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Microplastic and Associated Black Particles From Road-Tire Wear: Implications for Radiative Effects Across the Cryosphere and in the Atmosphere

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Key Points:

- Light absorbing particles, as blackened microplastic matter from road-tire wear, occur in snow of the Colorado Rocky Mountains (2013–2021)
- The mass of road-tire-wear particles annually generated is estimated to be 6,550 kt, of which about 655–1,965 kt might become airborne
- The results are relevant to radiation modeling of snow and ice surfaces as well as the atmosphere

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract The environmental effects of airborne micro- and nano-size plastic particles are poorly understood. Microscopy and chemical analyses of atmospherically deposited particles on snow surfaces at high elevation (2,865–3,690 m) in the Upper Colorado River basin (UCRB; Colorado Rocky Mountains) revealed the presence of black substances intimately associated with microplastic fibers, particles interpreted to have originated as tire matter. Identical and similar particles occur in shredded tires and road-surface samples. The substance responsible for the black color of all tires is carbon black, a graphitic light-absorbing tire additive produced by hydrocarbon combustion that homogeneously permeates the mixture of tire polymers and other additives. Such black tire matter may thus exert radiative effects closely similar to those of black carbon. The presence in snow of many organic compound types common to tires, measured by two-dimensional gas chromatography, suggests that atmospherically deposited black road-tire-wear matter is among the light-absorbing particulates that advance the onset and rate of snow melt in the UCRB. The mass of road-tire-wear particles shed from vehicles may be estimated by multiplying measured amounts of eroded tire-per-distance traveled by vehicular distances. Under a combination of measurements and assumptions about the amounts and radiative properties of atmospheric tire-wear particles, the radiative effects of these particles might add about 10%–30% to those effects from black carbon, an estimate ripe for revision. On regional and global scales, the amounts and effects of emitted and deposited tire-wear matter likely vary by factors of geographic source, transport pathway, and depositional setting.

Plain Language Summary A major source for airborne microplastic particles is road-tire wear. Microscopy and chemical analyses of wind-blown particles on dirty, high-elevation (2,865–3,690 m) snow surfaces in the Colorado Rocky Mountains revealed the presence of black substances intimately mixed with microplastics, particles interpreted as tire matter. Identical and similar particles occur in shredded tires and in samples collected from road surfaces. The black substance responsible for the black color of all tire particles is carbon black that homogeneously permeates the mixture of tire polymers and other additives. The mass of particles produced by road-tire wear may be estimated by multiplying measured amounts of eroded tire-per-distance traveled by vehicular travel distances. Under assumptions of amounts of tire-wear matter emitted to the atmosphere, their presence as light-absorbing particles likely contribute to melting of snow and ice as well as to warming the atmosphere.

1. Introduction

The presence of globally distributed fragments of plastic in marine, terrestrial, and atmospheric environments is generating multiple lines of research to determine their sources and many effects. As one area of concern, the influences of micro- and nano-plastic particles (MPs, <5 mm in any dimension) on radiative properties across the cryosphere and in the atmosphere remain poorly understood (e.g., Allen, S. et al., 2022; Allen, D. et al., 2022; Brahney et al., 2020; Evangeliou et al., 2020; Ming & Wang, 2021; Revell et al., 2021). Reasons exist to suspect that MPs can exert radiative forcing (RF) to heat the atmosphere and diminish albedo of snow and ice surfaces. Firstly, plastics absorb solar radiation as indicated by spectral methods such as reflectance, Raman, and FTIR (Brahney et al., 2020; Hidalgo-Ruz et al., 2012; Kokaly et al., 2017), regardless of type and color. Secondly, dark

colors (e.g., blue, red, green) of some MPs would increase their capacities to absorb light (Brahney et al., 2020; Evangeliou et al., 2020; Ming & Wang, 2021; Revell et al., 2021).

With respect to snow-surface albedo affected by atmospherically deposited dust, Ming and Wang (2021) noted that the relative RF effects of black carbon (BC) and MPs are unknown because of (a) their typical co-occurrence as separate classes of particulates and (b) the inability of commonly used methods to differentiate between them. In this study of airborne light-absorbing particles (LAPs) in snow, however, we suspected that some microplastic-particle (MP) particles and black, light-absorbing matter had a common origin. This common origin is road-tire wear, the particles from which are considered to be the dominant global source of MPs (Brahney et al., 2021; Evangeliou et al., 2020; Kole et al., 2017; Sommer et al., 2018; Vogelsang et al., 2020; Wagner et al., 2018).

