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## Significant formation of water-insoluble secondary organic aerosols in semi-arid urban environment

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[1] Diurnal variations of submicron elemental carbon (EC), water-soluble organic carbon (WSOC) and water-insoluble organic carbon (WIOC) were investigated at an urban background site in Cairo (Egypt) from 23 March to 14 April 2005. Well-marked diurnal patterns, with minima during the traffic-influenced morning period (6:00–9:00) and maxima during the intense photochemical period (12:00–15:00), were observed for the WSOC/EC ratio and, more surprisingly, for the WIOC/EC ratio. Such diurnal variations suggest significant formation of both water-soluble and water-insoluble secondary organic aerosols during the afternoon. Applying the EC-tracer method, freshly-formed secondary organic carbon (fresh SOC) was found to possibly account for more than 50% of OC concentrations measured during the 12:00–15:00 period, and this fresh SOC was calculated to be mainly (~60%) composed of water-insoluble species. The latter (unexpected) result could be due to low ambient relative humidity as well as to the importance of anthropogenic volatile organic compounds in Cairo. **Citation:** Favez, O., J. Sciare, H. Cachier, S. C. Alfaro, and M. M. Abdelwahab (2008), Significant formation of water-insoluble secondary organic aerosols in semi-arid urban environment, *Geophys. Res. Lett.*, *35*, L15801, doi:10.1029/2008GL034446.

### 1. Introduction

[2] Organic aerosols, which represent a large - if not major - fraction of fine airborne particles, are expected to induce significant effects on local to global climate [Kanakidou *et al.*, 2005]. In particular, they could act as cloud condensation nuclei (CCN) and are then considered as possible key contributors to the aerosol indirect effect and to the alteration of the water cycle [Broekhuizen *et al.*, 2004]. Hygroscopic properties of organic aerosols are notably linked to their chemical composition and water-solubility [Mircea *et al.*, 2005]. Therefore particular attention has been paid recently to water-soluble organic carbon (WSOC) comprised in ambient aerosols. In urban environments, and in the absence of biomass burning emissions, WSOC is considered to originate mainly from secondary processes. Indeed, primary emissions from fossil fuel combustion are reported to be mostly water-

insoluble, whereas (photochemical) oxidation mechanisms, responsible for the ageing of carbonaceous particles as well as the condensation of volatile organic compounds (VOCs) in the particulate phase, lead to more water-soluble species [Saxena and Hildemann, 1996; Decesari *et al.*, 2002]. Secondary organic aerosols (SOA) have also become a subject of great scientific concern as they could represent the majority of fine organic particles, even in heavily urbanized area [Zhang *et al.*, 2007]. However, SOA formation/transformation mechanisms are still poorly understood, leading to significant underestimations of their concentrations and climatic impacts by current atmospheric models [Volkamer *et al.*, 2006].

[3] For all these reasons, some experimental studies recently investigated the water-solubility of urban SOA [Miyazaki *et al.*, 2006; Salma *et al.*, 2007; Weber *et al.*, 2007]. These studies seem to indicate that SOA formed in anthropogenic-influenced air masses are composed almost exclusively of water-soluble species. Such results could find significant impacts in atmospheric science. They notably suggest that urban SOA concentrations may be adequately approximated by water-soluble organics, whose direct measurement in ambient air is now feasible [Sullivan *et al.*, 2004]. However, these studies were performed in few urban centres only, and it is not clear yet whether such findings can be extrapolated to all urban atmospheres. In particular, they focused on relatively humid urban environments (e.g., Tokyo) and, since mechanisms leading to the formation of SOA are expected to be dependent on ambient relative humidity [Seinfeld *et al.*, 2001], one could wonder whether SOA formed in relatively dry urban atmospheres are primarily composed of water-soluble species too.

[4] The aim of this paper is to investigate the water-solubility of submicron SOA formed in Cairo (Egypt), taken as an example of semi-arid urban atmosphere. Results presented hereafter suggest that freshly-formed SOA are mainly water-insoluble in such an environment. This finding is discussed in light of atmospheric conditions, but also of gaseous precursor origins (anthropogenic vs. biogenic).

