Reducing paving emissions and workers' exposure using novel mastic asphalt mixtures

Fabian Weissa, Philipp Balohb, Cornelia Pfallerb, Eylem Can Cetintasb, Anne Kasper-Gieblic, Anna Wonaschützc, Mariyan Dimitrovd, Bernhard Hofkoe, Helmut Rechbergere, Hinrich Grothea,∗

a Institute of Material Chemistry, Vienna University of Technology, Getreidemarkt 9 BC 01, 1060 Wien, Austria
b Institute for Chemical Technologies and Analytics, Vienna University of Technology, Getreidemarkt 9 BB 03, 1060 Wien, Austria
c Institute for Aerosol Physics and Environmental Physics, University of Vienna, Boltzmanngasse 5, 1090 Wien, Austria
d Institute of Aerosol Physics and Environmental Physics, University of Vienna, Boltzmanngasse 5, 1090 Wien, Austria
e Institute for Water Quality, Resource and Waste Management, Vienna University of Technology, Karlsplatz 13, 1040 Wien, Austria

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ABSTRACT
Mastic asphalt (MA) is an important construction material mainly used for infrastructure pavements as well as industrial and garage floors. As mastic asphalt construction sites are often limited in space, the prevalent form of construction is by manual paving. Therefore, the emissions caused by mastic asphalt are not only relevant for the environment, but also highly relevant for workers' health. In this study, we examine four mastic asphalt mixtures in terms of their emissions and worker exposure at various construction temperatures, ranging from 195 °C to 245 °C. We conducted three laboratory studies and a field study to evaluate the emissions away from the construction site, emissions in close proximity to the construction site, direct worker exposure and, for the very first time, also the absolute emissions in mass PM10 a.d. per mass of MA. The experiments show that a big part (80 wt %) of the PM10 a.d. emissions consists of particles smaller than 0.8 μm a.d. and are therefore very health relevant. Furthermore, we could show consistently in all our experiments that decreasing the paving temperature by 50 °C (by using a MA mixture workable below 200 °C) leads to an emission reduction of up to 70%.

1. Introduction

Asphalt or bitumen are the common names for the black, highly viscous fluid that is one product of crude oil refinement. One of its main uses today is for road construction as a composite with fine and coarse mineral aggregates forming asphalt mixtures. Nowadays, a wide variety of different mixtures exist, each optimized for a special application in construction, yet often with slight differences depending on the country's construction codes and material specifications. The mixtures may differ in terms of the bitumen grade, the mineralogical composition, the grading curve of mineral aggregates and several additives that alter the behavior of the overall mixture. In many cases the construction codes of a country evolve through a process that seeks out an optimization of cost, availability, safety, sustainability and durability of the material in question.

Here, a special kind of asphalt mixture was the subject of research: mastic asphalt (MA) [1]. MA has various applications for road and building construction and is often used where flexibility is needed and large compaction machines may not be used due to limitations in space. Different from other asphalt mixtures, mastic asphalt is usually paved manually. The work is heavy labor and the properties of the MA mixtures are very important since the application by hand calls for an optimal viscosity and overall workability. The standard mixtures MA8 (8 mm maximum nominal aggregate size) and MA11 (11 mm maximal nominal aggregate size) employed in this study are commonly used as pavement material in urban areas, for walk and bikeways, as well as on steel bridges. An on-going research project is aimed at improving this mixture. Improvements to MA were accomplished by lowering the production and paving temperature, using totally rounded aggregates instead of crushed aggregates and adding wax additive to the mixture. Due to the close proximity of the workers with the material, worker safety is one important topic in this research project.

Bitumen vapor and aerosol (bitumen constitutes ca. 9 wt% of MA) are categorized as class 2 carcinogens (carcinogenic in animal experiments) by the Deutsche Forschungsgemeinschaft [2] and reducing fumes of asphalt works is an important issue also in other applications like roofing [3] or highway paving with hot mix asphalt [4]. Elevated cancer mortality and other issues related with exposure of workers in
road construction were published during the years [5–7]. Hansen et al. [5] reports a three times higher lung cancer risk for mastic asphalt workers than for the general public. These finds are of special interest since the data is solely focused on the handling of mastic asphalt. This is particularly important because a comparison of different asphalts show that mastic asphalt workers have significantly higher exposure to organic matter than most other workers handling other sorts of asphalts [8,9]. For comparison, manual mastic asphalt work is related with organic matter than most other workers handling other sorts of asphalts particularly important because a comparison of different asphalts show that mastic asphalt workers have significantly higher exposure to organic matter than most other workers handling other sorts of asphalts [8,9].

