Atmospheric tar balls: primary droplets from biomass burning?

Á. Tóth[1], A. Hoffer[2], I. Nyírő-Kósa[2], M. Pósfai[1], and A. Gelencsér[1, 2]

[1] Department of Earth and Environmental Sciences, University of Pannonia, Veszprém, P.O. Box 158, H-8201, Hungary
[2] MTA-PE Air Chemistry Group of the Hungarian Academy of Sciences, University of Pannonia, Veszprém, P.O. Box 158, H-8201, Hungary
Keywords: tar ball, formation mechanism, primary emission, biomass burning.
Presenting author email: ubul.toth@gmail.com

Biomass burning is a globally important source of anthropogenic carbonaceous aerosol. Atmospheric tar balls occur in large numbers in relatively fresh biomass burning plumes (Pósfai *et al* 2003). These particles can be easily recognized by transmission electron microscopy (TEM) by their shape and composition. In contrast to other spherical aerosol types such as sulphates the amorphous, spherical tar ball particles are stable and do not volatilize under the electron beam. These particles differ from soot particles as well, since they do not contain a microstructure of concentric graphene-like layers. Due to their absorption properties tar ball particles have an important role in the Earth's radiation budget (Alexander *et al* 2008). These particles typically occur externally mixed with other particles in the atmosphere. According to elemental analyses of individual particles, tar balls consist mostly of carbon, with a minor amount of oxygen and may contain only traces of sulphur, potassium, chlorine and silicon. The average C/O molar ratio is about 10 (Pósfai, *et al* 2004).

It has been hypothesized that tar ball particles form in secondary processes from pyrolysis products in the atmosphere (Pósfai, *et al* 2004). However, these particles are homogenous, they do not contain an internal core and form a distinct group of particles with diameters up to 500 nm. On the basis of these features we now hypothesize that tar ball particles are ejected as droplets of tar, produced during the pyrolysis and are emitted from inside the vascular tissues and other pores of the burnt plants.

In this study we carried out a laboratory experiment to produce tar ball particles under conditions simulating the hypothesized formation mechanism but with the total exclusion of flame or fire. A closed all-glass apparatus was constructed in which droplets were generated from tar-water emulsion by bubbling purified air or nitrogen through it. The droplets were passed through a heated zone at 450 ^{o}C to simulate thermal processes occurring in the fire zone. Then the airflow was cooled and dried with excess dry air and the particles were collected on grids for TEM analysis.

The tar-water emulsions used in the experiments were prepared previously by dry distillation of various plants (e.g. needles of *Pinus ponderosa* and logs of Turkey oak).

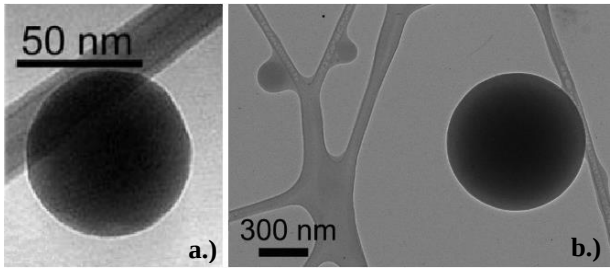

Figure 1. Bright-field TEM images of (**a.**) atmospheric tar ball (Pósfai *et al.*, 2004); (**b.**) laboratory-generated tar ball.

The morphology and elemental compositions of the particles were studied using TEM with energy-dispersive X-ray spectrometry (EDS). The generated particles were perfectly spherical and similarly to atmospheric tar ball particles, they were homogeneous (did not contain a condensation core) as it can be seen in *Figure 1b*. The C/O molar ratio of the laboratory-generated tar ball particles were also in the range reported for atmospheric tar balls. Our results imply that the suggested primary mechanism may be a feasible pathway of tar ball formation in biomass burning, but further studies are clearly warranted.

Present work was published in the frame of the project ***TÁMOP-4.2.2.A-11/1/KONV-2012-0064***. The project is realized with the support of the European Union, with the co-funding of European Social Fund.

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