### 2. Experiment

[5] Results presented in this paper were obtained in the frame of the Cairo Aerosol Characterization Experiment (CACHE) during the 2005 spring season (23 March to 14 April). Aerosol sampling was achieved at an urban background site (Cairo University, Giza) described in details by Favez *et al.* [2008]. Meteorological parameters, including relative humidity, were measured in the field (on a 5-min basis) using an Oregon Scientific WMR-918 weather

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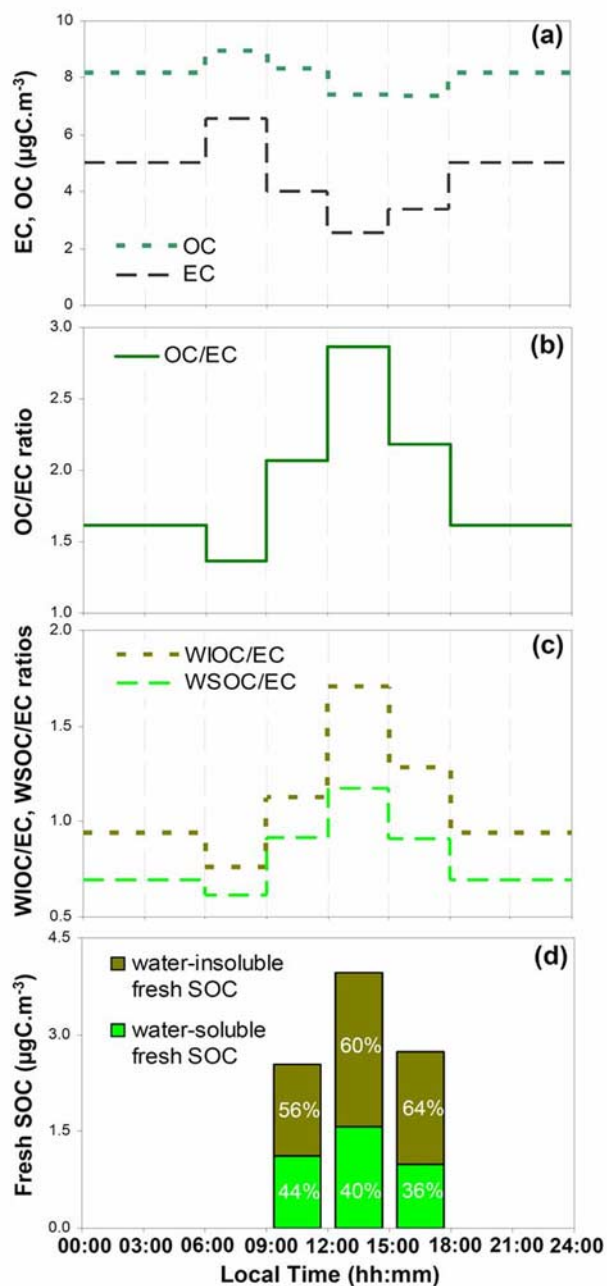
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station. A total number of 112 submicron aerosol samples were collected on pre-baked 47-mm quartz fibre filters (Whatman QMA) using a PM<sub>1</sub> sharp cut cyclone (URG corporation) running at a flow-rate of 1 m<sup>3</sup>.h<sup>-1</sup>. Sampling intervals were of 3 hours during daytime (6:00–9:00, 9:00–12:00, 12:00–15:00 and 15:00–18:00, local time) and of 12 hours during the night (18:00–6:00).