The light-absorbing capacities of common, airborne MP fibers and flakes, which are translucent, cloudy white, or colored, are small relative to equivalent amounts of some other airborne LAPs including BC (Bond et al., 2013; Flanner et al., 2021; He et al., 2018; Klimont et al., 2017; Petzold et al., 2013), certain minerals, such as ferric oxides (Alfaro et al., 2004; Moosmüller et al., 2012; Moskowitz et al., 2016; Reynolds et al., 2020), as well as dark rock and mineral particles (Reynolds et al., 2020). The relatively low light-absorbing capacity of plastic (some polymers) is illustrated for spectral reflectance by Kokaly et al. (2017). Ming and Wang (2021) suggested, nevertheless, that the mass of MPs today likely exceeds that of BC on the basis of a study by Dubaish and Liebezeit (2013). Focusing on the roles of LAPs to diminish the albedo of snow surfaces, Ming and Wang (2021) consequently inferred that MPs exert a greater RF influence on snow surfaces than do BC particles. The distinction among different LAPs is important for attribution to sources and degrees of RF because differently sourced LAP types and amounts would exert variable geographic effects across the cryosphere and in the atmosphere. A semantic clarification is that most components of a road tire–its rubber, as well as various synthetic polymers and elastomers–are plastics so that effectively all <5-mm tire-wear particles (TWPs) are microplastics, as defined by Hartmann et al. (2019).

Considering that road-tire wear is a major source of MPs, the compositions of road tires are central to understanding their RF effects. As summarized by Knight et al. (2020) and Zhang et al. (2023), road tires contain many different components: The tire tread of an all-season light-duty (passenger) vehicle may contain many different polymeric combinations of natural and styrene-butadiene rubbers (NR and SBR, respectively; about 30% by mass), many other hydrocarbon-based polymers and elastomers, fillers of carbon black, silica, chalk, clay, barite, talc, various antioxidants and antiozonates, as well as sulfur- and zinc-containing vulcanization agents (Baensch-Baltruschat et al., 2021; Eisentraut et al., 2018; Gao et al., 2022; Kole et al., 2017; Kreider et al., 2010; Lassen et al., 2015; Roy et al., 2020; Rødland et al., 2023; Sommer et al., 2018; van Mourik et al., 2016; Wagner et al., 2018). Heavy-duty-vehicle tires are similar to those of light-duty vehicles but differ in the amounts and types of polymers, elastomers, vulcanization agents, and fillers.

Additionally, at least 3,637 organic compounds have been detected within 400 analyzed tires on light- and heavy-duty vehicles (Molden, 2023a, 2023b). Boisseaux et al. (2024) also documented numerous organic compounds in tires. With respect to metals in these 400 tires, O'Loughlin et al. (2023) reported Al, Fe, and Mg as the most common metals and noted, in addition, that their and other studies have found the presence in tires of As, Ca, Cd, Cr, Cu, K, Mo, Na, Ni, Pb, S, Sb, Se, Si, Ti, V, and Zn. Zinc, in the range 0.06–1.55 weight % (Councell et al., 2004) and commonly estimated by average at approximately 1 weight %, has been considered as a diagnostic component for tire matter because it is used in nano-size particles as a vulcanization agent. Such a broad assumption of a zinc fingerprint, however, has been dismissed as unreliable for several reasons (e.g., O'Loughlin et al., 2023), among them: (a) Zinc readily leaches from tires (Degaffe & Turner, 2011; Selbes et al., 2015; Smolders & Degryse, 2002; Vashisth et al., 1998; Wik & Dave, 2009) and so may not always be detected in shredded tires, (b) in its small amounts, zinc nano-particles may be heterogeneously distributed and are thereby not always found when using narrow-target SEM-EDS methods, and (c) its variable abundance in tires (Harrison & Alghamdi, 2023; Pant & Harrison, 2013). A related point is that fully reliable chemical tracers for TWPs have not been currently adopted, whether in forms of metals or organic compounds (e.g., Harrison & Alghamdi, 2023). Disintegrated tires, such as those found along roadsides, release substances beneath the tread and undertread that include metals from belts and a variety of polymer fabrics from plies (e.g., rayon, polyester, or nylon cords).

Of importance with respect to cryospheric and atmospheric RF is carbon black, a graphitic, light-absorbing substance produced by combustion of hydrocarbons (Bandurin et al., 2022; Jäger et al., 1999). During

manufacture of a tire, carbon-black additive homogeneously permeates natural and synthetic polymers to impart uniformly the black color to tires (Barbin, 2018; Okel & Rueby, 2016). Between 22 and 40 weight % of tires for a light-duty vehicle is carbon black (Kole et al., 2017).

Many different methods have been used to determine the physicochemical properties of TWPs derived directly from road tires as well as those produced by agglomeration of abraded road and tire debris termed road- and tire-wear particles (RTWPs) (Beji et al., 2020, 2021; Camatini et al., 2001; Dahl et al., 2006; Dall'Osto et al., 2014; Gao et al., 2022; Goßmann et al., 2021; Järlskog, Jaramillo-Vogel, Rausch, Gustafsson, et al., 2022; Järlskog, Jaramillo-Vogel, Rausch, Perseguers, et al., 2022; Klöckner et al., 2019, 2021; Knight et al., 2020; Kovochich et al., 2021; Kreider et al., 2010; Kwak et al., 2013; Materić et al., 2020, 2021; Rausch et al., 2022; Rødland et al., 2022, 2023; Sommer et al., 2018; Wagner et al., 2018; Yan et al., 2021; Youn et al., 2021; Zhang et al., 2023). Variations in the physicochemical properties of TWPs and RTWPs arise from many factors, including different tire compositions, road surfaces and compositions, weathering, and driving conditions (Goßmann et al., 2021; Kim et al., 2021; Klöckner et al., 2019; Liu et al., 2022). Herein, we confine our subject to TWPs and RTWPs.