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2. Materials and methods

For the first laboratory study two mixtures of MA11 PMB (conventional and modified; see SI for details) were used. In laboratory study 3 and the study field three MA8 mixtures, eligible for road construction at lower than average paving temperatures (below 240 °C), were compared to a reference mixture. Powdered limestone was the filler component and a mixture of 82 wt% 90/10 penetration graded and 18 wt% 70/100 penetration graded bitumen was the binder component for all mixtures. The coarse fraction was varied from mixture to mixture as totally crushed (TC) aggregates of porphyritic origin and totally rounded (TR) limestone were used separately, but also in combination. The additional wax modifications were accomplished by the use of amide wax (AW). An EN 12697-35 [11] appropriate standard compulsory laboratory mixer (infraTest) with a capacity of 30 l and a reverse rotating function was used for all laboratory experiments. The mixtures used in the field study were produced at a local mixing plant.

The 4 mixtures used were:

Reference Mix: MA8 100% TC – production temperature: 245 °C
MA8 100% TC 3% AW – production temperature: 230 °C
MA8 50% TC 50% TR – production temperature: 220 °C
MA8 100% TR 3% AW – production temperature: 195 °C

These mixtures (for recipe details see SI) and the respective production temperatures were chosen after a test at the mixing plant, where construction workers tried applying them in a small area and evaluated their workability.

2.1. PM sampling

Particulate matter (PM) samples were collected on quartz fiber filters (Pallflex Tissuqartz 2500QAT-UP) using low-volume samplers (sample flow 2.3 m³/h) equipped with size selective inlets (PM10 - particulate matter with an aerodynamic diameter (a.d.) below 10 μm). Only during laboratory study 1 was no size cut installed. Size-segregated samples were collected using a low pressure impactor with size cuts at 12.8, 3.2, 0.8, 0.2, 0.06 and 0.015 g. Particulate matter with an aerodynamic diameter (a.d.) below 200 μm, allowing for a large reduction in emissions. Using low temperature mixtures is an important step towards promoting the safety of workers handling MA and towards reducing the environmental impact of MA road construction.

![Fig. 1. Size distribution of the modified MA mixture in laboratory study 1.](image-url)

Table 1

<table>
<thead>
<tr>
<th></th>
<th>Conventional MA11</th>
<th>PM10 [mg/m³]</th>
<th>OC [mg/m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filter 1 m</td>
<td>9.7</td>
<td>9.7</td>
<td></td>
</tr>
<tr>
<td>Filter 2.5 m</td>
<td>7.5</td>
<td>6.7</td>
<td></td>
</tr>
<tr>
<td>Modified MA11</td>
<td>2.7</td>
<td>2.4</td>
<td></td>
</tr>
<tr>
<td>Filter 1 m</td>
<td>2.3</td>
<td>2.1</td>
<td></td>
</tr>
</tbody>
</table>

