

Two-stroke scooters are a dominant source of air pollution in many cities

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Abstract

Fossil fuel powered vehicles emit significant particulate matter (PM), e.g. black carbon and primary organic aerosol (POA), and produce secondary organic aerosol (SOA). We quantify, for the first time, SOA production from two-stroke (2S) scooters. Passenger cars and trucks, particularly diesel vehicles, are thought to be the main vehicular PM sources. This needs re-thinking, as we show for the first time that elevated PM levels can be a consequence of ‘asymmetric pollution’ from 2S scooters; vehicles that constitute a small fraction of the fleet, but can dominate urban vehicular pollution through organic aerosol and aromatic emission factors up to 1000s of times higher than from other vehicle classes. Further, we demonstrate that oxidation processes producing SOA from vehicle exhaust also form potentially toxic ‘reactive oxygen species’.

PM damages health¹ and affects climate². Road vehicles are a significant source of PM, particularly in urban areas. A number of recent studies have shown that a large fraction, possibly the largest fraction, of vehicular PM is secondary produced via atmospheric oxidation of precursor gases in the exhaust³⁻⁵. Thus, understanding vehicular air pollution requires an assessment of SOA formation from different vehicle types. 2S scooters (powered two-wheeled vehicles with engine displacement $\leq 50 \text{ cm}^3$) are popular globally, particularly in Asia, Africa, and Southern Europe. Despite being high emitters of primary PM^{6,7}, regulations for scooters are generally less stringent than for other vehicles, e.g. in Europe having reached Euro 5/V (a fifth tranche of regulations), for passenger cars and trucks, vs. only Euro 2 for scooters (see supplementary information (SI), and ref. 8). Accordingly, a scientific report to

the European Commission suggests that scooters will emit more volatile organic compounds (VOCs) than all other vehicles combined in Europe by 2020 (ref. 9). Furthermore, high PM levels and toxic aromatic hydrocarbons, important SOA precursors¹⁰, have been observed in many cities, especially in Asia¹¹. Globally, organic aerosol (OA) dominates PM, with SOA accounting for the largest fraction⁴. Here, we show that 2S scooters emit significant amounts POA, aromatic VOCs and also produce significant SOA. We use the term ‘asymmetric polluter’ to describe these vehicles as their emission factors and evidence from air quality measurements before and after bans on scooters in Asian cities suggests they may dominate vehicular pollution despite relatively small numbers. Chemical analysis of the emissions shows that SOA is mainly produced via photo oxidation of aromatic VOCs, present in gasoline, from the exhaust. This shows that the known issue of incomplete fuel combustion during the 2S cycle is also responsible for SOA formation. Finally we present the first online measurements of aged exhaust showing that SOA formation also produces reactive oxygen species ‘ROS’ with potentially detrimental effects on our lungs.

Results

Emission factors

We investigated POA emissions and SOA formation from 2S scooters and their potential health effects. The oxidation of VOCs in 2S scooter emissions produces significant SOA (g carbon (C) kg⁻¹ fuel), with total OA on average 2.9 and 2.4 times higher than POA after aging for idling and driving 2S scooters, respectively (Figure

1, and Table S3, SI). In addition, substantial toxic aromatic emissions (up to ~40% of emitted VOC volume for the scooters of this study) of benzene, toluene, and C2-C4 alkylated benzenes, which are recognised SOA precursors^{10,12}, are present in the exhaust. Among the aromatics, benzene is of particular concern due to its carcinogenicity. Levels in the raw 2S scooter exhaust were as high as 300000 $\mu\text{g m}^{-3}$ or 146 ppm(v) from idling. The EU annual mean limit for the protection of human health is 5 $\mu\text{g m}^{-3}$ (ref.13), while the US National Institute for Occupational Safety and Health (NIOSH) recommends that workers wear special breathing equipment when exposed to benzene at levels exceeding 1 ppm for 15 minutes. Waiting in traffic behind a 2S scooter, e.g. at junctions and while the scooter is idling, may therefore be highly deleterious to health.