2. Study Aims

Two aims guided this study. One aim was to assess a possible source for observed black carbonaceous matter in snow directly from carbon-black-infused worn tires and asphaltic road surfaces. Road-tire-wear particles have been listed as a source of BC (Klimont et al., 2017), but the linkage among TWPs, forms of MPs, types of BC, and ultimately their combined RF contributions remains incompletely documented. Our immediate application was to comprehend better the airborne components that contribute to diminished albedo of snow surfaces during spring melt in the Upper Colorado River basin (UCRB), Colorado Rocky Mountains, leading to advanced onset and acceleration of snow melt (e.g., Abolafia-Rosenzweig et al., 2022; Deems et al., 2013; Painter et al., 2010, 2012). Broader applications may be considered for other cryogenic surfaces (Flanner et al., 2007, 2021; He, 2022; Liu et al., 2020; Skiles et al., 2018). The first aim underpins the second, which was to approximate masses of TWPs by combining measures of tire-wear mass per driven distance with estimates of tire-wear-particle airborne emission. Such approximations may be compared with regional and global scale estimates of atmospherically deposited MPs and BC.

3. Materials and Methods

Eighty-four snow-surface samples were collected from 14 high-elevation (2,865–3,690 m) sites in UCRB of the Colorado Rocky Mountains, USA (Figure S1 and Table S1 in Supporting Information S1). The samples were taken during late spring with small non-plastic shovels just before the complete disappearance of snow. As such, each sampled snow layer—the All Layers Merged (ALM) sample—contained particulate matter from all winter- and early spring-season airfall from individual dust storms and fugitive particulates. The snow samples were placed in plastic carboy jugs and then transported to a U.S. Geological Survey laboratory in Denver, Colorado. Melted snow was evaporated at 45-degrees C. To the extent possible, plant matter, such as pine needles, was removed by hand after drying. Processing samples from transport in carboys to drying on evaporation trays exposed snow to white plastic raising the possibility of contamination in the form of white particles. (The processing protocols were established before MPs were recognized.) Particles were collected from three asphaltic highway surfaces, two of which were on mountain passes, using a hand-held, commercial battery-powered vacuum cleaner employing a cyclone separator and by sweeping the road surface with a natural fiber bristle brush onto paper. Road-tire matter was drilled from cleaned surfaces of worn tire treads using a 15.875-mm (5/8-inch) spade bit. Shredded tire was collected after the outermost 3–5 mm of tread were drilled and discarded to minimize combining tire matter with mineral and bitumen crusts from asphaltic surfaces.

Observations of samples were made under a Keyence VX-7000 stereomicroscope at 80–700x and a scanning electron microscope (SEM; FEI Quanta 450 FEG) equipped with an energy-dispersive spectrometer (EDS; Oxford Instruments XMax^N 50). Operating conditions for secondary and backscatter-electron imaging and EDS collection were variable depending on the particle but resulted in imaging resolutions of 50 nm as well as detection of carbon and oxygen at the low energy end of the spectrum and first row transition elements at the high-energy end. Samples were prepared for SEM-EDS analyses by placing individually hand-picked particles on SEM stubs after they were imaged using stereomicroscopy.

Reflectance spectra were measured for one aliquot of each snow sample and one shredded tire. Each aliquot was sufficiently thick to fill a sample holder so that measured reflectance represented the total sample. Reflectance spectra were measured with an Analytical Spectral Devices Inc. FieldSpec3 standard resolution spectroradiometer, covering the wavelength range of 0.35–2.50 μm. Reflectance was measured relative to a Labsphere Spectralon® 99% reflectance reference panel. The average reflectance spectrum for each sample was converted to absolute reflectance, adjusting for the absorption properties of the reference panel (Kokaly & Skidmore, 2015). Channels below 0.37 μm were not used because of an increase in reflectance likely caused by stray, longer wavelength light. Total reflectance (Rtot) for each sample was computed by averaging the reflectance values for 2,131 channels across the wavelength range 0.37–2.50 μm. Calculations were made using the Material Identification and Characterization Algorithm, a module of the PRISM software system (Kokaly, 2011). Sources of uncertainty include the measurement protocol and instrumentation (sample loading, illumination, and spectrometer stability). To assess the uncertainty related to the measurement protocol, nine measurements of an internal standard were made across five measurement sessions (August 2014–July 2016). The mean Rtot for the internal standard was 0.5230 with a standard deviation of 0.0083. The largest uncertainty (95% confidence interval) for the Spectralon panel reflectance propagated to a 0.0127 uncertainty in reflectance for the internal standard.

