only involve winter months while both winter and summer months were included in the study by Heal et al. (2011), and (b) estimates of contemporary carbon using carbon-14 also contain sources of non fossil OC/OA other than biogenic SOC/SOA, whilst the estimates from the current study include only those components which correlate with the oxidation products of α -pinene or isoprene.

The results from NK indicate an average secondary/Other-OC organic component of $PM_{2.5}$ mass of $2.95 \,\mu g \, m^{-3}$, of which $1.63 \,\mu g \, m^{-3}$ (55%) is accounted for by the inclusion of oxidation products of α -pinene and isoprene. It seems very probable that production of other biogenic VOC and their oxidation products would correlate strongly with α -pinene and isoprene, which are recognised as two of the main precursors of biogenic secondary organic aerosol. This leads to the tentative conclusion that at least 55% of the secondary organic aerosol (SOA) is biogenic in origin, even during winter, consistent with the conclusions of both Heal et al. (2011) and Charron et al. (2013) that SOA in the south and Midlands of the UK is dominated by the biogenic component. By inference, up to 45% of SOA may arise from anthropogenic precursors such as toluene.

Comparison between CMB and AMS-PMF estimates

The AMS data collected at NK during winter 2012 has been analysed and apportioned using the PMF (Positive Matrix Factorisation) model based on the method used by Allan et al. (2010). Full details of the methods are available from Young et al. (2014). The results identified five source emission components that contribute to organic aerosols/matter (OA/OM), including one secondary component (OOA) and four pri-

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

ACPD

J. Yin et al.

Title Page

mary components, two biomass burning/solid fuel burning organic aerosol (SFOA) factors, cooking organic aerosol (COA) and traffic related/hydrocarbon-like organic aerosol (HOA). Comparison has been made with related OM components calculated from the CMB modelling by applying OM/OC ratios considered appropriate to the source, as illustrated in Tables 5 and 6 and Fig. 5. The two SFOA factors have been summed as this gave the strongest correlation to both the woodsmoke and woodsmoke plus coal contributions derived from the CMB. Table 6 used OC average estimates calculated from the daily CMB results, shown in Table S4, in order to be consistent with Fig. 5.

3.3.1 Woodsmoke particles

The mean concentration of SFOA ($1.63 \, \mu g \, m^{-3}$) derived from AMS-PMF was 2.0 times the CMB woodsmoke (CMB-WS) concentration estimate ($0.85 \, \mu g \, m^{-3}$) (Table 6), possibly due to the SFOA factor also including particles from other solid fuel combustion apart from wood burning. The AMS-SFOA value remained larger at 1.7 times the CMB value when the CMB coal combustion component was also included (CMB-WS + Coal: $0.97 \, \mu g \, m^{-3}$). Nevertheless good correlation was observed between CMB-WS and AMS-SFOA ($r^2 = 0.75$) (Table 5 and Fig. 5a), indicating that the SFOA component is closely related to woodsmoke. It is notable that a stronger correlation is observed between the AMS-SFOA and the CMB component (WS+Coal) ($r^2 = 0.78$), which may indicate other sources in the AMS component SFOA apart from woodsmoke. It is interesting to note that both CMB-WS and CMB-(WS+Coal) are correlated more closely to SFOA at low levels of those components (CMB-WS/WS+Coal < 0.9/1.2 and SFOA < 3.0) as compared to high levels when data points are more scattered from the regression line (Fig. 5a and b).

Previous work in the UK has obtained lower biomass smoke OC values for Birmingham EROS 2009–2010 (0.23 $\mu g\,m^{-3}$), London NK 2010/11 (0.33 $\mu g\,m^{-3}$) and Budbrooke, Warwickshire 2009–2010 (0.42 $\mu g\,m^{-3}$) (Harrison et al., 2012), whilst six Euro-

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

■ ■ Back Close

Full Screen / Esc

Printer-friendly Version



pean sites showed biomass smoke OC concentrations in the range of $0.039-3.1 \,\mu g \, m^{-3}$ annually and $0.048-7.7 \,\mu g \, m^{-3}$ for winter months (Gelencsér et al., 2007).

Based on the CMB-WS estimates, woodsmoke can represent on average about 15% and 28% of the OC, and 4% and 7% of the PM_{2.5} for NK and HAR respectively. The relative contributions of woodsmoke to OC are similar to those measured in Belgium by Maenhaut et al. (2012), and the woodsmoke contributions to PM_{2.5} are in line with the lower range calculated by Zhang et al. (2010) who conducted measurements at 15 urban/rural sites in the south-eastern US and estimated that the relative contribution of biomass burning to PM_{2.5} were 13% annually, 27% in winter and 2% in the summer months.

3.3.2 Food cooking particles

Many studies have indicated that food cooking is one of the most important aerosol emission sources in the indoor environment (Kamens et al., 1991; Zhao et al., 2007a; Buonanno et al., 2009; Wan et al., 2011; Massey et al., 2012). A detailed review has shown that cooking aerosol is a significant PM source for both indoors and outdoors (Abdullahi et al., 2013), and may arise from both residential and commercial food cooking. The AMS has been used to characterise PM and identify organic aerosols from cooking by means of application of PMF to mass spectral data (Kleeman et al., 2008; Allan et al., 2010; Huang et al., 2010; Williams et al., 2010; He et al., 2010 and 2011; Sun et al., 2011; Zhang et al., 2011; Mohr et al., 2009 and 2011; Clougherty et al., 2011), whilst CMB is able to calculate the food cooking concentration estimate using appropriate molecular markers (Zheng et al., 2002; Fraser et al., 2003; Schauer et al., 1996; Robinson et al., 2006; Wang et al., 2009). Food cooking estimates were calculated for the first time in the UK using a CMB model with this source profile at both sites NK and HAR. Earlier work (Yin et al., 2010) used only cholesterol as a tracer of meat cooking, suggesting much lower concentrations. AMS-PMF method data were available for the NK site only, for which the results were compared.

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

Back Close

Printer-friendly Version

Full Screen / Esc



The CMB-derived mean food cooking concentration estimates for OC and OM/PM $_{2.5}$ from averaging the daily CMB results are 0.32 and 0.56 $\mu g \, m^{-3}$ at NK (Tables S4 and 5), representing about 9 % of the fine OC and 4 % of the PM $_{2.5}$ mass. In comparison, the PMF apportioned results from the AMS data gave a value of 0.87 $\mu g \, m^{-3}$ for cooking organic aerosol, which is about 1.6 times the value of the PM $_{2.5}$ CMB estimate. A strong correlation ($r^2 = 0.80$, Pearson) was found for the daily food cooking estimates between the AMS-PMF and CMB datasets, with a gradient of 2.40 (the gradient is 1.76 with a small intercept of -0.13 after removing the high value point) (Table 5 and Fig. 5c). A much lower food cooking contribution was obtained from the CMB method for HAR (0.12 $\mu g \, m^{-3}$ or 1 % of PM $_{2.5}$), where no immediate local sources were present.

Similar food cooking concentration estimate ranges have been observed in many previous studies for outdoor measurements from both CMB and AMS-PMF source apportionment. Fraser et al. (2003) conducted source apportionment using CMB for both urban and background sites in Houston, Texas, and identified a PM_{2.5} source component of meat cooking of 0.9–1.3 μg m⁻³ at an urban and 0.7 μg m⁻³ at a background site. Robinson et al. (2006) used CMB to apportion ambient fine OC, and indicated that 10% or 0.32 µg m⁻³ of the OC is attributable to food cooking in Pittsburgh, Pennsylvania. Zheng et al. (2002) estimated that about 5-12% of the fine OC arose from meat cooking emissions in the South-Eastern US, whilst Lee et al. (2008) estimated that 12 % of the PM_{2.5} mass was from meat cooking in Korea. Sun et al. (2011) apportioned $PM_{1.0}AMS$ data using PMF and obtained 1.02 $\mu g\,m^{-3}$ of cooking organic aerosol in New York City, which contributes 30 % to the primary OA. Williams et al. (2010) analysed the AMS data collected in Southern California with PMF source apportionment, which identified 10.4% or 0.98 µg m⁻³ of cooking emissions from the organic aerosol in summer. Huang et al. (2010) concluded that about 24.4% of the OA was related to cooking emissions during the Beijing 2008 Olympic Games. There appears to be a systematic difference, with AMS studies estimating larger contributions to OM and PM_{2.5} than CMB studies. Summertime concentrations for cooked meat-related air particles

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

▶I

Back Close

Full Screen / Esc

Printer-friendly Version



are normally higher than in wintertime, presumably due to increased outdoor cooking activities and open kitchen windows.