Secondary organic aerosol yields

The contribution of the aromatics to SOA formation was estimated by calculating an apparent aerosol yield, y_{apparent} , assuming all SOA comes from aromatic precursors:

$$y_{\text{apparent}} = C_{\text{SOA}} / \sum_i \Delta_i \quad (1)$$

where C_{SOA} is the SOA produced ($\mu\text{g m}^{-3}$) for a given mass change in aromatic i (Δ_i , i = benzene, toluene, or C2-C4 alkylated benzenes). Apparent yields closely match average concentration-weighted literature aromatic SOA yields¹⁰ (Figure 2a, see also SI) for idling, complete ECE47 driving cycles, and ECE47 phase one (Ph1), indicating that most SOA is from aromatic precursors (Figure 2a). SOA from ECE47 phase two (Ph2) alone is underestimated by Eq. 1, suggesting SOA production from unidentified compounds, emitted by the hot engine. Note that the total emission

during a full cycle is dominated by Ph1, i.e. by cold engine emissions. Furthermore, a *Van Krevelen diagram* illustrates the aging of 2S scooter emissions, from oxygen (O:C~0 to O:C~0.6). This elemental composition is consistent with that of previously observed SOA from aromatic precursors¹⁴ (Figure 2b). We therefore conclude that SOA formation from 2S scooter emissions is likely from the oxidation of aromatics, in contrast to diesel SOA, which is predominantly from other precursors¹⁵.

Comparison to other vehicle types and ambient data

Figure 1 also shows laboratory and ambient measurements of POA; light aromatic; and benzene emission factors EFs from passenger cars and trucks (see SI for methodology and Table S4). Ambient data are from roadside/ tunnel measurements in the US, EU, and Asia, and are split according to the fraction of light duty and heavy duty vehicles (LDV and HDV) at the measurement site. Note that the general trend is for lower EFs in newer studies (Table S4), consistent with improvements in emission controls. Also shown are data from Indian in-use 2S auto rickshaws for comparison to the European scooters of this study. Caution is required in such a comparison however; although similar (both have 2S engines), these are a different vehicle class, and were furthermore tested during a different driving cycle. In general, ambient emission factors from Asian vehicles are in the same range as European and US vehicles, while emissions from in-use 2S rickshaws are slightly higher than from the European scooters of this study. POA emissions from 2S scooters are on average around 20 (maximum 2780) times higher than ambient (light duty dominant) values, and aged OA an average 53-771 times higher than laboratory studies on other vehicle types. It should be noted that absolute aerosol concentrations can influence emission

factors: higher measurement concentrations would lead to higher emission factors¹⁴. SOA formation is most significant from idling scooter emissions, while smaller at higher engine loads. However, POA emissions are higher under the latter conditions, and the aggregate POA+SOA emission at high load is comparable to that from idling.

Reactive oxygen species

We also examined the health implications of the 2S scooter SOA (other than those from the mass increase) using online measurements of particle-bound, water soluble reactive oxygen species (ROS), which are linked to negative health effects¹⁹. ROS are undetectable in POA, but accounts for 0.5-1% carbon in the aged OA, suggesting that PM emissions initially become increasingly toxic with aging (Figure 4). Increasing ROS is consistent with the increased O:C ratio of the aerosol and in line with a previous study showing increased oxidative potential with aging for 2S scooter emissions, albeit at aerosol and oxidant loadings much higher than under ambient conditions²⁰. After 1-2 hours of irradiation ROS stabilises or decreases, as reported previously for organic peroxides, likely due to decomposition processes^{21,22}.

Discussion

There are likely several reasons for these relatively large OA and aromatic emissions from 2S scooters. Firstly, 2S engines, unlike 4S, require addition of lubricant oils to the fuel, some of which is emitted in the exhaust. Secondly, during the 2S engine cycle some of the fresh fuel/air mixture passes directly through the engine¹⁶, increasing VOC emissions, which may explain the high SOA formation. Thirdly,

scooters generally utilise *rich combustion* (low air/ fuel ratio), improving drivability while producing higher CO, VOC and PM emissions (but lower NO_x). Accordingly, the VOC emissions measured here, in particular aromatics as found in raw gasoline, are also on average 124 and 11times higher from idling and driving 2S scooters, respectively, compared to those from other vehicles. Finally, scooter after-treatment systems are inherently inefficient due to their relatively small size and longer light-off times.