Organic carbon content of bulk dust was measured with an Elementar Soli TOC Cube (Elementar Analysensysteme GmbH, Langenselbold, Germany), under a gas-switching method, with temperature-dependent-oxidation. During the measurement of each dried sample, the temperature of the combustion oven was first ramped to 400°C in an oxygen environment and the resulting CO_2 was measured on the Soli TOC Cube near-infrared detector. This carbon was considered organic carbon (OC_{400}). Next, the carrier gas was switched from oxygen to nitrogen and the oven temperature ramped to 900°C. The resulting CO_2 corresponded to total inorganic carbon (TIC). The carrier gas was then switched back to oxygen with the oven remaining at 900°C and any remaining carbon—the residual oxidizable carbon (ROC)—was converted to CO_2 and measured. The total organic carbon (TOC) content of the sample is the sum of OC_{400} and ROC ($TOC = OC_{400} + ROC$). Organic compounds in six ALM samples were measured by two-dimensional gas chromatography (GCxGC; Liu & Phillips, 1991; Molden, 2023a, 2023b). Techniques for measuring masses of tires shredded during road tests were described by Molden (2023a) and applied to data of driven road distances listed by state, region, and the entire United States published by the Federal Highway Administration (2016, 2018, 2021). Twelve monthly pdf files covered each year tabulated herein. Annual summaries are listed in each report for December of a given year, and the 2012–2021 data are summarized in Federal Highway Administration (2021).

4. Observations and Results

Microplastics were observed in each snow sample covering water years 2013–2021 as cloudy white/translucent, blue, red/pink, green, gray, and black fibers as well as flakes, similar to many of those described previously in road dust (Table 2 in Yang et al. (2023)). The fibers were typically 5–10 μm in diameter, and many were shaped as ribbons twisted or curled along their lengths of tens to hundreds of micrometers suggesting mechanical deformation. The chemical compositions and origins of these particles were indeterminate by shape, size, or color. A black substance coated a few of the cloudy white/translucent fibers or was embedded within clusters of such fibers. Identical and similar, black-coated fibers were found in road-surface and shredded tire samples (Figure 1). A few fibers from road surfaces and tire shreds were also colored in blue and red/pink hues, and some of these colored fibers were partly and thinly coated by a black substance. The fibrous particles with associated black matter in snow were rare and not observed in every sample. Their sparse presence, nevertheless, generated the hypothesis that these particles consisted of tire-derived fibers partly coated by black tire matter. Because of possible contamination, statistical counting of microplastics was not done; only those fibrous particles having intimately associated black matter, possibly TWPs, and those consisting mostly of black matter, possibly RTWPs, were targets of further chemical investigation by EDS. In addition, all samples contained nearly equant black particles (typically <10 μm) that far outnumbered microplastic fibers and flakes and that lacked diagnostic features of their possible origins.

To test the hypothesis that some particles in snow consisted of tire-derived fibers partly coated by black tire matter, the chemical compositions of the fibers and their associated black matter were determined during SEM-EDS analysis (Figures S2–S4 in Supporting Information S1). Only carbon was detected in some cloudy white/translucent fibers that were coated by black matter, consistent with tire rubber. Other similar fibers, composed

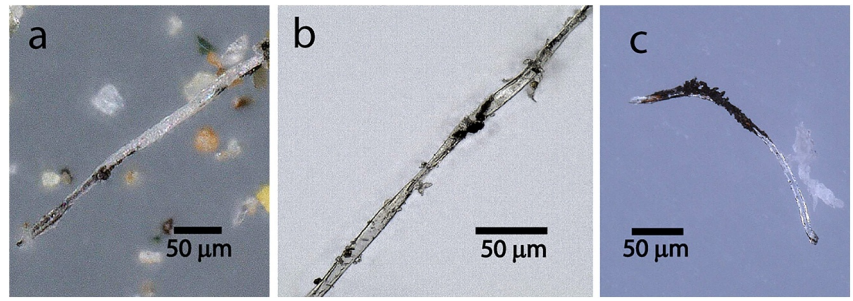

Figure 1. Photomicrographs of microplastic fibers with black substance interpreted as tire matter on their surfaces: (a) Particle from a snow sample from the Berthoud Pass (site Berth in Figure S1 in Supporting Information S1, 2019); (b) Particle from the road surface at McClure Pass (site McClP in Figure S1 in Supporting Information S1); (c) Fiber shred produced by drilling into the tread of an abandoned road-side tire.