3.3.3 Traffic related particles

The CMB mean concentration estimates (Table 4) of total traffic are $0.73 \, \mu g \, m^{-3}$ for OC and $1.26 \, \mu g \, m^{-3}$ for PM_{2.5} at North Kensington, while about a half of these levels were observed at rural Harwell (OC: $0.36 \, \mu g \, m^{-3}$ and PM_{2.5}: $0.61 \, \mu g \, m^{-3}$). The relative contribution of total traffic to OC and PM_{2.5} at the urban site is about 21 % and 8 %, and at the rural site about 16 % and 6 % respectively. Very strong correlations ($r^2 = 0.90$ –0.99) have been observed for the CMB traffic component and other related variables, aethalometer BC, measured EC and calculated primary organic carbon (obtained based on the method of Castro et al., 1999) at both urban and rural sites. The AMS-PMF derived component HOA is also strongly correlated with the CMB OM traffic component ($r^2 = 0.80$) for NK, and if the two outlying points are removed, the correlation improves ($r^2 = 0.86$) and the gradient reduces to 0.98. The absolute OM concentration levels are very similar from the two methods, showing a CMB traffic estimate of 0.98 $\mu g \, m^{-3}$ and an AMS value of 0.86 $\mu g \, m^{-3}$ (Table 6).

3.3.4 Secondary particles

The CMB component Other-OC/Other-OM is regarded as mostly secondary OC/OM (Yin et al., 2010). The Other-OM, with concentration estimates of $2.92\,\mu g\,m^{-3}$ at North Kensington and $1.85\,\mu g\,m^{-3}$ at Harwell (Table S4), represented about $46\,\%$ and $45\,\%$ of the total organic aerosol (OA) mass, and $19\,\%$ and $17\,\%$ of the PM_{2.5} mass at those two sites. The secondary component derived from the AMS-PMF results (OOA) is $0.99\,\mu g\,m^{-3}$ for the same period at NK, which is well below the CMB Other-OM level. Docherty et al. (2008) studied secondary organic aerosol (SOA) at Riverside, Southern California in the summer period using different methods, and showed very similar proportions of SOA/OA estimated by the CMB (77 %) and AMS-PMF (74±19 %) meth-

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

Back Close

Printer-friendly Version

Full Screen / Esc



ods, but with much higher relative contributions of SOA comparing with our UK NK site. However, the study also summarised results from previous studies in the Eastern LA Basin area with different methods, found that the SOA/OA ratios were mostly less than 50 % (range: about 15–50 %), with only one exception (75 %) by Schauer et al. (2002), and attributed those large differences to variations in sampling season, location, duration and methodology.

The CMB Other-OC/OM is strongly correlated with the calculated Sec-OC (obtained based on the elemental carbon tracer method of Castro et al., 1999) at both sites $(r^2 = 0.84 \text{ and } 0.62 \text{ for North Kensington and Harwell respectively})$, confirming that this component is mainly secondary organics, and lesser but still significant correlations were observed for Other-OM with secondary inorganic components, sulphate and nitrate. A strong relationship was found for AMS OOA with the CMB Other-OM $(r^2 = 0.68)$, Sec-OC $(r^2 = 0.64)$ and inorganic components SO_4^{2-} and NO_3^{-} $(r^2 = 0.79 \text{ and } 0.80)$ as expected (Table 5).

Regression analyses showed low to moderate correlation between the measured biogenic secondary marker 2-methyltetrols and the calculated Sec-OC/CMB Other-OC ($r^2 = 0.25$ –0.41), whilst higher correlations were found between the measured α -pinene oxidation products and Sec-OC/CMB Other-OC ($r^2 = 0.31$ –0.82). Pinic acid particularly showed strong correlation with Sec-OC/CMB Other-OC (Fig. 6). In comparison, no correlation was found between 2-methyltetrols and AMS OOA showing close to zero coefficients, whilst pinic acid again exhibited a good relationship with the AMS OOA component ($r^2 = 0.55$) (Fig. 6). The results of Kleindienst et al. (2007) from measurements of VOC oxidation products at a US site attribute far greater importance to α -pinene than isoprene as an SOA precursor during the cooler months of the year. The species of trees and shrubs present locally will also be influential.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Full Screen / Esc

Close

Back

Printer-friendly Version



There are few previous published studies that have compared the results between the AMS-PMF and the CMB methods, and this first comparison study in the UK at the NK site has shown some inconsistent results for individual primary component estimates, and a different split between the total primary and secondary source components. In a study in Mexico, Aiken et al. (2009) found similar average OA/OM apportionment values from the two methods AMS-PMF and CMB for HOA/Vehicle (4.5 μg m⁻³, 28 %/4.5 μg m⁻³, 29 %), BBOA/Woodsmoke (2.7 μg m⁻³, 17 %/1.7 μg m⁻³, 12 %) and OOA/Other-OM (7.4 μg m⁻³, 46 %/9.2 μg m⁻³, 58 %), but the source components calculated were different to our study.

Generally speaking, overall correlations between the CMB and PMF based estimates here are very good, but the quantitative agreement is lacking, with PMF estimates generally greater than CMB for the primary species (in particular, cooking and solid fuel burning) and CMB assigning a much larger proportion of organic matter to secondary aerosol. Agreement between the methods is relatively good for the traffic source, and fair for food cooking (Table 6). Measurements of levoglucosan and fine potassium made during the campaign (Crilley et al., 2014) give an estimate for biomass burning particles consistent with the CMB results, and other work based upon elemental carbon data (arising mostly from diesel emissions) suggests that the traffic estimate in the CMB model is reasonable (Pant et al., 2014). The AMS estimates of SFOA and HOA also correlate very well with a Single Particle Soot Photometer (SP2) -based apportionment of the black carbon particles present, based on the mass of individual particles and coating thicknesses (Liu et al., 2014).

It is important to remember that the CMB model is applied to organic carbon, which is apportioned into the eight categories listed at the top of Table 4. These are then converted into source contribution estimates to $PM_{2.5}$ (Table 4) using conversion factors established in earlier work (Yin et al., 2010 and references therein). The conversion of organic carbon to $PM_{2.5}$ mass is to allow for other elements (H, N, O) associated with

ACPD

Discussion Paper

Discussion Paper

Discussion Pape

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

I

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Discussion Paper

Discussion Paper

Discussion

Pape

© **()**

carbon in the organic compounds, as well as other chemical constituents associated with that source. For example, in the case of road traffic exhaust, there is a component of elemental carbon which is included, and for dust/soil, major contributions from inorganic constituents of soil (Si, Al etc.) are included. On the other hand, the AMS is apportioning the mass of organic matter, and the other constituents are not included. Table 6 shows a comparison of the CMB with the AMS data, attempting where possible to express the CMB results as solely the organic matter content so as to be comparable with the AMS data. The AMS factors do not include vegetation, natural gas and dust/soil in their apportionments, however this is not unexpected; vegetation and dust/soil particles are generally too large to be detected by the AMS and natural gas does not contribute a sufficient mass of particulate organics to the overall loading.

There are also a number of technical reasons why both techniques may deliver inaccurate estimates. The sum of the two sets of measurements when expressed as organic matter is greater for the CMB than the AMS (Table 6). The CMB model is applied to samples of $PM_{2.5}$ whereas the AMS samples particles up to around 0.8 μ m, and consequently fails to sample larger particles, which are possibly of different composition. It is conceivable that the larger estimate of SOA by CMB may be caused by condensation of secondary material onto supermicron particles.

One issue that may affect the AMS is an ambiguous collection efficiency (CE). A time-dependent and composition-dependent parameterisation of CE was used, with a value of 0.5 used for most of the data, in line with the parameterisation of Middlebrook et al. (2012). However, it is possible that if a certain particulate population is externally mixed with the inorganic fraction in the accumulation mode, it may exhibit a different collection efficiency. While the overall CE was validated against a DMPS, it is possible that if a fraction makes a low enough contribution to the total volume concentration, it may not be picked up through this test. In the event that cooking particles are liquid, which is deemed likely given that many of the constituents such as oleic acid are of this phase at room temperature, their concentration could be overestimated by the AMS by up to a factor of 2. Note that this will only apply to particles that have not diffused

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I ✓ ▶I

Close

Full Screen / Esc

Back

Printer-friendly Version

Interactive Discussion

Pape

onto the accumulation mode, so the level of overestimation could be less than 2, even if the true CE of pure-component particles is unity. However, this could account for the majority of the discrepancies with the primary particles.