[6] Filters were analysed for their organic carbon (OC), elemental carbon (EC), and WSOC contents. Water-insoluble organic carbon (WIOC) concentrations were then calculated as the difference between OC and WSOC. OC and EC analyses were achieved on 1.5 cm<sup>2</sup> punches using a thermo-optical carbon analyser (EC-OC Sunset Lab. Instrument) working in transmittance and implemented with the IMPROVE thermal program, as described by *Sciare et al.* [2003]. The detection limit and the analysis uncertainty of this instrument are of 0.2 μgC and 5%, respectively. WSOC analyses were conducted using a Total Organic Carbon analyser (Sievers TOC 900) following the analytical protocol detailed by *Favez et al.* [2008]. Briefly, extraction of the soluble carbonaceous fraction (WSOC) was performed on 2.54 cm<sup>2</sup> punches and in 25 mL of ultra pure water. The soaking step was conducted overnight (~16 hours) under soft shaking. Prior to analysis, the extract solution was filtered through Teflon (PTFE) filters (0.2 μm pore size diameter), in order to remove any suspended particle. The overall uncertainty of WSOC analyses is estimated here to be of 10%.

### 3. Secondary Organic Carbon and Partitioning Between WSOC and WIOC

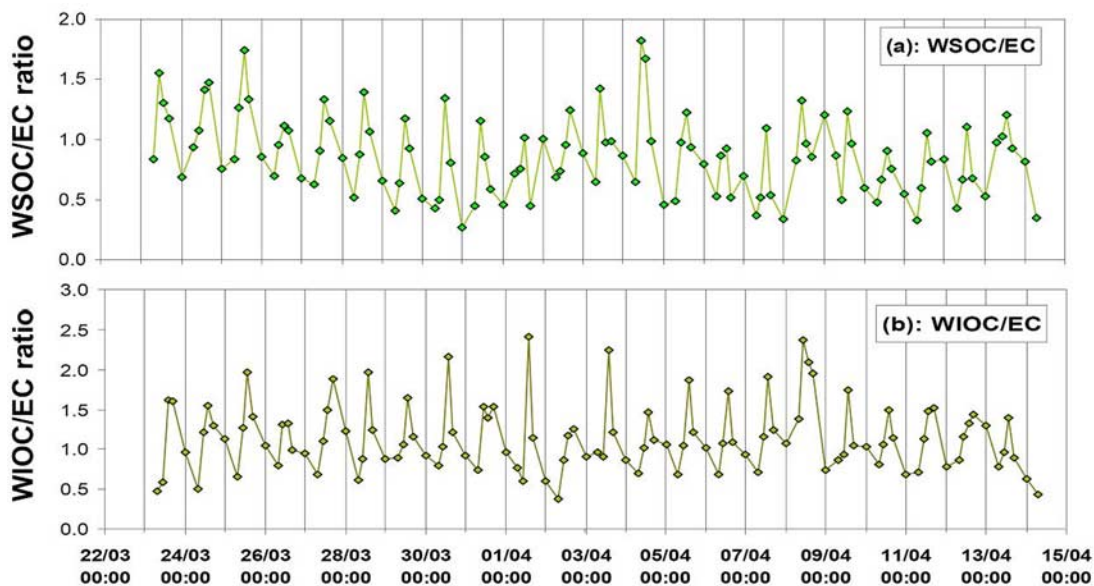
[7] For the whole period of study, diurnally averaged submicron EC, WIOC and WSOC concentrations were of  $4.5 \pm 2.1 \mu\text{gC.m}^{-3}$ ,  $4.8 \pm 2.3 \mu\text{gC.m}^{-3}$  and  $3.2 \pm 1.1 \mu\text{gC.m}^{-3}$  respectively. A mean WSOC/OC ratio of ~0.40 is then obtained. This value lies in the upper range of WSOC/OC ratios previously reported for urban environments [*Miyazaki et al.*, 2006; *Weber et al.*, 2007; *Salma et al.*, 2007; *Viana et al.*, 2007], illustrating the importance of water-soluble organics in Cairo, and possibly significant SOA formation in this megacity. The consequence of such secondary mechanisms can be observed through diurnal variations of carbonaceous aerosols. As shown in Figure 1a, EC concentrations displayed a clearly marked diurnal pattern, with maximum values ( $6.6 \pm 3.0 \mu\text{gC.m}^{-3}$ ) - obtained for the 6:00–9:00 period - being almost three times higher than minimum ones ( $2.6 \pm 1.1 \mu\text{gC.m}^{-3}$ , obtained for the 12:00–15:00 period). Compared to EC, the mean OC diurnal cycle fluctuated in a quite narrow range (Figure 1a). In turn, a well-marked diurnal pattern is obtained for the OC/EC ratio (Figure 1b), displaying minimum values ( $1.4 \pm 0.3$ ) during the 6:00–9:00 period and maximum values ( $2.9 \pm 0.5$ ) during the 12:00–15:00 period. Remarkably, this OC/EC diurnal cycle has been observed every day during the campaign. Primary emissions of fine carbonaceous aerosols in Cairo have been demonstrated to be largely dominated by mobile sources throughout the day [*Abu-Allaban et al.*, 2007; *Mahmoud et al.*, 2008]. Therefore, lowest OC/EC values measured during the morning period (i.e., the rush-hour 6:00–9:00 period) have to be attributed to primary traffic emissions, which are known to be characterized by relatively low OC/EC ratios. On the other hand, the increase