dominantly of carbon, contained oxygen, which may have resulted from photo-oxidation of NR and SBR (Mertz et al., 2012). The black matter coatings on fibers contained dominantly carbon with low amounts of oxygen typically along with variable contents of Si, Al, Mg, Ca, S, Ti, Cl, K, and Na. Among the possible sources of carbon in the black matter was carbon black in tire-derived particles and (or) fragments of road asphalt. Oxygen in the black matter may have been contained in common filler oxides (e.g., SiO_2, aluminosilicates, $CaCO_3$, and TiO_2). Overall, much of the compositional variability in the EDS spectra of the black matter might be attributed to various combinations of tire fillers (Roy et al., 2020). We did not attempt to chemically distinguish between particles from tire wear and those from asphalt, as mixtures of the two sources were possible. The lack of detected zinc in black matter coatings, if from tires, was problematic but may have been related to loss of zinc by leaching and (or) to its heterogeneous distribution. Nevertheless, zinc was detected in a road-sample particle, which resembled a RTWP by its characteristic rod shape (Kovochich et al., 2021). Zinc was also common in many black-matter coatings on translucent fibers from freshly shredded tire samples. In some such coatings, however, some analyzed spots yielded EDS spectra indicating the presence of zinc, whereas nearby analyzed spots failed to detect zinc.

The average total reflectance of the six snow samples varied by 23% (range, 0.2881–0.3746; Table 1). The shredded tire had an average total reflectance of 0.0280 (standard deviation, 0.005), and its spectrum was featureless except for displaying sightly increasing reflectance with increasing wavelength (Figure S5 in Supporting Information S1). Total organic carbon varied by 63% (range, 8.35–22.48 wt%; Table 1).

Results of pyrolysis gas chromatography (GCxGC) revealed (a) that each of the six snow samples contained similar numbers of specific organic compounds found in tires (average, 389; standard deviation, 14) and (b) that the number of these organic compounds constituted about half of the number of all organic compounds in each sample (average, 50%; standard deviation, 3%) (Table 1). The four organic compounds listed in Table 1 were

Table 1
Results From Organic Compound, Spectral Reflectance, and Total Organic Carbon Analyses

Site	WY	OC number	OC sum%	Fluorene	1,3-Pentadiene	2,4-Dimethylstyrene	Biphenyl	Rtot	TOC
McClure Pass	14	396	53	64.2	440.0	1.6	11.4	0.3601	8.35
McClure Pass	16	369	49	77.9	269.3	14.6	11.9	0.3378	20.5
Wolf Creek Pass	15	402	47	54.6	0	0	15.2	0.3110	13.58
Wolf Creek Pass	16	377	48	29.9	0	8.5	9.3	0.2881	22.48
Berthoud Pass	16	386	56	0.2	1223.1	29.0	14.1	0.3282	12.44
Grand Mesa	16	405	49	49.3	1.2	1.3	12.0	0.3746	8.52

Note. WY, water year of sample collection. OC Number, numbers of specific organic compounds in a snow sample determined by the GCxGC method that are in the road-tire-composition database (Molden, 2023a, 2023b); OC SUM%, the proportion of the number of organic compounds detected in a snow sample also found in the road-tire-composition database to the number of all organic compounds in that sample; concentrations of the four listed organic compounds in ng mg^{-1}; Rtot, average spectral reflectance across 0.37–2.5 µm wavelengths; TOC, total organic carbon in weight %.

overall the most prevalent in the tire database, potentially toxic, and of low risk of transformation in the environment.

5. Discussion

Results from the three approaches are interpreted together to reveal the presence of road-tire-wear fragments in the UCRB snow samples. Importantly, the GCxGC method differs from the other two approaches in its capacity to sense particles of all sizes, including those much smaller than were individually imaged by stereomicroscopy and analyzed by SEM-EDS. Although the GCxGC method does not differentiate among particle-size classes, such small particles include the nano-size component, which is highly susceptible to airborne emission from roads and subsequent atmospheric transport (Dahl et al., 2006; Dall'Osto et al., 2014; Harrison & Alghamdi, 2023; Panko et al., 2013). The numbers of organic compounds found in tires and their sum percentages relative to all organic compounds varied little despite the large range in total organic carbon. This result indicates that tire matter was supplied as a relatively constant fraction in relation to all organic compounds in other carbonaceous substances, such as soil-plant matter, independent of site location.

The foregoing interpretations of tire matter in UCRB snow led to the following assessment of the amounts of atmospherically transported TWPs and their potential radiative importance over larger areas and selected time spans. Potential RF effects of TWPs can be partly assessed by estimates of (a) their produced masses by location, year, and season, (b) the proportion of their mass and (or) number lifted into the atmosphere, and (c) their airborne transport and depositional fates, along with an assumed radiative efficiency for them (not done here). Different methods have been used to estimate TWP-MP mass (e.g., Brahney et al., 2020, 2021; Evangeliou et al., 2020, 2022; Kole et al., 2017; Wagner et al., 2018), including (a) sampling followed by various counting and (or) instrumental detection methods, (b) modeling, (c) measurements of tire-mass lost over tire lifetimes, and (d) measures of tire wear applied to road-travel distances. Our approach centered on the fourth method using a recent measure of tire-wear mass to estimate the amounts of TWPs thoroughly blackened by graphitic carbon black.