PMF analysis is subject to inherent uncertainties associated with rotational ambigu-5 ity (Allan et al., 2010). This can result in an amount of the mass being misattributed between factors and it is conceptually possible that some secondary material could be wrongly interpreted as solid fuel burning; indeed, there is evidence for this occurring to an extent in the CTOF instrument, which suffers from this phenomenon more than the HR-AMS used here (Young et al., 2014). Dall'Osto et al. (2014) showed that the COA factor from AMS data in Cork. Ireland exhibited an association with other urban aerosol sources, possibly indicating that it contained a contribution from these rather than cooking. These issues should be manifested as rotational freedoms within the solution sets and by varying the fpeak parameter, a tangible amount of uncertainty in the PMF outputs can be attributed to this (see Table S3) but this in itself is not large enough to explain the discrepancies. It is worth noting in particular that the ambiguity identified using this method consists mainly of a redistribution of mass between the primary factors, so would not explain in isolation an overestimation of both SFOA and COA. Nevertheless, it could be that this might explain at least part of the discrepancies reported.

There are a number of problems that may cause the CMB model to be inaccurate. Firstly, there is a general uncertainty surrounding how representative the source profiles assumed are of the aerosols encountered in this environment, but the sensitivity study of CMB profiles discussed above was intended to probe and minimise such effects. That said, it is recognised that the application of CMB to the secondary fraction is inherently highly uncertain, owing to the broad range of precursors and the complexity of the chemistry. It is also possible that some of the marker compounds are being lost from the aerosols between emission and measurement, either through repartitioning to the gas phase as the aerosol undergoes dilution in the atmosphere, chemical reactions through atmospheric processing, or evaporating from the filter during or after sampling.

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

Title Page **Abstract** Introduction Conclusions References **Tables Figures** Back Close

Printer-friendly Version

Conclusions

The CMB and AMS-PMF methods use entirely different processes to apportion organic carbon and organic matter respectively to source categories. The CMB method is able to attribute carbon to a larger number of sources, but depends upon prior knowledge of source profiles, which must add a significant element of uncertainty. The AMS-PMF method makes no a priori assumptions, but depends upon the PMF to separate components with many similarities in their mass spectra, and is able to apportion into a smaller number of classes.

The use of other marker elements/compounds to estimate source contributions from biomass burning and road traffic gives some confidence in the estimates from the CMB approach. It also attributes carbon to sources not recognised by AMS-PMF, which must be in some way included in the factors output by this method.

Although the average mass estimates for primary components differ appreciably between the CMB and AMS-PMF results, the estimated daily average concentrations for each generic source category show generally good correlations. In summary, the PMF estimations were higher than CMB by over a factor of two for solid fuel burning and cooking, slightly higher for traffic and lower for the secondaries. While no single issue with either technique can explain the discrepancies, they are within the scope of a combination of known problems and ambiguities (such as AMS collection efficiency, PMF rotation, inhomogeneous distribution of components across size fractions, Partisol collection artefacts and uncertainties in CMB profiles). Work needs to be performed to better constrain all of these issues.

The Supplement related to this article is available online at doi:10.5194/acpd-14-24523-2014-supplement.

Discussion

Paper

ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in **Southern England**

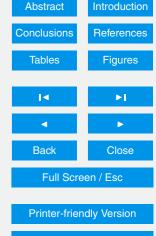
J. Yin et al.

Title Page

Discussion Paper

Discussion Pape

Interactive Discussion





Acknowledgements. Funding was received from the Natural Environment Research Council as part of the ClearfLo project (NERC NE/H003142/1). Raw data are available from the authors on request.

References

- Abdullahi, K. L., Delgado-Saborit, J. M., and Harrison, R. M.: Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: a review, Atmos. Environ., 71, 260–294, 2013.
 - Aiken, A. C., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D., Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner, E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L.: Mexico City aerosol analysis during MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) Part 1: Fine particle composition and organic source apportionment, Atmos. Chem. Phys., 9, 6633–6653, doi:10.5194/acp-9-6633-2009, 2009.
 - Allan, J. D., Williams, P. I., Morgan, W. T., Martin, C. L., Flynn, M. J., Lee, J., Nemitz, E., Phillips, G. J., Gallagher, M. W., and Coe, H.: Contributions from transport, solid fuel burning and cooking to primary organic aerosols in two UK cities, Atmos. Chem. Phys., 10, 647–668, doi:10.5194/acp-10-647-2010, 2010.
- Allen, A. G., Nemitz, E., Shi, J. P., Harrison, R. M., and Greenwood, J. C.: Size distributions of trace metals in atmospheric aerosols in the United Kingdom, Atmos. Environ., 35, 4581–4591, 2001.
 - Bigi, A. and Harrison, R. M.: Analysis of the air pollution climate at a central urban background site, Atmos. Environ., 44, 2004–2012, 2010.
- Birmili, W., Allen, A. G., Bary, F., and Harrison, R. M.: Trace metal concentrations and water solubility in size-fractionated atmospheric particles and influence of road traffic, Environ. Sci. Technol., 40, 1144–1153, 2006.
 - Böge, O., Miao, Y., Plewka, A., and Herrmann, H.: Formation of secondary organic particle phase compounds from isoprene gas-phase oxidation products: an aerosol chamber and field study, Atmos. Environ., 40, 2501–2509, 2006.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

I

I

Back Close

Printer-friendly Version

Full Screen / Esc



ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

- Title Page

 Abstract Introduction

 Conclusions References

 Tables Figures

 I

 I

 I

 Back Close

 Full Screen / Esc
 - Printer-friendly Version

Interactive Discussion

© BY

- Bressi, M., Sciare, J., Ghersi, V., Bonnaire, N., Nicolas, J. B., Petit, J.-E., Moukhtar, S., Rosso, A., Mihalopoulos, N., and Féron, A.: A one-year comprehensive chemical characterisation of fine aerosol (PM_{2.5}) at urban, suburban and rural background sites in the region of Paris (France), Atmos. Chem. Phys., 13, 7825–7844, doi:10.5194/acp-13-7825-2013, 2013.
- Buonanno, G., Morawska, L., and Stabile, L.: Particle emission factors during cooking activities, Atmos. Environ., 43, 3235–3242, 2009.
 - Canagaratna, M. R., Jayne, J. T., Jimenez, J. L., Allan, J. D., Alfarra, M. R., Zhang, Q., Onasch, T. B., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L. R., Trimborn, A. M., Northway, M. J., DeCarlo, P. F., Kolb, C. E., Davidovits, P., and Worsnop, D. R.: Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer, Mass Spectrom. Rev., 26, 185–222, doi:10.1002/Mas.20115, 2007.
 - Canonaco, F., Crippa, M., Slowik, J. G., Baltensperger, U., and Prévôt, A. S. H.: SoFi, an IGOR-based interface for the efficient use of the generalized multilinear engine (ME-2) for the source apportionment: ME-2 application to aerosol mass spectrometer data, Atmos. Meas. Tech., 6, 3649–3661, doi:10.5194/amt-6-3649-2013, 2013.
 - Castro, L. M., Pio, C. A., Harrison, R. M., and Smith, D. J. T.: Carbonaceous aerosol in urban and rural European atmospheres: Estimation of secondaryorganic carbon concentrations, Atmos. Environ., 33, 2771–2781, 1999.
 - Chan, Y. C., Simpson, R. W., McTainsh, G. H., Vowles, P. D., Cohen, D. D., and Bailey, G. M.: Characterisation of chemicalspecies in PM_{2.5} and PM₁₀ aerosols in Brisbane, Australia, Atmos. Environ., 31, 3773–3785, 1997.

- Charron, A., Degrendele, C., Laongsri, B., and Harrison, R. M.: Receptor modelling of secondary and carbonaceous particulate matter at a southern UK site, Atmos. Chem. Phys., 13, 1879–1894, doi:10.5194/acp-13-1879-2013, 2013.
- Claeys, M., Graham, B., Vas, G., Wang, W., Vermeylen, R., Pashynska, V., Cafmeyer, J., Guyon, P., Andreae, M. O., Artaxo, P., and Maenhaut, W.: Formation of secondary organic aerosols through photooxidation of isoprene, Science, 303, 1173–1176, 2004.
 - Clements, A. L. and Seinfeld, J. H.: Detection and quantification of 2-methyltetrols in ambient aerosol in the southeastern United States, Atmos. Environ., 41, 1825–1830, 2007.
- Clougherty, J. E., Houseman, E. A., and Levy, J. I.: Source apportionment of indoor residential fine particulate matter using land use regression and constrained factor analysis, Indoor Air, 21, 53–66, 2011.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

