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Receptor modelling
of fine particles in
Southern England

J. Yin et al.

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

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

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 re-

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



sults were highly correlated ($r^2 = 0.69\text{--}0.91$). 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 $\text{PM}_{2.5}$ (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 $\text{PM}_{2.5}$ 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 ($\text{PM}_{2.5}$) 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,

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



**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 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., 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 2-methylerythritol: oxidation products of isoprene) (Claeys 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.,

**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



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.

2 Methodology

2.1 Site location and aerosol sampling

5 Sampling was carried out during the winter ClearLo 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 10 (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.

15 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 20 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 25 with the new procedures which are described in greater detail.

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



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



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_{10} 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.

**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



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 $C_3H_7^+$ and $C_2OH_3^+$) 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 nitrogen-containing peaks were not deemed to have been successfully retrieved using PIKA. Five factors were identified: oxygenated OA (OOA), cooking OA (COA), hydrocarbon-like 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).

2.2.4 Rotational ambiguity

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 distinguishing 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 be seen as indicative of the amount of rotational ambiguity present in the solutions (Allan et al., 2010). However, the solution for $f_{\text{peak}} = 0$ 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.

2.2.5 The CMB model

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

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



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 ng m^{-3} and 94.5 ng m^{-3}), hopanes ($0.25\text{--}0.50 \text{ ng m}^{-3}$ and $0.079\text{--}0.36 \text{ ng m}^{-3}$) and PAHs ($0.10\text{--}0.67 \text{ ng m}^{-3}$ and $0.044\text{--}0.51 \text{ ng m}^{-3}$) 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: $0.08\text{--}0.18 \text{ ng m}^{-3}$; PAHs: $0.06\text{--}0.27 \text{ ng m}^{-3}$) and rural site CPSS (Levo: 7.7 ng m^{-3} ; hopanes: $0.07\text{--}0.15 \text{ ng m}^{-3}$; PAHs: $0.05\text{--}0.21 \text{ ng m}^{-3}$) in 2007–2008 (Harrison and Yin, 2010), but lower levels were observed for n-alkanes from the current study ($0.58\text{--}2.1 \text{ ng m}^{-3}$ and $1.2\text{--}3.7 \text{ ng m}^{-3}$ for NK and HAR), presumably due to a seasonal effect, since earlier results ($1.0\text{--}5.2 \text{ ng m}^{-3}$ and $1.8\text{--}4.7 \text{ ng m}^{-3}$ 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 ($0.73\text{--}1.9 \text{ ng m}^{-3}$) and CPSS ($0.47\text{--}1.7 \text{ ng m}^{-3}$).

3.2 CMB model results

Source contributions to fine particulate OC and $\text{PM}_{2.5}$ 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).

3.2.1 Source contributions to fine OC

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

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



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 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 PM_{2.5} 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 air mass ori-

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



5
10
15
20
25

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 ($1.63 \mu\text{g m}^{-3}$) derived from AMS-PMF was 2.0 times the CMB woodsmoke (CMB-WS) concentration estimate ($0.85 \mu\text{g m}^{-3}$) (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: $0.97 \mu\text{g m}^{-3}$). 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 ($0.23 \mu\text{g m}^{-3}$), London NK 2010/11 ($0.33 \mu\text{g m}^{-3}$) and Budbrooke, Warwickshire 2009–2010 ($0.42 \mu\text{g m}^{-3}$) (Harrison et al., 2012), whilst six Euro-

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](#)

pean sites showed biomass smoke OC concentrations in the range of 0.039–3.1 $\mu\text{g m}^{-3}$ annually and 0.048–7.7 $\mu\text{g m}^{-3}$ 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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ 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.

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



**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



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 ($r^2 = 0.84$ and 0.62 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^- ($r^2 = 0.79$ 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.

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 ambiguity (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 f_{peak} 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.

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](#)

The Partisol sampler used to collect samples for the CMB estimation is subject to both positive (adsorption of vapour on the filter) and negative (evaporation of semi-volatile material from the filter) artefacts.

5 Conclusions

5 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.

15 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.