Differences in chemical composition of the asphalt mixtures were explored using a Laser Ablation Aerosol Particle Time of Flight Mass Spectrometer (LAAPTOF, AeroMegt GmbH). The LAAPTOF single particle mass spectrometer samples aerosol through an inlet with a critical orifice of 100 μm diameter (flow rate of 75 cm³/min⁻¹). An aerodynamic lens focuses the particles into a narrow beam, separating them from the ambient gas and transitioning them into a vacuum (around 10⁻⁷ mbar). Single particles pass through two detection laser beams (wavelength 405 nm), creating a scattered light signal during transit. The light signal of the second detection laser triggers a 193 nm excimer laser pulse (ATLEX 300 I, ATL Lasertechnik GmbH, Germany), which ionizes the detected particle. The produced ions pass into a bipolar time-of-flight mass spectrometer (B-TOF, TOFWerk AG), and their flight time is measured. For each particle a positive and a negative mass spectrum is recorded. The lower and upper size detection limits of the LAAPTOF, determined by PSL calibration, are 390 nm and 1540 nm, respectively. The overall detection efficiency of the LAAPTOF is about 2.5% [13]. The Time-of-Flight mass spectrometer was calibrated with an aerosol generated by nebulizing a suspension of Carbon Black (Elftex 124, Cabot Corporation) in an 80/20 water/isopropanol mixture. The LAAPTOF single particle spectra (just like those from other laser ablation single particle instruments [1-4]) allow for qualitative determination of particle composition deduced from the ionic fragments produced by the ablation, and for analysis of the particle population, e.g. groups of differing chemical composition. The LAAPTOF mass spectra were processed, calibrated, and converted to stick spectra using software provided by AeroMegt GmbH. The large number of individual spectra was grouped using a fuzzy c-means algorithm [15]. As the aerosol turned out to be homogeneous, a single cluster center is reported here.
Fig. 2. Representative spectrum for conventional and modified MA. The representative spectrum of the wax (see Fig. 3) is included for comparison. Main panel: $-100 < m/z < +100$, with suggested peak identification, insert: $m/z$ – 380 to 0; marker peaks for the wax are visible in the modified, but not in the conventional MA spectrum.

Fig. 3. Representative spectrum of the wax. a) range $m/z$ $-400$ to $+400$, b) range $m/z$ $-100$ to $+100$ with suggested peak identification.

Fig. 4. Representative spectra of conventional and modified bitumen; the representative spectrum of the wax (see Fig. 3) is included for comparison. Main panel: $m/z$ $-100$ to $+100$, with suggested peak identification, insert: $m/z$ $-380$ to 0; marker peaks for the wax are visible in the modified, but not in the conventional bitumen spectrum.
Table 2
Absolute emission of mastic asphalt mixtures; percentage relates to the emission of the reference mix.

<table>
<thead>
<tr>
<th>MA mixture</th>
<th>PM10 [mg/kg]</th>
<th>[%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>MA 100% TC 245°C</td>
<td>5.52</td>
<td>100</td>
</tr>
<tr>
<td>MA 100% TC 3% AW 230°C</td>
<td>4.47</td>
<td>81</td>
</tr>
<tr>
<td>MA 50% TC 50% TR 220°C</td>
<td>3.97</td>
<td>72</td>
</tr>
<tr>
<td>MA 100% TR 3% AW 195°C</td>
<td>1.61</td>
<td>29</td>
</tr>
</tbody>
</table>

Table 3
Values from field study sampling; percentage relates to the value of the reference mix for each column.

<table>
<thead>
<tr>
<th>MA mixture</th>
<th>Close proximity</th>
<th>Personal exposure</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PM10 [μg/m³]</td>
<td>[%]</td>
</tr>
<tr>
<td>MA 100% TC 245°C</td>
<td>3,79</td>
<td>100</td>
</tr>
<tr>
<td>MA 100% TC AW 3% 230°C</td>
<td>182</td>
<td>48</td>
</tr>
<tr>
<td>MA 50% TC 50% TR 220°C</td>
<td>131</td>
<td>35</td>
</tr>
<tr>
<td>MA 100% TR AW 3% 195°C</td>
<td>60,5</td>
<td>16</td>
</tr>
</tbody>
</table>

Fig. 5. Particle size distribution during construction work at the field study; lowest temperature is missing because the collected mass for each size channel is too low.

Fig. 6. Field study – Background measurement in grey; sampling station of the Vienna ambient air monitoring network in black; construction days: Aug. 8, 9, 12 and 16. as a representative spectrum for each sample. The aggregate spectrum – the maximum signal of all spectra at each m/z value – was used to evaluate specific peak locations.

3. Experiments

Three laboratory studies and a field study were set up: (i) laboratory study 1 was aimed at characterizing the emissions from conventional as well as from modified asphalt in a controlled environment. (ii) laboratory study 2 was designed to investigate qualitative chemical differences in the emissions of wax-modified and non-modified bitumen. (iii) laboratory study 3 focused on determining the absolute emissions of the asphalt mixtures used per kilogram of mass. (iv) During the field study, conditions next to an actual pavement construction site were monitored (ambient concentrations and personal monitoring) for in-situ quantification of emissions over the entire construction period.