- Title Page

 Abstract Introduction

 Conclusions References

 Tables Figures

 I

 ▶ I

 Back Close

 Full Screen / Esc
 - Printer-friendly Version

Interactive Discussion



- Crilley, L. R., Bloss, W. J., Yin, J., Beddows, D. C. S., Harrison, R. M., Allan, J. D., Young, D. E., Flynn, M., Williams, P., Zotter, P., Prevot, A. S. H., Heal, M. R., Barlow, J. F., Halios, C. H., Lee, J. D., Helfter, C., Green, D., Szidat, S., and Mohr, C.: Sources and contributions of wood smoke during winter In London: assessing local and regional influences, Atmos. Chem. Phys., submitted, 2014.
- Dall'Osto, M., Hellebust, S., Healy, R. M., O'Connor, I. P., Kourtchev, I., Sodeau, J. R., Ovadnevaite, J., Ceburnis, D., O'Dowd, C. D., and Wenger, J. C.: Apportionment of urban aerosol sources in Cork (Ireland) by synergistic measurement techniques, Sci. Total Environ., 493, 197–208, 2014.
- DeCarlo, P. F., Kimmel, J. R., Trimborn, A., Northway, M. J., Jayne, J. T., Aiken, A. C., Gonin, M., Fuhrer, K., Horvath, T., Docherty, K. S., Worsnop, D. R., and Jimenez, J. L.: Field-deployable, high-resolution, time-of-flight aerosol mass spectrometer, Anal. Chem., 78, 8281–8289, doi:10.1021/ac061249n, 2006.
- Docherty, K. S., Stone, E. A., Ulbrich, I. M., DeCarlo, P. F., Snyder, D. C., Schauer, J. J., Peltier, R. E., Weber, R. J., Murphy, S. M., Seinfeld, J. H., Grover, B. D., Eatough, D. J., and Jimenez, J. L.: Apportionment of primary and secondary organic aerosols in Southern California during the 2005 study of organic aerosols in Riverside (SOAR-1), Environ. Sci. Technol., 42, 7655–7662, 2008.
- El Haddad, I., Marchand, N., Wortham, H., Piot, C., Besombes, J.-L., Cozic, J., Chauvel, C., Armengaud, A., Robin, D., and Jaffrezo, J.-L.: Primary sources of PM_{2.5} organic aerosol in an industrial Mediterranean city, Marseille, Atmos. Chem. Phys., 11, 2039–2058, doi:10.5194/acp-11-2039-2011, 2011a.

- El Haddad, I., Marchand, N., Temime-Roussel, B., Wortham, H., Piot, C., Besombes, J.-L., Baduel, C., Voisin, D., Armengaud, A., and Jaffrezo, J.-L.: Insights into the secondary fraction of the organic aerosol in a Mediterranean urban area: Marseille, Atmos. Chem. Phys., 11, 2059–2079, doi:10.5194/acp-11-2059-2011, 2011b.
- Fine, P. M., Cass, G. R., and Simoneit, B. R. T.: Chemical characterization of fine particle emissions from the fireplace combustion of wood types grown in the Midwestern and Western United States, Environ. Eng. Sci., 21, 387–409, 2004.
- Fraser, M. P., Yue, Z. W., and Buzcu, B.: Source apportionment of fine particulate matter in Houston, TX, using organic molecular markers, Atmos. Environ., 37, 2117–2123, 2003.
 - Gelencsér, A., May, B., Simpson, D., Sa'nchez-Ochoa, A., Kasper-Giebl, A., Puxbaum, H., Caseiro, A., Pio, C. A., and Legrand, M.: Source apportionment of PM_{2.5} organic aerosol over

/S.

Europe: primary/secondary, natural/anthropogenic, and fossil/biogenic origin, J. Geophys. Res., 112, D23S04, doi:10.1029/2006JD008094, 2007.

Guenther, A., Hewitt, C. N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., Mckay, W. A., Pierec, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., and Zimmerman, P.: A global model of natural volatile organic compounds emissions, J. Geophys. Res., 100, 8873–8892, 1995.

Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, Th. F., Monod, A., Prévôt, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmos. Chem. Phys., 9, 5155–5236, doi:10.5194/acp-9-5155-2009, 2009.

Harper, S. L., Walling, J. F., Holland, D. M., and Pranger, L. J.: Simplex optimization of multielement ultrasonic extraction of atmospheric particulates, Anal. Chem., 55, 1553–1557, 1983.

Harrison, R. M. and Yin, J.: Chemical speciation of PM_{2.5} particles at urban background and rural sites in the UK atmosphere, J. Environ. Monitor., 12, 1404–1414, 2010.

Harrison, R. M., Jones, A. M., and Lawrence, R. G.: A pragmatic mass closure model for airborne particulate matter at urban background and roadside sites, Atmos. Environ., 37, 4927–4933, 2003.

Harrison, R. M., Jones, A. M., and Lawrence, R. G.: Major component composition of PM₁₀ and PM_{2.5} from roadside and urban background sites, Atmos. Environ., 38, 4531–4538, 2004.

Harrison, R. M., Beddows, D. C. S., Hu, L., and Yin, J.: Comparison of methods for evaluation of wood smoke and estimation of UK ambient concentrations, Atmos. Chem. Phys., 12, 8271–8283, doi:10.5194/acp-12-8271-2012, 2012.

Hasheminassab, S., Daher, N., Schauer, J. J., and Sioutas, C.: Source apportionment and organic compound characterization of ambient ultrafine particulate matter (PM) in the Los Angeles Basin, Atmos. Environ., 79, 529–539, 2013.

He, L.-Y., Lin, Y., Huang, X.-F., Guo, S., Xue, L., Su, Q., Hu, M., Luan, S.-J., and Zhang, Y.-H.: Characterization of high-resolution aerosol mass spectra of primary organic aerosol emissions from Chinese cooking and biomass burning, Atmos. Chem. Phys., 10, 11535–11543, doi:10.5194/acp-10-11535-2010, 2010.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

I

I

Back Close

Printer-friendly Version

Full Screen / Esc



ACPD

14, 24523–24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

- Title Page

 Abstract Introduction

 Conclusions References

 Tables Figures

 I

 I

 Back Close

 Full Screen / Esc
 - Printer-friendly Version

Interactive Discussion

© () BY

- He, L. Y., Huang, X. F., Xue, L., Hu, M., Lin, Y., Zheng, J., Zhang, R., and Zhang, Y. H.: Submicron aerosol analysis and organic source apportionment in an urban atmosphere in Pearl River Delta of china using high-resolution aerosol mass spectrometry, J. Geophys. Res., 116, D12304, doi:10.1029/2010JD014566, 2011.
- Heal, M. R., Naysmith, P., Cook, G. T., Xu, S., Raventos Duran, T., and Harrison, R. M.: Application of ¹⁴C analyses to source apportionment of carbonaceous PM_{2.5} in the UK, Atmos. Environ., 45, 2341–2348, 2011.
 - Huang, X.-F., He, L.-Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L., Zeng, L.-W., Liu, X.-G., Zhang, Y.-H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.: Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer, Atmos. Chem. Phys., 10, 8933–8945, doi:10.5194/acp-10-8933-2010, 2010.

10

- Hyder, M., Genberg, J., and Jönsson, J. Å.: Application of hollowfiberliquid phase microextraction for pinicacid and pinonicacidanalysisfromorganicaerosols, Anal. Chim. Acta, 713, 79–85, 2012.
- Kamens, R., Lee, C. T., Wiener, R., and Leith, D.: A study of characterize indoor particles in three non-smoking homes, Atmos. Environ. A-Gen., 25, 939–948, 1991.
- Kleeman, M. J., Robert, M. A., Riddle, S. G., Fine, P. M., Hays, M. D., Schauer, J. J., and Hannigan, M. P.: Size distribution of trace organic species emitted from biomass combustion and meat charbroiling, Atmos. Environ., 42, 3059–3075, 2008.
- Kleindienst, T. E, Jaoui, M., Lewandowski, M., Offenberg, J. H., Lewis, C. W., Bhave, P. V., and Edney, E. O.: Estimates of the contributions of biogenic and anthropogenic hydrocarbons to secondary organic aerosolat a southeastern US location, Atmos. Environ., 41, 8288–8300, 2007.
- Kourtchev, I., Ruuskanen, T., Maenhaut, W., Kulmala, M., and Claeys, M.: Observation of 2-methyltetrols and related photo-oxidation products of isoprene in boreal forest aerosols from HyytiKourtchev, I., Ruuskanen, T., Maenhaut, W., Kulmala, M., and Claeys, M.: Observation of 2-methyltetrols and related photo-oxidation products of isoprene in boreal forest aerosols from Hyytiälä, Finland, Atmos. Chem. Phys., 5, 2761–2770, doi:10.5194/acp-5-2761-2005, 2005.
- Kourtchev, I., Copolovici, L., Claeys, M., and Maenhaut, W.: Characterization of Atmospheric aerosols at a forested site in central Europe, Environ. Sci. Technol., 43, 4665–4671, 2009.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