**Figure 1.** Mean diurnal cycles of (a) OC and EC concentrations, (b) OC/EC ratio, (c) WSOC/EC and WIOC/EC ratios, and (d) partitioning between the water-soluble and water-insoluble fractions among of “fresh” secondary organic carbon.

of the OC/EC ratio observed during the afternoon can thus be mainly attributed to secondary processes leading to SOA.

[8] In order to evaluate concentrations of secondary organic carbon (SOC), the EC-tracer method [*Turpin and Huntzicker*, 1991] has been applied to our results. This indirect method commonly used to evaluate primary and secondary organic aerosols postulates that EC is a good tracer of primary OC because both fractions are mostly emitted from the same combustion sources. According to this method, primary organic carbon may be estimated considering an OC/EC ratio characteristic of primary emis-



**Figure 2.** Temporal variations of the (a) WSOC/EC and (b) WIOC/EC mass ratios during the whole period of study.

sions. Due to the lack of information on such OC/EC ratio specific for Cairo emissions, it is not possible to precisely calculate SOC concentrations here. However SOC formed during a particular day (fresh SOC) may be evaluated considering the OC/EC ratio measured the same day during the rush-hour period, as follows:

$$\text{SOC} = \text{OC}_{\text{total}} - (\text{OC}/\text{EC})_{6:00-9:00} \times \text{EC} - \text{OC}_{\text{primary, non combustion}} \quad (1)$$

where  $\text{OC}_{\text{primary, non combustion}}$  stands for non combustion OC primary emissions (e.g., marine primary organic aerosols or plant debris) and may be neglected for submicron aerosols investigated here. Applying this methodology, it appears that, on average for the whole period of study, more than 50% of OC concentrations measured during the intense photochemical afternoon period (12:00–15:00) might originate from fresh SOA formation. This finding is in accordance with recent studies showing that mechanisms leading to SOA may be very intense and rapid in urban atmospheres [Volkamer *et al.*, 2006].

[9] Significant SOA formation through photochemical processes is expected to increase the contribution of WSOC among carbonaceous particles. The WSOC/EC ratio is actually found here to display a well marked diurnal pattern, with maxima observed during the 12:00–15:00 period and being higher by a factor 2 than minima, obtained for the 6:00–9:00 period (Figure 1c). Less expected is the diurnal pattern obtained for the WIOC/EC ratio, which is found to be more than twice higher on average during the afternoon than during the 6:00–9:00 period (Figure 1c). It is to note that such diurnal variations have been observed every day during the field campaign (Figure 2). This finding seems to indicate that a large fraction of fresh SOC is composed of water-insoluble organics in Cairo.