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24550

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



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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



**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



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**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](#)

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**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



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**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



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**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



- Lanz, V. A., Alfara, M. R., Baltensperger, U., Buchmann, B., Hueglin, C., Szidat, S., Wehrl, 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.
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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



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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



**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



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Receptor modelling of fine particles in Southern England

J. Yin et al.

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) OC & EC SO ₄ ²⁻ , NO ₃ ⁻ , Cl ⁻ , Na ⁺ , K ⁺ , Mg ²⁺ , Ca ²⁺ , NH ₄ ⁺	GCMS OCEC analyser Dionex	Daily
	TOF-AMS	PM _{0.8}	Mass size segregated Or- ganic 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) OC & EC SO ₄ ²⁻ , NO ₃ ⁻ , Cl ⁻ , Na ⁺ , K ⁺ , Mg ²⁺ , Ca ²⁺ , NH ₄ ⁺	GCMS OCEC analyser Dionex	Daily

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Receptor modelling of fine particles in Southern England

J. Yin et al.

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	0.8	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 ₁	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 ₁	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 Seinfeld, 2007
Southeastern US, rural	Jun 2004	1.6	4.9	6.5	–	–	–	Clements and Seinfeld, 2007

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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

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

Source Name		OC			$\text{PM}_{2.5}$			OC/ $\text{PM}_{2.5}$ or OC/OM CF^c
		NK ^a	NK ^b	HAR ^a	NK ^a	NK ^b	HAR ^a	
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/ $\text{PM}_{2.5}$	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

Receptor modelling of fine particles in Southern England

J. Yin et al.

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

Pair of Variables	NK2012		r^2
	Slope	Intercept	
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 NO_3^-	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

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Receptor modelling of fine particles in Southern England

J. Yin et al.

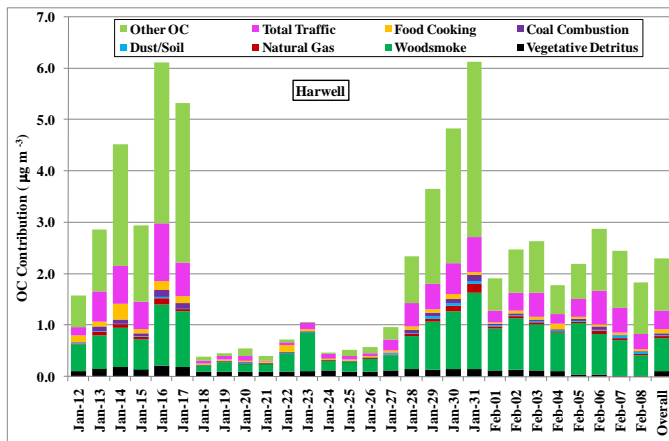
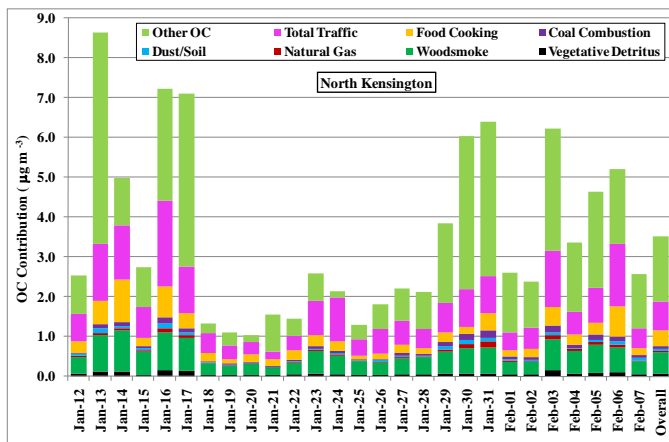


Figure 1. Daily OC source contributions at NK and HAR.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Receptor modelling of fine particles in Southern England