Precise estimation of a relative contribution to vehicular PM and aromatics from 2S scooters is difficult since vehicle regulations vary by country. Another complication arises from the possibility of large contributions to OA from a small number of super-polluting vehicles (of all types). However, many scooters will likely fall into this super-polluting category, especially as a considerable number of scooters are in operation in some regions without any form of emissions control (note that all scooters presented in this study are equipped with two-way oxidation catalysts, which reduce carbon monoxide and VOCs) and because emissions may be further exacerbated by poor maintenance and tampering, rife for scooters¹⁷. Furthermore, ambient data in Figure 1 likely include a number of such super polluting vehicles. Therefore our results suggest that 2S scooters are ‘asymmetric polluters’ of OA and aromatics compared to other vehicles. Using the average 2S scooter EF (ECE 47 driving cycle) in Figure 1 suggests that 2S scooters contribute to around 60% of roadside POA in Bangkok where they account for 10% of fuel consumption (Figure 3). In a more extreme case (comparing the 75th percentile for scooters and 25th percentile for ambient light duty dominated) 2S scooters would contribute over 96% to roadside POA. Note that these values are based on the European scooters of this study. As Figure 1 shows, emissions from some in-use Asian 2S vehicles may be

higher, by a factor of three. Since other Asian vehicles are not expected to be more polluting based on Figure 1, higher emissions from in-use Asian 2S vehicles would strengthen our conclusion that 2S scooters dominate urban pollution in the region. Estimation of contributions to aged OA are more difficult as vehicular SOA has not been systematically quantified under ambient conditions. However, smog chamber measurements suggest average aged OA contributions to ambient vehicular PM of 85 % (comparing to LDVs meeting Euro 5) or 98% (comparing to LDVs not meeting Euro 5) from 2S scooters. Meanwhile, in the EU, 2S scooters consume only 1 % of vehicle fuel, Figure 3. Even with these low numbers, scooters may be the major source of some of the vehicle related pollutants, especially in Southern Europe, and our data suggest that reducing the numbers of these vehicles would cost-effectively mitigate vehicle OA and aromatic emissions, given the alternatives available (electric and 4S). In this regard China has taken the lead, banning or restricting scooters in many cities since the late 1990s¹⁸, leading to large decreases in the traffic-related aromatic emissions in some Chinese cities (Figure 3b). Strikingly, roadside aromatics are now higher in Dongguan, where scooters are not banned, than 60 km away in Guangzhou, even though the traffic volume is much higher in Guangzhou (Figure 3c). This result is statistically significant: year-to-year BTEX concentrations in Guangzhou were 229 $\mu\text{g m}^{-3}$ in 1996, 244 $\mu\text{g m}^{-3}$ in 1999, 290 $\mu\text{g m}^{-3}$ in 2000, and 150 $\mu\text{g m}^{-3}$ in 2002, average $228 \pm 68 \mu\text{g m}^{-3}$, vs. 37 $\mu\text{g m}^{-3}$ after the scooter ban in 2005, for example.

Our data suggest that 2S scooters are a significant, and in many cities the largest, source of vehicular PM and toxic SOA and aromatic hydrocarbons, despite being a relatively small fraction of the total fleet. Therefore, given the alternative technologies

available, restrictions on 2S scooters, already implemented in China, could improve air quality in many cities around the globe.

Methodology

We combine results from two measurement campaigns where 2S scooter exhaust was injected through a heated inlet into smog chambers^{3,23,25} to produce SOA via photochemistry. During the first study an in-use Euro 1 (E1) and a new Euro 2 (E2a) 2S scooter were run in idle or simulated low power. During the second campaign emissions from a different Euro 2 2S scooter (E2b) were sampled during ECE47 driving cycles². Table S1 provides specifications of these vehicles. *European exhaust emission standards* are shown in Table S2.