Herein, vehicle road-travel distances for different scenarios of time span and area have been multiplied by a road-measured amount of tire-mass loss per distance traveled. That amount is 0.000067 kg km^{-1} for four new tires on a light-duty vehicle (Molden, 2023a). In one set of examples, travel distances in the U.S. were taken from the Federal Highway Administration (2016, 2018, 2021, as indicated in Section 3). Tire-mass-loss estimates provided in this study greatly underestimate actual losses primarily because they do not account for heavy-duty vehicles, such as pick-ups, SUVs, and trucks, as well as aggressive driving behavior. In addition, this factor discounts wear rates of old tires because new tires shed more debris than do used ones. The contribution of the latter influence is likely small, considering legacy tire-wear MPs in the global plastic cycle discussed in the following.

Because of interest in particulate matter on snow, we estimated tire-wear-particle mass generated within the 11 conterminous states of the western U.S. during November through May of 2016, the period of regional high-elevation snow cover that captures atmospheric dust and provides critical water sources for downstream consumption. With 6.47×10^8 km traveled by vehicles in the 11 states during the 7-month period of 2016, that estimate is 43.3 tonnes of TWPs (6.47×10^8 km $\times$ 0.000067 kg km^{-1} = 43,349 kg).

Another scenario considers total vehicle distance–50.05 trillion km–in the U.S. during the 10-year period of 2012–2021 (Federal Highway Administration, 2021). Such distance would have produced minimally about 3,353.3 kilotonnes (kt) of TWPs. During 2016, total vehicle distances in the U.S. (5.11 trillion km; Federal Highway Administration, 2016) would have yielded about 342 kt of TWPs. During 2018 in the U.S., total vehicle distances of about 5.215 trillion km (Federal Highway Administration, 2018) would have produced 349 kt of TWPs.

Many uncertainties accompany the foregoing estimates. As one matter, measured tire-mass losses range widely as summarized by Lee et al. (2020): 0.5×10^{-4} to 1.32×10^{-4} kg km^{-1} for passenger cars, 0.5×10^{-4} to 7×10^{-4} kg km^{-1} for lightweight trucks, 2.67×10^{-4} to 7×10^{-4} kg km^{-1} for buses, and 5.17×10^{-4} to 11×10^{-4} kg km^{-1} for heavyweight trucks. Values depend on many factors including tire types and age, setting (laboratory compared with road tests), road conditions, and driving situations (e.g., different speeds, as well as straight-line at constant speed, braking, and cornering). Moreover, more light trucks now operate in the U.S. than passenger vehicles (Federal Highway Administration, 2023). In the future, another factor might be an increasing

Table 2
Estimates of Annual Atmospheric Microplastics

Source	Geographic extent	Range or factor	Atmospheric MPs kt/yr
Evangeliou et al. (2020) PM10	Global (deposited)	Mean	284
Evangeliou et al. (2020) PM10	Global (deposited)	Low	102
Evangeliou et al. (2020) PM10	Global (deposited)	High	787
Brahney et al. (2021) 4–250 μm	Global (deposited)	Mean	96
Brahney et al. (2021) 4–250 μm	Global (deposited)	Low	63
Brahney et al. (2021) 4–250 μm	Global (deposited)	High	110
Kole et al. (2017)	Global (emitted)	10%	592
Brahney et al. (2020)	U.S. (deposited) mostly 2018	Total	22
This study	U.S. (emitted) 2018	10%	34.7

Note. Estimated global annual MP masses in kilotonnes (kt) deposited from the atmosphere (Brahney et al., 2020; Evangeliou et al., 2020) and annual MP atmospheric emissions from road-tire wear globally (Kole et al., 2017) and in 2018 for the U.S. (This study). Factor refers to a 10% multiplier of total road-tire-wear-generated particle masses to estimate masses entrained in the atmosphere. All values represent global masses except those for the U.S. as noted. In this study, Molden's (2023a) factors of 0.000067 kg km^{-1} for tire-wear-mass loss and 10% airborne emission were applied to estimates of vehicular distances in the U.S. 2018 entry (Federal Highway Administration, 2018). The estimate for Kole et al.'s atmospheric MPs is based on 10% of their global emissions estimate of 5,918 kt. The estimate by Brahney et al. (2020) for MP deposition across the contiguous U.S. (22 kt) was derived from sample collections made late 2017 to early 2019 at remote sites in western U.S. and thus is comparable in time span to the estimate for "This study; U.S. (emitted) 2018." The amount attributed by Brahney et al. (2020) to tire wear (84% of this 22-kt estimate) is 18.5 kt yr^{-1}. The entry for "This study" includes road travel in Alaska and Hawai'i that together contributed about 0.5% to the total U.S. distances.

number of electric vehicles, each of which produces more TWPs per traveled distance than an internal combustion vehicle because of the greater masses of the former.