J. Yin et al.

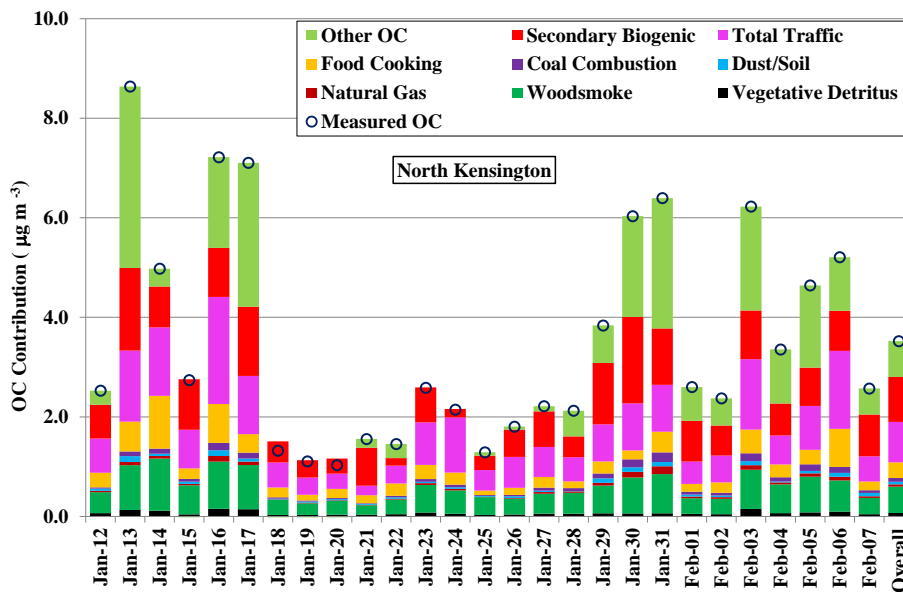


Figure 2. Daily OC source contribution estimates with secondary biogenic components at NK.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Receptor modelling of fine particles in Southern England

J. Yin et al.

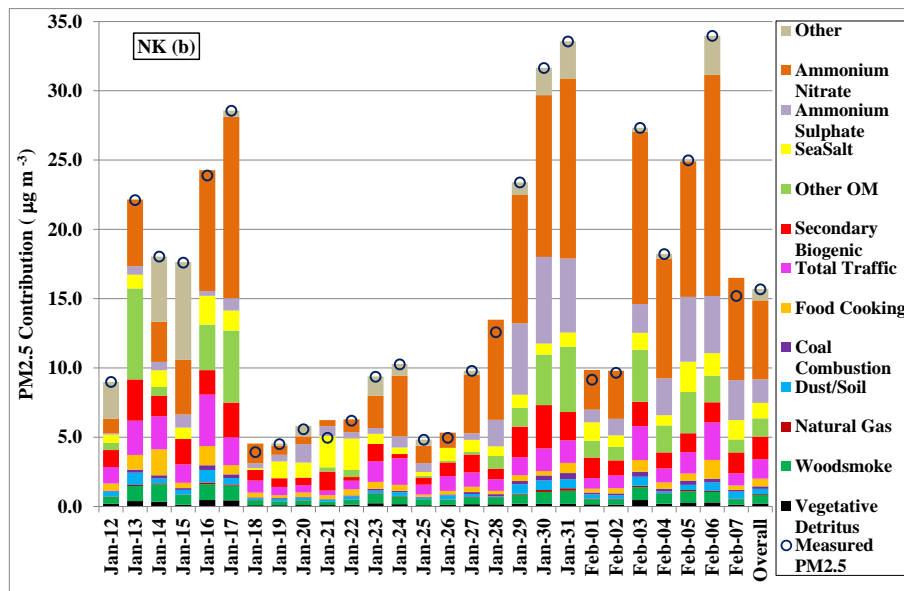


Figure 3. Daily PM_{2.5} source contribution estimates with secondary biogenic components at NK.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Receptor modelling of fine particles in Southern England

J. Yin et al.

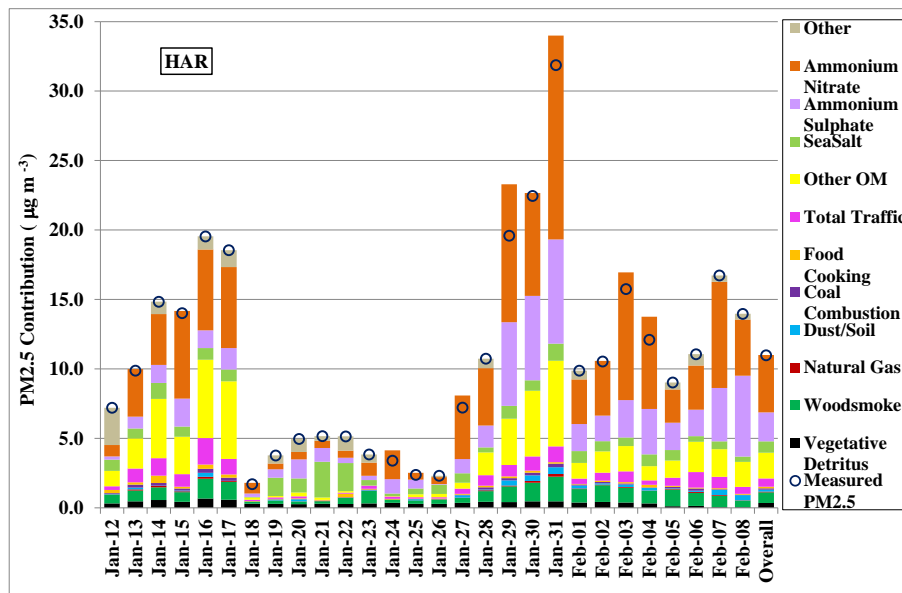


Figure 4. Daily PM_{2.5} source contribution estimates at HAR.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Receptor modelling of fine particles in Southern England