All experiments were under high NO_x conditions (where the main reactions of peroxy radicals (RO₂) are with NO rather than other RO₂ radicals see SI and Table S3).

Average OH concentrations were $\sim 5 \cdot 10^6 \text{ cm}^{-3}$. OH concentrations were from the decay of a nine times deuterated butanol (butanol-D9, 98% Aldrich) tracer as measured using a quadrupole PTR-M.S (idling 2S scooters) or PTR-ToF-MS (Ionicon Analytik, driving cycle 2S scooters), see also Barmet et al., 2012 (ref 28).

Idling scooter experiments

Emissions were introduced into the 27 m³ Paul Scherrer Institute Teflon environmental chamber²³. The external temperature of the scooter exhaust was monitored (Thermocouple type K, Messelemente) and after an initial warming period of several minutes (consisting of idling or applying low power) the emissions were

injected only when the external exhaust temperature was stable at idle or at simulated low power. Table S3 provides the operating conditions, smog chamber OA concentrations and aerosol emission factors of this study used in Fig. 1. OA was monitored high resolution time of flight aerosol mass spectrometer (HR-ToF-AMS, Aerodyne. Unity collection efficiency is assumed since emitted particles likely consist of spherical oil-like droplets with low bounce. After an initial spike in the OA concentration following sample injection, a time of at least twenty minutes was allowed for equilibration. The concentration of OA after this point was taken as the initial POA emission. A battery of 80 100W UV black lights (ErgoLine “Cleo Performance”, Solarium lights), was used to initiate photo-oxidation and SOA formation. Experiments were carried out with a steady injection of NO (<20 ml min⁻¹) whereby NO was maintained at around 2-3 ppb(v). Relative humidity inside the smog chamber was between 40-60% for all experiments, and temperature was maintained at 25 °C.

OA was corrected for wall losses using

$$OA_{WLC}(t) = \frac{OA_{Meas}(t)}{\exp(-kt)}, \quad (2)$$

where $OA_{WLC}(t)$ and $OA_{Meas}(t)$ are the wall loss corrected and measured organic matter concentrations, respectively, as a function of time t , and k is the first order mass loss rate constant determined from an exponential fit of BC data.

Volatile organic compounds inside the smog chamber were quantified with a quadrupole proton transfer reaction mass spectrometer (PTR-MS), while carbon monoxide was quantified with a dedicated CO monitor (Aerolaser, CO-Monitor AL5002) and total gas phase hydrocarbons were measured from the chamber using a

flame ionization detector (FID, J.U.M model VE 7). Additional measurements at the tailpipe were performed by transferring emissions through a heated line (191 °C) to a Fourier transformed infrared spectrometer (FTIR, MKS Multigas analyzer 2030) for online measurements (at 1 Hz) of small hydrocarbons, nitrogen containing species (NO, NO₂, N₂O, NH₃ and HCN) and other oxygenated small organics (formaldehyde, acetaldehyde), as well as CO and CO₂.

Online reactive oxygen species measurements

Online particle bound ROS analysis utilised the fluorescence probe 2,7-dichlorofluorescein (DCFH) in solution. Particles were collected and continuously extracted on a wetted hydrophilic filter. The particle collector samples air at 5 litres per minute and collects particles larger than aerodynamic diameter 50 nm with greater than 95% efficiency. . Particles are collected and extracted in an aqueous solution of horseradish peroxidase (HRP) (0.5 units per ml) allowing immediate reaction of ROS on collection. The concentration of ROS is characterised following subsequent reaction of the oxidised HRP with DCFH (5 µM) for 10 minutes at 40 °C, yielding the fluorescent product DCF in the continuous flow set-up. The concentration of DCF is measured using fluorescence spectroscopy in a flow-through cell and calibrated to ROS concentration with hydrogen peroxide. ROS data in Fig. 4 are normalised to the total carbon per m³, determined from high resolution fitting of aerosol mass spectrometer data, and presented as a percentage.