- Title Page

 Abstract Introduction

 Conclusions References

 Tables Figures

 I ← ►I

 ← ► Back Close

 Full Screen / Esc
- Printer-friendly Version

Interactive Discussion

© **()**

- Lanz, V. A., Alfarra, M. R., Baltensperger, U., Buchmann, B., Hueglin, C., Szidat, S., Wehrli, M. N., Wacker, L., Weimer, S., Caseiro, A., Puxbaum, H., and Prevot, A. S. H.: Source attribution of submicron organic aerosols during wintertime inversions by advanced factor analysis of aerosol mass spectra, Environ. Sci. Technol., 42, 214–220, doi:10.1021/Es0707207, 2008.
- Lee, H., Park, S. S., Kim, K. W., and Kim, Y. J.: Source identification of PM_{2.5} particles measured in Gwangju, Korea, Atmos. Res., 88, 199–211, 2008.
- Lewandowski, M., Jaoui, M., Offenberg, J. H., Kleindienst, T. E., Edney, E. O., Sheesley, R. J., and Schauer, J. J.: Primary and secondary contributions to ambient PM in the Midwestern United States, Environ. Sci. Technol., 42, 3303–3309, 2008.
- Liu, D., Allan, J. D., Young, D. E., Coe, H., Beddows, D., Fleming, Z. L., Flynn, M. J., Gallagher, M. W., Harrison, R. M., Lee, J., Prevot, A. S. H., Taylor, J. W., Yin, J., Williams, P. I., and Zotter, P.: Size distribution, mixing state and source apportionment of black carbon aerosol in London during wintertime, Atmos. Chem. Phys., 14, 10061–10084, doi:10.5194/acp-14-10061-2014, 2014.
- Lough, G. C., Schauer, J. J., Tortorelli, J., Mani, E., Lawson, D. R., Clark, N. N., and Gabele, P. A.: Development of molecular marker source profiles for emissions from on-road gasoline and diesel vehicle fleets, JAWMA, 57, 1190–1199, 2007.
- Maenhaut, W., Vermeylen, R., Claeys, M., Vercauteren, J., Matheeussen, C., and Roekens, E.: Assessment of the contribution from wood burning to the PM₁₀ aerosol in Flanders, Belgium, Sci. Total Environ., 437, 226–236, 2012.

- Massey, D., Kulshrestha, A., Masih, J., and Taneja, A.: Seasonal trends of PM_{10} , $PM_{5.0}$, $PM_{2.5}$ and $PM_{1.0}$ in indoor and outdoor environments of residential homes located in North-Central India, Build. Environ., 47, 223–231, 2012.
- Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of composition-dependent collection efficiencies for the aerodyne aerosol mass spectrometer using field data, Aerosol Sci. Tech., 46, 258–271, doi:10.1080/02786826.2011.620041, 2012.
 - Mohr, C., Huffman, J. A., Cubison, M. J., Aiken, A. C., Docherty, K. S., Kimmel, J. R., Ulbrich, I. M., Hannigan, M., and Jimenez, J. L.: Characterization of primary organic aerosol emissions from meat cooking, trash burning, and motor vehicles with high-resolution aerosol mass spectrometry and comparison with ambient and chamber observations, Environ. Sci. Technol., 43, 2443–2449, 2009.

- 14
 - 14, 24523-24572, 2014

ACPD

Receptor modelling of fine particles in Southern England

J. Yin et al.

- - Printer-friendly Version

Full Screen / Esc



- Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U., and Prévôt, A. S. H.: Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data, Atmos. Chem. Phys., 12, 1649–1665, doi:10.5194/acp-12-1649-2012, 2012.
- Paatero, P.: Least squares formulation of robust non-negative factor analysis, Chemometr. Intell. Lab., 37, 23–35, 1997.
- Paatero, P.: The multilinear engine a table-driven, least squares program for solving multilinear problems, including the n-way parallel factor analysis model, J. Comput. Graph. Stat., 8, 854–888, 1999.
- Paatero, P., Hopke, P. K., Song, X. H., and Ramadan, Z.: Understanding and controlling rotations in factor analytic models, Chemometr. Intell. Lab., 60, 253–264, doi:10.1016/S0169-7439(01)00200-3, 2002.
- Pancras, J. P., Landis, M. S., Norris, G. A., Vedantham, R., and Dvonch, J. T.: Source apportionment of ambient fine particulate matter in Dearborn, Michigan using hourly resolved PM chemical composition data, Sci. Total Environ., 448, 2–13, 2013.
- Pant, P. and Harrison, R. M.: Estimation of the contribution of road traffic emissions to particulate matter concentrations from field measurements: a review, Atmos. Environ., 77, 78–97, 2013.
- Pant, P., Yin, J., and Harrison, R. M.: Sensitivity of a chemical mass balance model to different molecular marker traffic source profiles, Atmos. Environ., 82, 238–249, 2014.
 - Pio, C., Cerqueira, M., Harrison, R. M., Nunes, T., Mirante, F., Alves, C., Oliveira, C., Sanchez de la Campa, A., Artinano, B., and Matos, M.: OC/EC ratio observations in europe: re-thinking the approach for apportionment between primary and secondary organic carbon, Atmos. Environ., 45, 6121–6132, 2011.
- Plewka, A., Gnauk, T., Brüggemann, E., and Herrmann, H.: Biogenic contributions to the chemical composition of airborne particles in a coniferous forest in Germany, Atmos. Environ., 40, S103–S115, 2006.
- Presto, A. A., Huff-Hartz, K. E., and Donahue, N. M.: Secondary organic aerosol production from terpeneozonolysis. 1., Effect of UV radiation, Environ. Sci. Technol., 39, 7036–7045, 2005.
- Puxbaum, H., Caseiro, A., Sanchez-Ochoa, A., Kasper-Giebl, A., Claeys, M., Gelencser, A., Legrand, M., Preunkert, S., and Pio, C.: Levoglucosan levels at background sites in Europe

Paper

14, 24523-24572, 2014

ACPD

Receptor modelling of fine particles in Southern England

J. Yin et al.

- Title Page

 Abstract Introduction

 Conclusions References

 Tables Figures

 I ◀ ▶I

 Back Close
 - Printer-friendly Version

Full Screen / Esc



- for assessing the impact of biomass combustion on the European aerosol background, J. Geophys. Res., 112, D23S05, doi:10.1029/2006JD008114, 2007.
- Robinson, A. L., Subramanlan, R., Donahue, N. M., Bernardo-Bricker, A., and Rogge, W. F.: Source apportionment of molecular markers and organic aerosol. 3. Food cooking emissions, Environ. Sci. Technol., 40, 7820–7827, 2006.
- Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., and Simoneit, B. R. T.: Sources of fine organic aerosol. 4. Particulate abrasion products from leaf surfaces of urban plants, Environ. Sci. Technol., 27, 2700–2711, 1993a.
- Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., and Simoneit, B. R. T.: Sources of fine organic aerosol. 5. Natural-gas home appliances, Environ. Sci. Technol., 27, 2736–2744, 1993b.
- Schauer, J. J., Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., and Simoneit, B. R. T.: Source apportionment of airborne particulate matter using organic compounds as tracers, Atmos. Environ., 30, 3837–3855, 1996.
- Schauer, J. J.: Source Contributions to Atmospheric Organic Compound Concentrations: Emissions Measurement and Model Predictions, Environmental Engineering Science, California Institute of Technology, Pasadena, P400, 1998.
 - Schauer, J. J., Kleeman, M. J., Cass, G. R., and Simoneit, B. R. T.: Measurement of emissions from air pollution sources. 2. C₁ through C₃₀ organic compounds from medium duty diesel trucks, Environ. Sci. Technol., 33, 1578–1587, 1999.
 - Schauer, J. J., Fraser, M. P., Cass, G. R., and Simoneit, B. R. T.: Source reconciliation of atmospheric gas-phpase and particle-phase pollutants during severe photochemical smog episode, Environ. Sci. Technol., 26, 3806–3814, 2002.
 - Stone, E. A., Snyder, D. C., Sheesley, R. J., Sullivan, A. P., Weber, R. J., and Schauer, J. J.: Source apportionment of fine organic aerosol in Mexico City during the MILAGRO experiment 2006, Atmos. Chem. Phys., 8, 1249–1259, doi:10.5194/acp-8-1249-2008, 2008.
 - Stone, E. A., Zhou, J., Snyder, D. C., Rutter, A. P., Mieritz, M., and Schauer, J. J.: A comparison of summertime secondary organic aerosol source contributions at contrasting urban locations, Environ. Sci. Technol., 43, 3448–3454, 2009.
- Sun, Y.-L., Zhang, Q., Schwab, J. J., Demerjian, K. L., Chen, W.-N., Bae, M.-S., Hung, H.-M., Hogrefe, O., Frank, B., Rattigan, O. V., and Lin, Y.-C.: Characterization of the sources and processes of organic and inorganic aerosols in New York city with a high-resolution time-of-