3.1. Laboratory study 1

This study was conducted in an enclosed laboratory area at TU Vienna IVWS/RELLA Halle on two days, Sep. 23, 2014, and Dec. 12, 2014. The room was equipped with a laboratory mixer, where two different MA11 PMB mixes (see SI) were prepared by mixing for 30 min. Particulate matter (PM) samplers were placed at a distance of 1 m and 2.5 m from the laboratory mixer. The LAAPTOF inlet was located about 2 m away from the emission source. Due to the extremely high concentrations (up to 10 mg/m³) of emitted aerosol from the asphalt mixer, the contributions from pre-existing aerosol particles were negligible. With the LAAPTOF, over 30 separate measurement samples (5 min each, yielding a total of several ten thousand spectra) were taken. For this study, a representative measurement for each day was chosen for detailed analysis: Sep. 23, 2014, 13:32–14:34, and Dec. 12, 2014, 14:19–14:21.

3.2. Laboratory study 2

For laboratory study 2, conducted at the University of Vienna aerosol laboratory on Dec. 1, 2016, and Dec. 6, 2016, the experimental setup consisted of a hot plate, stirrer, temperature gauge, and a can containing the sample. Two types of sample were used: i) conventional bitumen (CB) consisting of a mixture of 82 wt% 90/10 penetration graded and 18 wt% 70/100 penetration graded bitumen. ii) the same bitumen with 3 wt% AW as a wax additive (modified bitumen MB) and iii) the wax by itself (AW). The hotplate with the sample was placed into a fume hood and heated at a fast rate (roughly 10 °C/min) to 140 °C, then slowly (roughly 1 °C/min) to 235 °C. Throughout the heating process, the LAAPTOF sampled aerosol particles from the fume hood through an 8 mm polyurethane tube of about 6 m length. The inlet of the tube was located about 20 cm from the sample container. 23 samples (5 min each) of aerosol particles emitted by the heated bitumen and wax at different temperatures were taken. For detailed analysis, samples at two temperatures were chosen: 200 °C, representing the paving temperature of the modified asphalt, and 230 °C, representing the paving temperature of the conventional asphalt. As the heating process of the bitumen/additive was hard to track due to the phase change of the sample (from almost solid to very viscous, turning gradually more liquid from the bottom of the container upward), leading almost certainly to an uneven heating of the sample, these temperatures are rough estimates only. We assume an uncertainty in sample temperature of at least ± 10 °C. For the additive, a measurement at one representative temperature of 210 °C (AW210) was chosen for analysis.

3.3. Laboratory study 3

Laboratory study 3 was conducted at TU Vienna IVWS/RELLA Halle (24.02.2017, 01.03.2017–03.03.2017). For these experiments four mixtures (as described in Material and Methods) were used. For each experiment (Fig. S3 shows the procedure) one mixture was prepared in the laboratory mixer in the morning and one in the afternoon (one recipe a day). The mixture was transferred into a standard wooden
bucket as used by asphalt workers on construction sites. This bucket was weighed and then carried to another room (~20 m away), where the temperature of the mix was measured. The bucket was emptied onto a steel plated wooden plate covered with aluminum foil. The asphalt mix was covered with a fume hood with a filter sampler attached. During a period of 30 min the filter was loaded with air drawn through the fume hood to sample particulate matter evolving from the asphalt mixtures. After 30 min the hood was removed and the temperature of the mix was measured again to ensure that cooling had occurred and that further emissions could be regarded as negligible (samples were below 80 °C after 30 min). As the sample flow was 37.7 l/min, the volume of the fume hood was purged 9.4 times during the sampling interval of 30 min. To ensure a mixing within the fume head, the ambient air was let in on all sides on the bottom of the fume hood. As no cleaning of the ambient air entering the fume hood was performed, parallel PM sampling was performed in close proximity to determine background concentrations for comparison.