J. Yin et al.

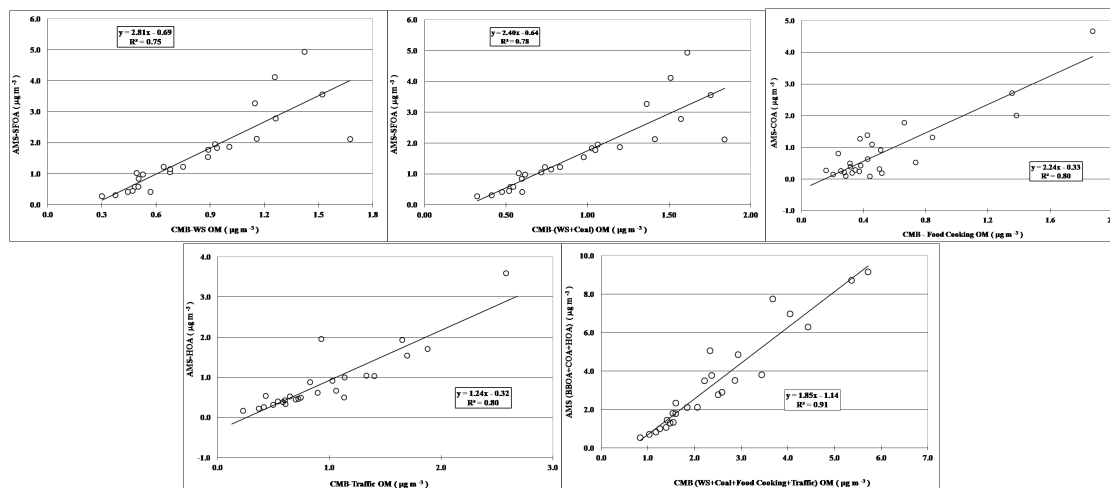


Figure 5. Primary component comparison between the AMS-PMF and CMB methods at NK.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Receptor modelling
of fine particles in
Southern England

J. Yin et al.

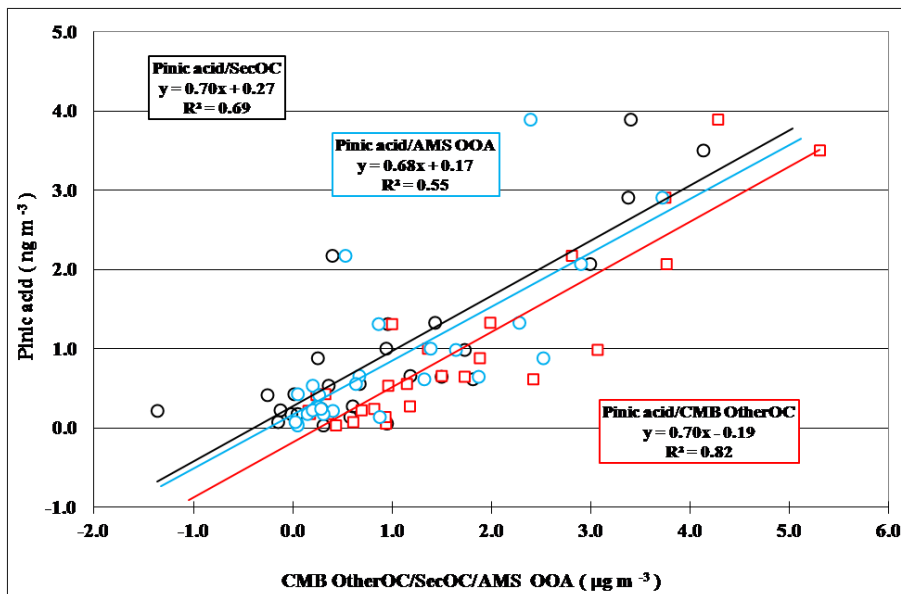


Figure 6. Secondary component comparison at NK showing relationships between pinic acid concentrations and estimated secondary OC from the elemental carbon tracer method (SecOC) and the CMB model (Other-OC), and with the AMS OOA factor.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)