- ACPD
- 14, 24523–24572, 2014
- Receptor modelling of fine particles in Southern England
 - J. Yin et al.
- Title Page

 Abstract Introduction

 Conclusions References

 Tables Figures

 I

 I

 Back Close

 Full Screen / Esc
- Printer-friendly Version
 - Interactive Discussion
 - © O

- flight aerosol mass apectrometer, Atmos. Chem. Phys., 11, 1581–1602, doi:10.5194/acp-11-1581-2011, 2011.
- Thorpe, A. and Harrison, R. M.: Sources and properties of non-exhaust particulate matter from road traffic: a review, Sci. Total Environ., 400, 270–282, 2008.
- ⁵ Turpin, B. J., Saxena, P., and Andrews, E.: Measuring and simulating particulate organics in the atmosphere: problems and prospects, Atmos. Environ., 34, 2983–3013, 2000.
 - Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, Atmos. Chem. Phys., 9, 2891–2918, doi:10.5194/acp-9-2891-2009, 2009.
 - Utembe, S. R., Watson, L. A., Shallcross, D. E., and Jenkin, M. E.: A common representative intermediates (CRI) mechanism for VOC degradation, Part 3: development of a secondary organic aerosol module, Atmos. Environ., 43, 1982–1990, 2009.
 - Wagener, S., Langner, M., Hansen, U., Moriske, H. J., and Endlicher, W. R.: Spatial and seasonal variations of biogenic tracer compounds in ambient PM₁₀ and PM₁ samples in Berlin, Germany, Atmos. Environ., 47, 33–42, 2012a.
 - Wagener, S., Langner, M., Hansen, U., Moriske, H. J., and Endlicher, W. R.: Source apportionment of organic compounds in Berlin using positive matrix factorization assessing the impact of biogenic aerosol and biomass burning on urban particulate matter, Sci. Total Environ., 435–436, 392–401, 2012b.
 - Wan, M. P., Wu, C. L., Sze To, G. N., Chan, T. C., and Chao, C. Y. H.: Ultrafine particles and PM_{2.5} generated from cooking in homes, Atmos. Environ., 45, 6141–6148, 2011.
 - Wang, Q., Shao, M., Zhang, Y., Wei, Y., Hu, M., and Guo, S.: Source apportionment of fine organic aerosols in Beijing, Atmos. Chem. Phys., 9, 8573–8585, doi:10.5194/acp-9-8573-2009, 2009.
- Williams, B. J., Goldstein, A. H., Kreisberg, N. M., Hering, S. V., Worsnop, D. R., Ulbrich, I. M., Docherty, K. S., and Jimenez, J. L.: Major components of atmospheric organic aerosol in southern California as determined by hourly measurements of source marker compounds, Atmos. Chem. Phys., 10, 11577–11603, doi:10.5194/acp-10-11577-2010, 2010.
 - Yin, J., Allen, A. G., Harrison, R. M., Jennings, S. G., Wright, E., Fitzpatrick, M, Healy, T., Barry, E., Ceburnis, D., and McCusker, D.: Major component composition of urban PM₁₀ and PM_{2.5} in Ireland, Atmos. Res., 78, 149–165, 2005.

- ACPD
- 14, 24523-24572, 2014
- Receptor modelling of fine particles in Southern England
 - J. Yin et al.
- Title Page

 Abstract Introduction

 Conclusions References

 Tables Figures

 I ← ►I

 Back Close

 Full Screen / Esc
 - Printer-friendly Version
 - Interactive Discussion
 - © BY

- Yin, J., Harrison, R. M., Chen, Q., Rutter, A., and Schauer, J. J.: Source apportionment of fine particles at urban background and rural sites in the UK tmosphere, Atmos. Environ., 44, 841–851, 2010.
- Young, D. E., Allan, J. D., Williams, P. I., Green, D. C., Flynn, M. J., Harrison, R. M., Yin, J., Gallagher, M. W., and Coe, H.: Investigating the annual behaviour of submicron secondary inorganic and organic aerosols in London, Atmos. Chem. Phys. Discuss., 14, 18739–18784, doi:10.5194/acpd-14-18739-2014, 2014.
- Yue, Z. and Fraser, M. P.: Polar organic compounds measured in fine particulate matter during TexAZS 2000, Atmos. Environ., 38, 3253–3261, 2004.
- Zhang, Q., Jimenez, J., Ganagaratna, M., Ulbrich, I., Ng, N., Worsnop, D., and Sun, Y.: Understanding atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, Anal. Bioanal. Chem., 401, 3045–3067, 2011.
- Zhang, X., Hecobian, A., Zheng, M., Frank, N. H., and Weber, R. J.: Biomass burning impact on PM_{2.5} over the southeastern US during 2007: integrating chemically speciated FRM filter measurements, MODIS fire counts and PMF analysis, Atmos. Chem. Phys., 10, 6839–6853, doi:10.5194/acp-10-6839-2010, 2010.
- Zhang, Y. X., Schauer, J. J, Zhang, Y. H., Shao, M., Wie, Y. J., Liu, Y., and Zeng, L. M.: Characteristics of particulate carbon emissions from real world Chinese coal combustion, Environ. Sci. Technol., 42, 5068–5073, 2008.
- Zhao, Y., Hu, M., Slanina, S., and Zhang, Y.: Chemical compositions of fine particulate organic matter emitted from Chinese cooking, Environ. Sci. Technol., 41, 99–105, 2007a.
 - Zhao, Y., Hu, M., Slanina, S., and Zhang, Y.: The molecular distribution of fine particulate organic matter emitted from Western-style fast food cooking, Atmos. Environ., 41, 8163–8171, 2007b.
- Zheng, M., Cass, G. R., Schauer, J. J., and Edgerton, E. S.: Source apportionment of PM_{2.5} in the southeastern United States using solvent-extractable organic compounds as tracers, Environ. Sci. Technol., 36, 2361–2371, 2002.

Receptor modelling of fine particles in

ACPD

14, 24523-24572, 2014

J. Yin et al.

Title Page

Southern England

| Tille Lage | | | | | |
|-------------|--------------|--|--|--|--|
| Abstract | Introduction | | | | |
| Conclusions | References | | | | |
| Tables | Figures | | | | |
| . I∢ | ►I | | | | |
| • | • | | | | |
| Back | Close | | | | |

Printer-friendly Version

Full Screen / Esc



Table 1. Air sampling and analytical instrumentation.

| Site | Instrument/ Filter media | Particle size | Chemical analyses /Measurements | Analyser/ Methods | Sample inter- vals |
|------|-----------------------------|-------------------|--|--------------------------|-----------------------|
| NK | Partisol/ PTFE filter | PM _{2.5} | PM mass Al, Si, Fe Ti, Mn, Ni, Cu, Zn, Ba | Balance XRF ICP-MS | Daily |
| | Digitel/ Quartz filter | PM _{2.5} | Organic markers (including secondary) | GCMS | Daily |
| | | | OC & EC SO ₄ ²⁻ , NO ₃ ⁻ , Cl ⁻ , Na ⁺ , K ⁺ , Mg ²⁺ , Ca ²⁺ , NH ₄ ⁴ | OCEC analyser Dionex | |
| | TOF-AMS | $PM_{0.8}$ | Mass size segregated Organic aerosol OOA, BBOA, HOA, COA | PMF | 1–10 min |
| HAR | Partisol/ PTFE filter | PM _{2.5} | PM mass Al, Si, Fe Ti, Mn, Ni, Cu, Zn, Ba | Balance XRF ICP-MS | Daily |
| | Digitel/ Quartz filter | PM _{2.5} | Organic markers (not including secondary) | GCMS | Daily |
| | | | OC & EC SO ₄ ²⁻ , NO ₃ ⁻ , CI ⁻ , Na ⁺ , K ⁺ , Mg ²⁺ , Ca ²⁺ , NH ₄ ⁺ | OCEC analyser Dionex | |

Discussion Paper

ACPD

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

| Title Page | | | | | |
|-------------------|--------------|--|--|--|--|
| Abstract | Introduction | | | | |
| Conclusions | References | | | | |
| Tables | Figures | | | | |
| | | | | | |
| . I◀ | ►I | | | | |
| • | • | | | | |
| Back | Close | | | | |
| Full Screen / Esc | | | | | |

Printer-friendly Version

Interactive Discussion



Table 2. Concentration summary of measured components at NK and HAR.