3.4. Field study

Field measurements were conducted during the construction of a test road of 300 m length with the respective asphalt mixes. This testing field was in a residential area of Vienna. The experiments took place on five days of August 2016 (8.8.2016, 9.8.2016, 11.8.2016, 12.8.2016, 16.08.2016). On each day only one specific mix was applied at the construction site (except for the reference mix, which was applied on two days). Two PM10 filter samplers and a cascade impactor for size segregated sampling were placed next to the segment under construction, approx. 0.5 m off the road. Additional measurements aimed to evaluate the workers’ exposure to particulate matter. Two of the workers were equipped with personal PM10 samplers. One of the workers’ tasks was to apply MA8 onto the street (Worker 1), and the other’s was to fill the buckets at the asphalt boiler (Worker 2) (see SI for a picture). To determine ambient particulate matter concentrations (PM10) characteristic for the site and to account for a possible influence of the paving activities on the local PM10 levels, sampling with an automated system was performed for a slightly longer time period (5.8.2016–19.8.2016) at a distance of approx. 100 m from the construction site.

4. Results and discussion

4.1. Laboratory study 1

Table 1 shows the median values of the particulate matter sampling in laboratory study 1. Particulate matter concentrations determined in close vicinity to the laboratory mixer showed significant differences between the conventional MA11 mix at 245 °C and the modified MA11 mix at 190 °C (see Table 1). Although the sampling set-up does not justify deducing an emission flux, the difference between the measurements can be taken as a strong indication of reduced emissions of particulate matter mass (up to 70% for both distances sampled, 1 m and 2.5 m) for the modified MA mix. Most of the particulate matter sampled was organic carbon (>88%). We did not find particles of the mineral fraction in the sampled aerosol particles via LAAPTOF MS. The particle size distribution shown in Fig. 1 was determined during the mixing of the modified MA and shows that the emitted particles in this laboratory experiment are mostly below 0.8 μm a.d. In fact, 80% of all sampled mass could be attributed to the PM1 fraction, which leads to the following conclusion: 80% of all sampled mass is respirable and therefore highly health relevant [16].

Fig. 2 (main panel) shows the representative stick spectra for conventional and modified MA11. The spectral peaks indicate a mostly organic carbonaceous aerosol (m/z +12 to +15, +24 to +29, +36 to +41 and +48 to +58, as well as ~24 to ~26, with prominent peaks at +27, +29, +37, +43 [17]). The combination of peaks at ~97 and ~33 indicates sulfate [18], which exhibits a larger signal intensity in the conventional than in the modified asphalt. At first glance this seems to be the only qualitative chemical difference between conventional and modified MA.

4.2. Laboratory study 2

The laboratory experiments on bitumen and wax additive (AW), however, provided more insight: Fig. 3 shows the representative spectrum for AW. At low m/z (Fig. 3b), a clear organic carbon signature is visible, along with evidence of ambient aerosol (sulfate peak at m/z ~97, nitrate peaks at m/z ~46 and ~62), which likely served as a condensation surface for hot vapor emitted from the sample. At high m/z (Fig. 3a) two very conspicuous peaks are evident. The mean peak locations are m/z ~255.85 and m/z ~283.87 (FWHM 1.88 and 2.05, respectively). We call these peaks “m/z 256” and “m/z 284” for the remainder of this discussion.

To gain insight into the extent to which the wax can be detected in the modified bitumen mixture, the presence or absence of these spectral peaks in the bitumen samples needed to be determined. A more pronounced “elemental carbon signature” (peaks at m/z ~12, ~24, ~36, ~48, etc., as well as m/z ~12, ~24, and ~36) is visible in the bitumen samples, which was not observed in the AW (Fig. 4). The inset in Fig. 4 shows the same spectra in the mass range m/z ~380 to 0, with a logarithmic y-axis for better distinction. It is obvious that the spectral peaks m/z ~256 and ~284 are present in the modified, but not in the conventional bitumen.

Finally, the question of whether the two peaks could be identified in the modified asphalt had to be settled. If so, a clear chemical signature of the additive would be identifiable in the modified asphalt emissions. This is shown in Fig. 2 (insert). While the peaks m/z ~256 and ~284 are less conspicuous than in the modified bitumen samples, they are clearly above the noise level for the modified asphalt, and equally clearly absent in the conventional asphalt. Thus the additive has been identified in the modified asphalt emissions through its marker peaks m/z ~256 and m/z 284.