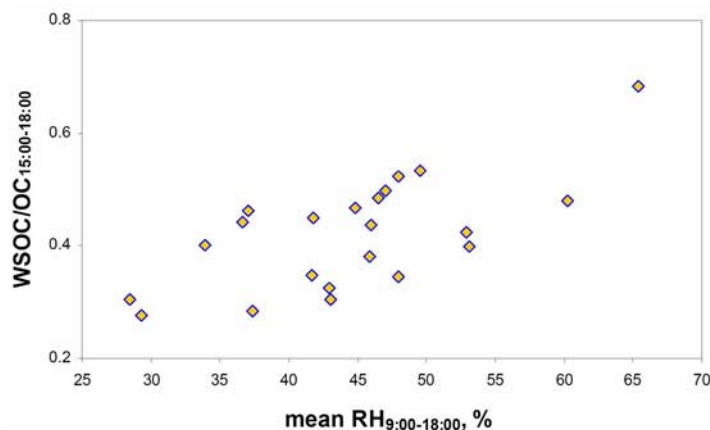
[10] In order to estimate the partitioning between WSOC and WIOC among fresh SOC, OC was replaced by WSOC (and then WIOC) in equation (1). Results, presented in Figure 1d, suggest that up to 60% of fresh SOC formed in Cairo is water-insoluble. To the best of our knowledge, this

is the first time that such an important contribution is derived from field data, previous works having related SOA formation solely through the WSOC fraction. In the following section we demonstrate that results presented here can not be attributed to sampling artefacts.

#### 4. Sampling Artefacts

[11] The collection of aerosol using filtration techniques is known to be submitted to negative sampling artefacts (i.e., losses of semi-volatile organic matter from the filter during the aerosol collection) as well as to positive sampling artefacts (i.e., adsorption of gaseous organics on the filter). In this study, negative artefacts are expected to be minimized for daytime samplings due to the shortness of collection periods (3 hours), assumed to correspond to little changes in the gas-particle partitioning equilibrium. They are also expected to be limited for filters collected during the night due to relatively low temperatures ( $13.5 \pm 3.6^\circ\text{C}$  on average for the 18:00–6:00 period).

[12] Although positive sampling artefacts can not be excluded, they are assumed to poorly affect OC/EC ratios presented here, due to high EC concentrations prevailing in Cairo. Novakov *et al.* [2005] showed that in urban environments, OC/EC ratios are not significantly affected by sampling artefacts for EC concentrations above  $1.5 \mu\text{gC}\cdot\text{m}^{-3}$ . EC concentrations measured in this study are on average three times higher than this threshold value. Moreover, highest OC/EC ratios observed during the afternoon ( $2.9 \pm 0.5$  on average) are still in the range of artefact-corrected OC/EC ratios proposed by Novakov *et al.* [2005] for urban atmospheres ( $2.2 \pm 1.5$ ). For these reasons we assume positive sampling artefacts to be limited here (<20% of total OC). Furthermore, it should be noted that positive artefacts are expected to increase the WSOC/OC ratio [Salma *et al.*, 2007], and may thus not be responsible for the large contribution of WIOC to secondary organics observed in this study. This seems to be confirmed by results obtained from low pressure cascade impactor samplings (LPI, 13-stages Dekati) performed during the campaign



**Figure 3.** Positive linear correlation ( $r = 0.68$ ,  $n = 22$ ) obtained between the WSOC/OC ratio measured at the end of the day (15:00–18:00) and mean daytime ambient relative humidity.

(data not shown). Indeed, impaction techniques are commonly assumed to be barely submitted to positive artefacts, and submicron WSOC/OC ratios measured for LPI samplings (three daytime samplings) are slightly lower than those obtained for PM<sub>1</sub> filters collected during the same periods ( $0.28 \pm 0.03$  and  $0.32 \pm 0.04$  on average for LPI samples and PM<sub>1</sub> filters respectively). Therefore, although positive artefacts cannot be ruled out, they should preferentially affect WSOC rather than WIOC measurements, and could then not be responsible for the here-above calculated predominance of WIOC among secondary organics. As exposed below, large amounts of water-insoluble SoA could be due to meteorological conditions as well as to the nature of gaseous precursors.

