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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Persistent primary organic tar particles during the regional wintertime hazes in North China: insights into their aging and optical changes" *by* Lei Liu et al.

Anonymous Referee #2

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This study describes and compares wintertime aerosol particles in a rural location on the North China Plane with aerosol in Beijing (downwind from the rural site): aerosol concentration, chemical composition and individual-particle properties were measured, and optical properties simulated. The measurement campaign covered a period when conditions changed from relatively clean to heavily polluted, and the aerosol at the assumed source (the rural site) and in the urban haze could be compared. The major finding is that refractory primary organic particles from coal and biomass burning dominate at the rural site, and similar but coated particles are the major constituents in the aged haze. The inorganic coatings change the optical properties of the aerosol, by enhancing their absorption. The paper is sound and fairly well written. I have three

concerns, one of which is partly a terminological issue.

(1) Primary Organic Tar (POT). I am slightly uncomfortable with the use of the term "primary organic tar" (and its acronym, POT) for the observed class of particles. From the results it seems a correct interpretation that these particles are primary organics emitted by burning of coal and biomass. However, this is an interpretation and not an observation, as is now presented right at the start of the paper. The properties we know from TEM include the morphologies of the particles (some spherical and others irregular), their C-rich nature (although the particles were collected on a thick carbon+plastic film and therefore we do not know how much C they contain), and their thickness from SEM which suggests they were quite viscous when they arrived onto the grids. Indeed, these properties and the high concentrations at the locality where burning of coal is a major source of particles suggests they are primary organics. However, I am not sure they can be called "tar", since there is no study of the speciation of their components (although the authors refer to Xu et al. 2019 who used AMS and identified three groups of burning-related factors). Admittedly, "tar" is a vaguely defined substance. However, the atmospheric chemistry literature is already replete with vaguely defined terms (and acronyms), and introducing a new one, such as POT may not help to clear the confusion. For the spherical types of such particles already exists a widely used term, "tar ball". And about tar balls we know they are primary (Tóth et al. ACP 14, 6669–6675, 2014), indeed made of tar (Tóth et al. ACP 18, 10407–10418, 2018), and even their optical properties are known (Hoffer et al. ACP 16, 239–246, 2016). By using POT, I feel the definition of tar ball is diluted and thereby the utility of the term diminished. In summary, I believe it may be more prudent to call the POT particles just burning-related primary organics (PO), and it could be mentioned that the spherical ones appear to be tar balls. But, in the end, it is up to the authors what they wish to call these particles.

(2) Optical properties. The calculations for coated absorbing organic particles are interesting but I do not completely understand Figure 10. (1) What is the explanation for the insensitivity of ACS enhancement to the Dp/Dc size ratios larger than about 1.5?

(2) What is the reason for the apparently random variation of ACS enhancement with Dc? I wonder if any optical measurements were done under hazy conditions during the sampling campaign? If such results are available, it would be interesting to compare them with the models.

(3) Size changes. If the particles were transported from the rural site to Beijing, and during transport secondary aerosol formed on them, it does not see logical that their sizes decreased. The explanation offered is that large particles deposited but this process does not seem feasible for the particles in this size range. Can you please provide some more discussion on this issue?

Minor comments:

29: "average particle-to-core ratios.."- it is unclear what ratios; probably "..diameter ratios" would be correct

32: replace "we using.." with "Using Mie theory we estimated.. "

61: chromatography

64-66. subject of sentence unclear

85 "properties" instead of "information"

178-179 "five grids.. were selected" - I guess "five grid meshes. . . were selected" would be correct

191: primary organic tar, POT: I do not understand the utility of this name and abbreviation. First, tar should be organic anyway, so "organic tar" is a tautology. Second, how do you know it is tar?

195-197: Hoffer et al. ACP 16, 239–246, 2016 found 1.84 - 0.21i for tar balls - I wonder if this would make a large difference in your calculations, compared to the value by Alexander et al.

246-249: unclear sentence, please reword

266-267 and text that follows: "TEM observations show a distinct group of spherical and irregular primary organic particles.." - I think TEM observations indeed show the morphologies of the particles but they do not provide direct information on their "organic" nature (only their elemental composition), let alone their primary origin. Please try to distinguish observations from interpretation.

278: Could it be just PO?

295: "agrees" instead of "consists"

372: influences

376-377: how were the absorption cross sections estimated?

405-409: this seems important but I do not understand the statement completely.

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