Driving cycle scooter experiments

The Paul Scherrer Institute mobile smog chamber³ was deployed, and experiments conducted, in a certified chassis dynamometer test cell (Vehicle Emissions Laboratories, Joint Research Centre of the European Commission, JRC-Ispra, Italy)^{29,30}. Emissions from 2S scooters were sampled at the tailpipe during full ECE47 driving cycles, during phase one only of the ECE47 (first four modules of the driving cycle, Ph1), and during phase two only of the ECE47 (final four modules of the driving cycle, Ph2). The emissions were transferred to the smog chamber via a heated inlet system (150 °C) and Dekati ejector dilutor. UV lights were switched on after several minutes to initiate photochemistry.

OA concentrations were measured with a HR-ToF-AMS (Aerodyne), while black carbon was quantified with an aethalometer (AE33, Aerosol d.o.o.). The exponential decay rate of black carbon k was used in Eq. 2 to correct for particle losses to the walls. Gas phase compounds were monitored with a proton transfer reaction time of flight mass spectrometer (PTR-ToF-MS, Ionicon), while CO₂ and CO were measured using a cavity ring down spectrometer (Picarro, G2401) and total hydrocarbons were measured with a flame ionisation detector (Horiba, THC Monitor APHA-370).

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Competing interest statement

The authors declare no competing financial interests.

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Figure 1: Emission factors from two-stroke scooters and other vehicles measured in the laboratory and under ambient (road-side/ tunnel) conditions. Emission factors (EF) plotted as box-and-whiskers (median line, red; 25th and 75th percentile, box; 10th and 90th percentile, whiskers) of a) POA, b) aged OA (POA+SOA formation), c) benzene and d) light aromatics (benzene, toluene, and C2-C4 alkylated

benzenes,). Points shown next to the box and whiskers are the individual data points, coloured depending on measurement region for ambient data. 2S scooters (this study) were run in idle or during driving cycles (ECE47). Data on the other vehicles shown are from the literature (please see SI, Table S4) for light duty and heavy duty vehicles (LDV and HDV). LDV data is further divided between vehicles meeting Euro 5 and those not meeting Euro 5, labelled <Euro 5 in parenthesis. Ambient data are split according to a contribution of HDVs to the data of higher than or lower than 50%. Note that, many of the higher ambient values are from older vehicle studies (see Table S4)

Figure 2: Contribution of aromatic oxidation to two-stroke scooter secondary aerosol formation a) Apparent SOA mass yields, $y_{apparent}$ (Eq. 1), as a function of suspended OA (C_{OA}). Error bars show the sensitivity of $y_{apparent}$ to the chamber wall loss factor, \pm one standard deviation. $y_{apparent}$ for a Euro 1 and two Euro 2 2S scooters) are shown in red, blue and orange, respectively. Ph 1 and Ph 2 are the first and second phases of the ECE47 driving cycle, I and LP refer to Idling and simulated Low Power, respectively. A predicted yield, concentration weighted, for the mixture of all aromatics (please refer to SI), is given in green triangles. b) Elemental ratios of OA emissions for the Euro 1 and a Euro 2 scooter as a function of photochemical age. Elemental ratios observed for xylene¹³ and ambient²⁶ SOA are shown, orange and purple, respectively.

Figure 3: Ambient and model data on two-stroke scooters a) Share of total fuel consumption by 2S in 2005, 2010 and 2015 from the Greenhouse gas Air pollution Interactions and Synergies, GAINS, model²⁶. b) Roadside benzene, toluene, ethyl-

benzene, and xylene (BTEX) before and after banning/ restricting 2S scooters in two Chinese cities c) Roadside BTEX and number of all vehicles in three Chinese cities.

Figure 4: Reactive oxygen species in two-stroke scooter emissions The percentage of water soluble reactive oxygen species (ROS) and elemental O:C ratios of organic aerosol as a function of time after lights on in the smog chamber from Euro 1 (red) and Euro 2 (blue) 2S scooter exhaust emissions. ROS concentration measured in moles hydrogen peroxide equivalents is normalised to the molar organic carbon concentration per m^3 inside the smog chamber to give a percentage.