4.3. Laboratory study 3

The goal of laboratory study 3 was to assess the emission caused by an MA mix in relation to mass. With this experiment we were able to measure absolute amounts of aerosol coming from the hot asphalt mixtures for the first time. The results are listed in Table 2. The data shows the same trend as laboratory study 1 and, when looking at a similar temperature difference (245 °C–195 °C vs. 240 °C–190 °C), the emission reduction that took place is in the same range (70%). These absolute emission values have to be regarded as lower limits as a loss of particles may have occurred already within the fume hood. Sampling of blank filters through the empty fume hood prior to and after the actual measurement sampling suggests an underestimation of emission values of roughly 10%. Furthermore, in the experiment the MA was applied onto the surface by emptying the bucket and letting the MA spread by itself. On construction sites the MA is emptied onto a surface and then spread manually by a worker, which leads to a faster creation of surface area and a certain manual mixing of the MA. The difference in these two procedures may lead to higher particle emission of the MA on the construction site than the laboratory study suggests.

4.4. Field study

The measurements of the field study, shown in Table 3, confirm the trend observed in the laboratory studies. Measurements at the curb site, next to the paving activities, showed a clear dependence of particulate matter concentrations on the temperature of the respective asphalt mixtures, with higher temperatures leading to elevated mass concentrations. In all cases, mass concentrations on site (ranging from 60 to
Personal exposure of construction workers yielded mass concentrations reaching up to nearly 4 mg/m³ (Table 3). Again, elevated working temperatures of the asphalt mixtures led to higher mass concentrations. No connection between the worker filling the buckets at the asphalt boiler and the one applying the mix to the road is evident. The reported values compare well to other studies on similar construction work. For example, Rühl et al. [9] reported aerosol exposure values ranging from 1.74 to 3.12 mg/m³ and Spickenheuer et al. [19] reported values between 1.8 and 4.92 mg/m³ for manual outdoor work with mastic asphalt in similar job tasks as the workers in this study. Still, we have to keep in mind that our numbers rely on a limited number of measurements and can be influenced by the behavior (e.g., movements) of the worker and local influences (see Table 3 - worker 2 has higher exposure at 230 °C than at 245 °C/meteorological data during the field study is listed in the SI).

The background sampling before, during and after construction allows the possible influence of the construction activities on the local environment to be evaluated. The results are shown in Fig. 6, which summarizes the mass concentrations determined at the site approx. 100 m away from the actual construction activities as well as at a sampling site of the Vienna ambient air monitoring network roughly 1 km away from the measurement site, taken as a reference to show that concentrations determined at our background site reflect the general conditions observed in Vienna. Agreement between the mass concentrations observed at the two sites was high ($r^2 = 0.8847$). The construction days were August 8, 9, 12 and 16. Fig. 6 shows that there is no obvious difference in concentration, so we conclude that the construction activities had no impact on the local ambient air quality experienced by the general public.

5. Conclusion

Paving 1 km (width 3.5 m; depth 3.5 cm) using the reference mixture at 245 °C results in 1.623 kg PM 10 a.d. just from the paving process (no production, transport, storage etc.). When using the MA8 100% TR 3% AW at 195 °C as pavement, the resulting emission is 0.473 kg PM 10 a.d.. Therefore, per km pavement savings of 1.15 kg PM 10 a.d. emission is possible, which is obviously a small value compared to the emission of cars using the road [20]. Furthermore, the ambient air measurements in a distance of 100 m to the construction site showed no relationship between air quality and the construction activities. Nonetheless, the implications for worker health are relevant in any case. Mastic asphalt emission proved to be of rather small particular size (over 70 wt% of particles are below 1 μm a.d.). This suggests deep penetration and long retention time in the human lung [21–23]. Workplace concentrations remained below the threshold of 10 mg/m³ suggested by the BITUMEN forum [24]. Still, if we look at Rumler et al. [25], we see that the impact of bitumen fumes and aerosols on workers’ health increases with increased exposure time even if the suggested threshold is not exceeded. Several other publications [26–28] have reported on the problems arising in MA construction. Using an MA mixture workable below 200 °C that cuts the emission by 70% constitutes a significant advantage for the environment, the workers and also the employers.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.buildenv.2018.03.060.

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