| | | N | | HAR | | | |
|-------------------------------|-------|--------|-----------------|-------|--------|-----------------|--|
| Components | Mean | Median | 10 %ile/90 %ile | Mean | Median | 10 %ile/90 %ile | |
| PM _{2.5} | 15.7 | 12.6 | 4.9/29.8 | 11.0 | 10.2 | 3.1/19.6 | |
| $(\mu g m^{-3})$ | | | | | | | |
| ÖC | 3.5 | 2.6 | 1.3/6.7 | 2.3 | 2.1 | 0.46/5.0 | |
| EC | 1.5 | 1.2 | 0.67/2.7 | 0.68 | 0.56 | 0.12/1.3 | |
| CI ⁻ | 0.67 | 0.60 | 0.33/1.3 | 0.50 | 0.45 | 0.23/0.76 | |
| SO ₄ ²⁻ | 1.8 | 1.3 | 0.58/3.8 | 1.7 | 1.3 | 0.46/4.4 | |
| $NO_3^{\frac{4}{3}}$ | 3.5 | 2.6 | 0.69/7.6 | 3.2 | 2.7 | 0.42/6.3 | |
| Al | 0.044 | 0.035 | 0.017/0.086 | 0.027 | 0.019 | 0.008/0.060 | |
| Si | 0.14 | 0.12 | 0.056/0.25 | 0.077 | 0.056 | 0.023/0.15 | |
| Levo | 73.9 | 69.5 | 42.7/118 | 94.5 | 99.0 | 27.2/152 | |
| $(ng m^{-3})$ | | | | | | | |
| C ₂₉ | 2.1 | 1.7 | 1.2/4.0 | 3.7 | 3.6 | 1.6/5.4 | |
| C ₃₁ | 1.7 | 1.3 | 0.84/3.1 | 2.9 | 3.0 | 0.93/4.3 | |
| C ₃₃ | 0.95 | 0.75 | 0.43/2.0 | 1.2 | 1.2 | 0.54/2.0 | |
| C ₃₅ | 0.58 | 0.47 | 0.28/1.1 | 3.1 | 3.5 | 0.4/4.3 | |
| 17α TNohop | 0.25 | 0.19 | 0.14/0.36 | 0.079 | 0.075 | 0.042/0.12 | |
| $17\beta\alpha$ Nohop | 0.50 | 0.41 | 0.24/0.84 | 0.36 | 0.36 | 0.25/0.47 | |
| 17 <i>αβ</i> Hop | 0.33 | 0.26 | 0.20/0.39 | 0.17 | 0.16 | 0.14/0.22 | |
| B(k)F | 0.67 | 0.50 | 0.15/1.4 | 0.49 | 0.38 | 0.053/1.0 | |
| B(b)F | 0.54 | 0.39 | 0.15/1.1 | 0.51 | 0.48 | 0.12/0.94 | |
| B(e)P | 0.48 | 0.35 | 0.14/0.94 | 0.33 | 0.29 | 0.048/0.66 | |
| IP | 0.40 | 0.28 | 0.11/0.84 | 0.29 | 0.23 | 0.041/0.62 | |
| PIC | 0.10 | 0.081 | 0.045/0.17 | 0.044 | 0.034 | 0.005/0.10 | |
| B(ghi)PER | 0.47 | 0.35 | 0.15/0.94 | 0.25 | 0.20 | 0.039/0.52 | |
| PalmA | 60.2 | 50.1 | 29.0/110 | 19.0 | 13.7 | 7.2/34.5 | |
| LinoA | 7.9 | 2.0 | 0.17/14.2 | 6.7 | 0.66 | 0.31/6.3 | |
| OleiA | 11.8 | 2.3 | 1.1/22.4 | 3.0 | 1.6 | 0.66/3.2 | |
| SteaA | 26.6 | 22.7 | 12.6/43.1 | 10.7 | 7.3 | 5.4/18.7 | |
| MethT | 0.19 | 0.13 | 0.05/0.27 | _ | _ | _ | |
| MethE | 0.31 | 0.26 | 0.12/0.39 | _ | _ | _ | |
| PinoA | 1.3 | 0.96 | 0.55/2.7 | _ | _ | _ | |
| PinicA | 0.94 | 0.56 | 0.11/2.5 | _ | _ | _ | |

Note: PM_{2.5} mass and inorganic constituents in μg m⁻³; Organic markers in ng m⁻³.

Table 3. Comparison of data for mean concentrations of secondary biogenic molecular markers.

| Site | Season/ Fraction | 2-Methyl -threitol | 2-Methyl -erythritol | 2-Methyl -tetrols | Pinonic acid | Pinic acid | Pinonic + Pinic acids | References |
|--|------------------------------------|-----------------------|-------------------------|----------------------|-----------------|---------------|-----------------------------|----------------------------------|
| NK, London, UK, urban back- ground | Jan-Feb 2012/ PM _{2.5} | 0.19 | 0.31 | 0.50 | 1.3 | 0.94 | 2.3 | _ |
| HV (high vegetation site), Berlin, Germany | Feb-Oct 2010/ PM ₁₀ | 8.0 | 1.4 | 2.2 | 13.2 | 3.9 | 17.1 | Wagener et al., 2012a, b |
| LV (roadside, low vegetation), Berlin, Germany | Feb-Oct 2010/ PM ₁₀ | 0.6 | 1.2 | 1.8 | 8.4 | 2.2 | 10.6 | Wagener et al., 2012a, b |
| RegV (background, regional influence), Berlin, Germany | Feb-Oct 2010/ PM ₁₀ | 0.8 | 1.2 | 2.0 | 15.3 | 5.6 | 20.9 | Wagener et al., 2012a, b |
| HV (high vegetation site), Berlin, Germany | Feb-Oct 2010/ PM ₁ | 0.6 | 1.1 | 1.7 | 11.9 | 3.1 | 15.0 | Wagener et al., 2012a, b |
| LV (roadside, low vegetation), Berlin, Germany | Feb-Oct 2010/ PM ₄ | 0.3 | 0.8 | 1.1 | 6.9 | 2.3 | 9.2 | Wagener et al., 2012a, b |
| Vavihill, background, southern Sweden | 2008–2009/ PM ₁₀ | _ | - | _ | 3.02 | 3.03 | 6.1 | Hyder et al., 2012 |
| Hyytiälä, Finland, rural with forests surrounded | summer 2004/ PM ₄ | 5.1 | 21.2 | 26.3 | - | - | - | Kourtchev et al., 2005 |
| Hyytiälä, Finland, rural with forests surrounded | autumn 2004/ PM ₁ | 0.18 | 0.29 | 0.47 | _ | - | _ | Kourtchev et al., 2005 |
| Southeastern US, urban & urban background | Jun 2004 | 4.8 | 11.9 | 16.7 | _ | - | _ | Clements and Se- infeld, 2007 |
| Southeastern US, rural | Jun 2004 | 1.6 | 4.9 | 6.5 | - | - | - | Clements and Se- infeld, 2007 |

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

■ Back Close

Full Screen / Esc

Printer-friendly Version



Table 4. Source contribution estimates (SCE) ($\mu g \, m^{-3}$) and standard deviation (S.D.) for fine particulate OC and PM_{2.5} at NK and HAR from the CMB model.

| | | | ОС | | | PM _{2.5} | | OC/PM _{2.5} or |
|-------------------------------|------|--------|--------|------------------|--------|-------------------|-------|-------------------------|
| Source Name | | NK^a | NK^b | HAR ^a | NK^a | NKb | HAR | OC/OM CF° |
| Vegetation | SCE | 0.069 | 0.069 | 0.11 | 0.21 | 0.21 | 0.35 | 0.324 |
| | S.D. | 0.010 | 0.010 | 0.015 | 0.030 | 0.030 | 0.048 | _ |
| Woodsmoke | SCE | 0.53 | 0.53 | 0.64 | 0.64 | 0.64 | 0.76 | 0.836 |
| | S.D. | 0.11 | 0.11 | 0.14 | 0.14 | 0.14 | 0.16 | _ |
| Natural Gas | SCE | 0.046 | 0.046 | 0.042 | 0.054 | 0.054 | 0.049 | 0.849 |
| | S.D. | 0.009 | 0.009 | 0.007 | 0.011 | 0.011 | 0.008 | _ |
| Dust/Soil | SCE | 0.044 | 0.044 | 0.016 | 0.34 | 0.34 | 0.12 | 0.131 |
| | S.D. | 0.036 | 0.036 | 0.015 | 0.27 | 0.27 | 0.11 | _ |
| Coal | SCE | 0.074 | 0.074 | 0.041 | 0.17 | 0.17 | 0.094 | 0.432 |
| | S.D. | 0.020 | 0.020 | 0.009 | 0.046 | 0.046 | 0.021 | _ |
| Food Cooking | SCE | 0.39 | 0.39 | 0.072 | 0.69 | 0.69 | 0.13 | 0.566 |
| · · | S.D. | 0.066 | 0.066 | 0.013 | 0.12 | 0.12 | 0.023 | _ |
| Total Traffic | SCE | 0.73 | 0.73 | 0.36 | 1.26 | 1.26 | 0.61 | 0.579 |
| | S.D. | 0.35 | 0.35 | 0.16 | 0.86 | 0.86 | 0.29 | _ |
| Biogenic Secondary | SCE | _ | 0.90 | _ | _ | 1.63 | _ | 0.556 |
| , | S.D. | _ | 0.17 | _ | _ | 0.31 | _ | _ |
| Other-OC/OM | SCE | 1.64 | 0.73 | 1.02 | 2.95 | 1.32 | 1.84 | 0.556 |
| | S.D. | _ | _ | _ | _ | _ | _ | _ |
| Sea Salt | SCE | _ | _ | _ | 1.1 | 1.1 | 0.82 | _ |
| | S.D. | _ | _ | _ | 0.020 | 0.020 | 0.020 | _ |
| Ammonium Sulphate | SCE | _ | _ | _ | 2.2 | 2.2 | 2.1 | _ |
| | S.D. | _ | _ | _ | 0.028 | 0.028 | 0.028 | _ |
| Ammonium Nitrate | SCE | _ | _ | _ | 5.8 | 5.8 | 4.1 | _ |
| | S.D. | _ | _ | _ | 0.072 | 0.072 | 0.072 | _ |
| Measured OC/PM _{2.5} | Mass | 3.5 | 3.5 | 2.3 | 15.7 | 15.7 | 11.0 | _ |