Determining the proportion of TWPs entrained by wind into the atmosphere remains a conundrum, involving factors of particle size and shape along with emissivity factors from different conditions of place and time (e.g., Brahney et al., 2021; Evangeliou et al., 2020; Kole et al., 2017). Estimates of this proportion vary widely. In one assessment, Grigoratos and Martini (2014; cited by Kole et al., 2017) broadly approximated this proportion at 1%–30%. Recent measurements of airborne TWPs emitted directly and instantaneously from tire contact on a road surface yielded values of 10% by mass and 90% by number (Molden, 2023a). In Table 1, this measured 10% value is used as the airborne component of estimated tire-wear-particle masses for this study and for those of Kole et al. (2017). The 10% factor for airborne emission of tire matter is considered conservative because it accounts for immediate generation of tire particles when the rubber meets the road and not additional suspension from road surfaces by traffic turbulence (see Chen et al., 2023) and much later emission of legacy MPs discussed in the following.

For the 2018-U.S. case, the estimate for TWP-MPs under 10% airborne generation is 34.9 kt, which is 1.6-times greater than all MP deposition in the U.S. estimated by Brahney et al. (2020) at 22 kt annually (determined mostly from 2018 sampling) for a particle-size range of 4–250 μm (Table 2). By modeling, 84% of MP deposition in the western U.S. was attributed to road-dust sources by Brahney et al. (2021), so that their annual estimate for road-tire-produced, deposited MPs would be about 18.5 kt. For the winter-late spring period in the western U.S. during 2016, roughly 4.3 tonnes of road-tire-wear-MPs would have undergone atmospheric transport under 10% airborne generation.

With respect to sources, an important consideration is the global plastic cycle (Bank & Hansson, 2019), by which MPs can reside temporarily in a variety of settings before entrainment into the atmosphere. A source for such emission is wave action and bubble bursting that produce sea-spray aerosols, which contain MPs originally deposited on land and then transported by runoff into oceans. Estimates for the masses and numbers of MPs liberated from ocean surfaces vary greatly (Allen et al., 2020; Allen, D. et al., 2022; Allen, S. et al., 2022; Evangeliou et al., 2022; Fu et al., 2023; Harb et al., 2023; Shaw et al., 2023; Trianic et al., 2020; Yang et al., 2022). The amounts of such resuspended legacy TWP-MPs from ocean surfaces, nevertheless, are a key factor and major uncertainty for assessing their RF influence by possibly augmenting atmospheric loading of

TWPs directly from roads (Brahney et al., 2021). Among additional sources of atmospheric MPs are emissions from agricultural fields treated with MP-bearing waste or temporarily covered with plastic sheeting, trash incineration, disintegrated macroplastics, and sewage treatment plants (e.g., summarized by Bergmann et al. (2019), Brahney et al. (2021), Mormile et al. (2017)).

The extent to which the RF effects of carbon black-permeated tire fragments resemble those of BC of any form or origin is an important unknown, depending upon differences in their physicochemical properties. Importantly, carbon black for tires is produced under mostly uniform conditions by hydrocarbon combustion. Accordingly, reflectance spectra of two carbon black standards are closely similar with reflectance values <0.03 out of 1.0 for a purely reflective surface (Clark, 1983; Kokaly et al., 2017; Figures S6 and S7 in Supporting Information S1). In contrast, BC is generated under many different conditions (Lack et al., 2014; Petzold et al., 2013). Nevertheless, black tire fragments and BC from traditionally recognized sources (fossil-fuel combustion produced by domestic activities, such as cooking stoves; industrial, vehicle, and ship exhaust emissions; as well as burned vegetation) might possess similar RF effects because both classes are produced by combustion as agglomerates of nano-size particles.

The airborne release of black matter from tires indicates that some amount of black atmospheric LAPs is from road-worn tires thereby augmenting BC atmospheric loading. Importantly, radiative absorption of a worn-tire particle would involve nearly all of the surface area of the particle because graphitic carbon-black homogeneously permeates tire matter. Assuming that an entire carbon-black permeated tire particle absorbs solar energy in a nearly uniform manner, as illustrated in Figure S5 in Supporting Information S1, its contribution to atmospheric and cryosphere RF would be larger than determined solely by its carbon-black content. As such, our estimates of produced road-tire-wear particles amplify the earlier supposition of a BC source from road-tire wear (Klimont et al., 2017).

In the case of Kole et al.'s (2017) global estimate, the annual contribution of TWP-MPs to the atmosphere would have amounted to 592 kt assuming 10% airborne emission and would have added about 8%–9% as black LAPs to total BC emissions (6,600–7,200 kt) during 2000–2010 (Klimont et al., 2017). Assuming 30% airborne emission as suggested possible by Kole et al. (2017), annual atmospheric road-tire-wear-particle masses would have amounted to approximately 1776 kt, which, considered as black LAPs, would have represented 27% of total BC emissions during 2015, even discounting emission of legacy microplastics.

Considering that Kole et al.'s (2017) frequently cited approximation for global emissions of TWPs was derived from different types of estimates covering about 17 years up to 2016, a revision might now be timely. Their estimate of 5,917.52 kt found an average annual emission factor of 0.81 kg per person in the global population. Global population has increased 10.422% from 2016 to January 2024. Applying that increase to the original estimate yields a current revision for global tire-wear generation of approximately 6,550 kt and 655–1,965 kt for a respective 10%–30% spread in an assumed airborne component.