## 5. Impacts of Relative Humidity and of Gaseous Precursors

[13] Several studies have shown that SOA formation through VOC condensation/sorption and subsequent aerosol phase reactions in ambient air could be driven by aqueous phase reactions [e.g., *Heald et al.*, 2006]. Moreover, some laboratory experiments have identified relative humidity (RH) as a key factor controlling SOA formation, the lower RH the less efficient the condensation of hydrophilic compounds in the particulate phase [e.g., *Seinfeld et al.*, 2001]. It could thus be envisaged that changes in ambient RH, and subsequently in the aerosol liquid water content, may lead to competing effects between the formation of highly oxidized polar compounds (more hygroscopic) and the formation of longer-chained oligomers (less hygroscopic). Low RH prevailing in Cairo during daytime (<45% for the 9:00–18:00 period, on average for the whole period of study) might then be partly responsible for the relatively low contribution of WSOC to fresh SOC observed here. This hypothesis seems to be confirmed by the positive correlation ( $r = 0.68$ ) obtained between ambient RH (averaged during the photochemically active period: 9:00–18:00) and the WSOC/OC ratio measured at the end of the day (15:00–18:00 period, allowing the integration of the whole production of freshly-formed SOA), as shown in Figure 3.

[14] The origin of organic gaseous precursors (anthropogenic or biogenic) can also be put forward to explain the high contribution of WIOC to fresh SOC calculated here. It

is commonly assumed that anthropogenic VOCs (AVOCs) are less polar and less hydrophilic than biogenic VOCs (BVOCs) [*Song et al.*, 2007]. By extension, SOA originating from AVOCs might also be less hydrophilic (and less water-soluble) than biogenic SOA. This hypothesis is consistent with recent studies showing that WSOC aerosols found in urban environments show much more similarities (in structure and in radiocarbon content) with biogenic SOA than with anthropogenic SOA [*Baltensperger et al.*, 2005; *Weber et al.*, 2007]. These results suggest then that SOA formed in environments characterized by relatively high AVOC/BVOC ratios could be less water-soluble than SOA formed in urban environments with lowest AVOC/BVOC ratios. In Cairo, levels of AVOCs are in the upper range of those reported for the most polluted megacities in the world [*Khoder*, 2007]. Regarding BVOCs, no data is available yet. However, modelling studies [*Lathière et al.*, 2005; *Kanakidou et al.*, 2005] have clearly shown that the areas surrounding Cairo are not significant sources of isoprene or terpenes by comparison with mid-latitudes regions, where most of SOA studies were performed [*Zhang et al.*, 2007]. Therefore, the ratio between AVOCs and BVOCs is expected to be significantly higher (possibly one order of magnitude) in Cairo than in urban centres where WSOC of secondary origin have been investigated previously (United States, Europe and Eastern Asia). This could partly explain the much larger contribution of WIOC to SOC observed in our work, compared to previous ones (<25% in Tokyo) [*Miyazaki et al.*, 2006].

[15] Finally, investigating secondary organics in Los Angeles, *Kleeman et al.* [2007] have demonstrated that RH does not have significant influence on the mechanisms leading to anthropogenic SOA, whereas the formation of biogenic SOA could be limited under low RH. It could then be envisaged that large amounts of water-insoluble SOA observed here are related to the combination of both low RH and high AVOC concentrations occurring in Cairo.

## 6. Concluding Remarks

[16] The major contribution of this paper is to show that in urban atmospheres characterized by relatively low RH and high AVOC/BVOC ratios, SOA could be mainly

composed of water-insoluble species. This finding clearly indicates that WSOC may not be considered as an adequate quantitative tracer for SOA in every urban environment. This paper also suggests a strong influence of relative humidity on the partitioning between water-soluble and water-insoluble organics in ambient air. Mechanisms controlling WSOC content in SOA are poorly documented and such findings clearly point out the need for further characterization of the water-solubility of SOA, in source regions as well as in city-plumes. Finally, it is hypothesized here that high levels of AVOCs found in Cairo could enhance the formation of water-insoluble SOA. The use of artefact-free advanced methods (e.g., aerosol mass spectrometer and radiocarbon measurements) and the investigation of VOCs in semi-arid urban atmospheres should help bridge the gap between the nature of gaseous precursors (anthropogenic vs. biogenic) and the water-solubility of SOA.

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