Note: Figures in bold were not statistically different from zero; a - Modelled without biogenic secondary source profile; b - Modelled with biogenic secondary source profile; c - Conversion factor

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

I

I

Back Close

Full Screen / Esc

Printer-friendly Version



Table 5. RMA regression results for CMB and AMS-PMF organic matter estimates and related variables.

| | | NK2012 | |
|---|-------|-----------|-------|
| Pair of Variables | Slope | Intercept | r^2 |
| AMS-SFOA/CMB-WS | 2.81 | -0.69 | 0.75 |
| AMS-SFOA/CMB-(WS+Coal) | 2.40 | -0.64 | 0.78 |
| AMS-COA/CMB Food Cooking | 2.24 | -0.33 | 0.80 |
| AMS-HOA/CMB Traffic | 1.24 | -0.32 | 0.80 |
| AMS (SFOA+COA+HOA)/CMB (WS+Coal+Food+Traffic) | 1.85 | -1.14 | 0.91 |
| AMS-OOA/CMB Other-OM | 0.39 | -0.06 | 0.69 |
| AMS-OOA/Sec-OC | 0.81 | 0.18 | 0.64 |
| AMS OOA/Measured SO ₄ ²⁻ | 0.71 | -0.27 | 0.79 |
| AMS OOA/Measured NO3 | 0.33 | -0.16 | 0.80 |
| CMB Other-OC/Sec-OC | 0.99 | 0.66 | 0.84 |
| CMB Other-OM/Measured SO ₄ ²⁻ | 1.13 | 0.78 | 0.39 |
| CMB Other-OM/Measured NO_3^{-} | 0.58 | 0.84 | 0.42 |

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Full Screen / Esc

Printer-friendly Version



Table 6. Comparison of apportionment of organic matter by the AMS-PMF and CMB method at NK ($\mu g \, m^{-3}$).

| Source | CMB Estimate | AMS-PMF Estimate |
|-----------------|--------------|------------------|
| Biomass burning | 0.85 | 1.63 |
| Coal | 0.12 | _ |
| Food cooking | 0.56 | 0.87 |
| Traffic | 0.98 | 0.86 |
| Vegetation | 0.11 | _ |
| Natural gas | 0.055 | _ |
| Dust/soil | 0.10 | _ |
| Primary-total | 2.77 | 3.36 |
| Secondary | 2.92 | 0.99 |
| TOTAL | 5.69 | 4.35 |

Receptor modelling of fine particles in Southern England

J. Yin et al.

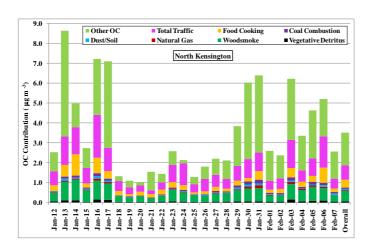
Full Screen / Esc

Close

Back

Printer-friendly Version





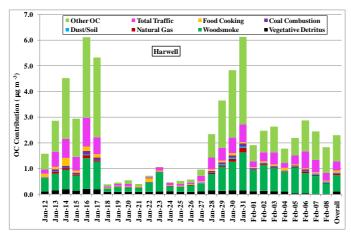


Figure 1. Daily OC source contributions at NK and HAR.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.



Printer-friendly Version





Discussion Paper

Discussion Paper

Printer-friendly Version

Interactive Discussion



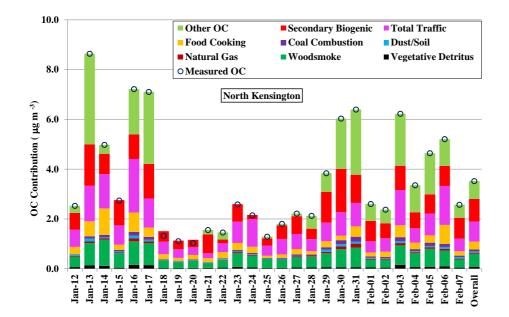


Figure 2. Daily OC source contribution estimates with secondary biogenic components at NK.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.

■ Coal

Combustion
Dust/Soil

■ Natural Gas

■ Woodsmoke

■ Vegetative

Detritus

OMeasured PM2.5

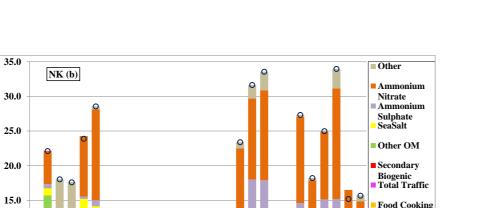


Figure 3. Daily $PM_{2.5}$ source contribution estimates with secondary biogenic components at NK.

Jan-28 Jan-29 Jan-30 Jan-31 Feb-01 Feb-02 Feb-03

Jan-22 Jan-23 Jan-24 Jan-25 Jan-26

PM2.5 Contribution (µg m ³)

5.0

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I

I

I

Back Close

Full Screen / Esc

Printer-friendly Version





ACPD

Receptor modelling of fine particles in **Southern England**

J. Yin et al.



Printer-friendly Version Interactive Discussion



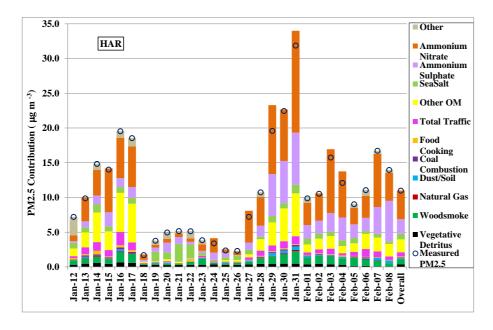


Figure 4. Daily PM_{2.5} source contribution estimates at HAR.

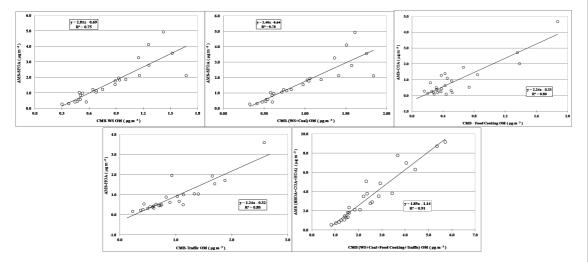


Figure 5. Primary component comparison between the AMS-PMF and CMB methods at NK.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in Southern England

J. Yin et al.





Printer-friendly Version



Discussion Paper

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



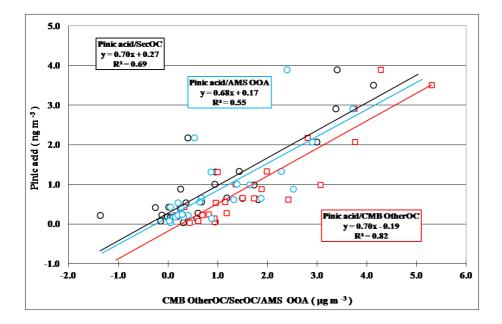


Figure 6. Secondary component comparison at NK showing relationships between pinic acid concentrations and estimated secondary OC from the elemental carbon tracer method (Sec-OC) and the CMB model (Other-OC), and with the AMS OOA factor.

ACPD

14, 24523-24572, 2014

Receptor modelling of fine particles in **Southern England**

J. Yin et al.