A tire-wear-mass estimate of 6,290 kt generated for a population of nearly 7.765 B in 2019 would have added 629–1,887 kt of black LAPs (under the 10%–30% airborne assumption) to the estimated global BC emissions at 5,800 kt (https://www.ccacoalition.org/short-lived-climate-pollutants/black-carbon; accessed 24 January 2024; O'Rourke et al., 2021) Such a range in mass would thereby represent about 11%–33% of these BC emissions.

An important but poorly understood factor for assessing RF by tire-wear products is the size range, shapes, and consequent aerodynamics of particulates and related surface area, in addition to amounts (Kole et al., 2017; Xiao et al., 2023). In particular, nano-size particles are a recognized but understudied component of tire-wear MPs (Allen, D. et al., 2022; Allen S. et al., 2022; Beji et al., 2020, 2021; Dahl et al., 2006; Dall'Osto et al., 2014; Materić et al., 2020, 2021; Mathissen et al., 2011; Schwaferts et al., 2019; Thorpe & Harrison, 2008; Wagner et al., 2018).

6. Implications for Radiative Effects on Snow Cover in the Colorado Rocky Mountains

The four selected organic compounds listed in Table 1 are too few for their amounts to estimate masses of tire matter and thereby to quantify possible influence of black tire matter on spectral reflectance (Rtot in Table 1). Nevertheless, the GCxGC data might serve as a proxy for tire matter. To simulate a possible tire-matter proxy, the organic compound concentrations in each of the four samples, which contained all four target organic compounds, were summed. The highest concentration-sum sample had lowest Rtot, the lowest concentration-sum sample had

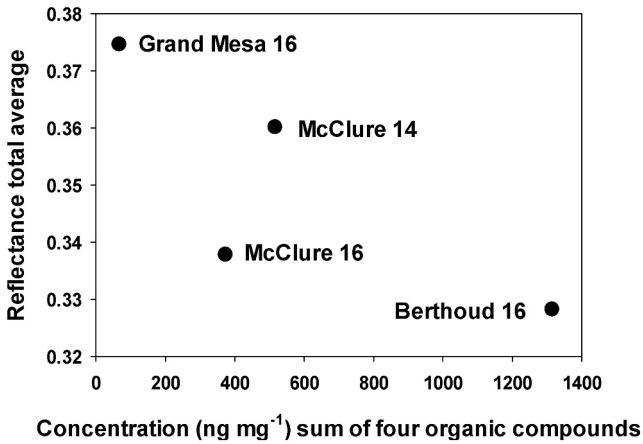

Figure 2. Plot of average total reflectance versus the sum of four organic carbon compounds (Table 1) detected in four snow samples designated by sites and water years.

highest Rtot, and the data for other two samples plotted in between (Figure 2). Such a relation would be expected if the concentration sum of the four targets proxied black tire matter. It is important to emphasize that the summed concentrations are considered as underestimates for the following reasons: (a) Not all of the organic compounds in a tire were sought in the analyses, and (b) some of the target chemicals might have transformed in the environment, so that only a proportion of them were detected in a sample. In addition, the concentrations of the identified organic compounds were not all quantified using standards for those exact compounds.

The high numbers of tire-related organic compounds and their proportions to non-tire organic compounds (about 50%) indicate that tire matter was present in the snow samples and perhaps in sufficient amounts to contribute to RF of the snow surfaces. Moreover, the number of tire-related organic compounds was nearly constant suggesting nearly uniform atmospheric inputs of tire-matter types across the study region during 2014–2016. Altogether, the results imply that road- and tire-derived particulates in snow can contribute to diminished snow-surface albedo, thereby advancing the onset and rate of snowmelt in the Colorado Rocky Mountains. Several other kinds of light absorbing, carbonaceous particulates measured by TOC (Table 1), including vegetal matter, insect parts, soot from burned wood, coal, biochar, and industrial/transport-exhaust effluents, likely also diminished Rtot.

7. Broad Implications

The RF effects of TWPs depend on source locations, transport pathways, and distances, including residence times and distributions of temporarily sequestered particles in soil and oceans. Additional research is needed to understand RF effects of road-tire-wear particles and their relations to those of BC. Along with better characterization of black-tire-particle optical properties, more tests to recognize sub-micrometer fragments of tire matter in the atmosphere and on cryospheric surfaces would be useful (see Panko et al., 2013). Furthermore, model outputs for atmospheric transport of particulates imply regionally dense concentrations of MPs (Brahney et al., 2021; Shaw et al., 2023) and BC (Klimont et al., 2017) on marine surfaces. The airborne emission of plastics by ocean-spray processes is another source to consider when evaluating atmospheric loading and subsequent deposition of micro- and nano-plastic particles generated directly from roads.

Given that some MP particles can undergo far-distant atmospheric transport, including to high elevations of snow-covered mountains, polar latitudes, and other remote locations (e.g., Allen et al., 2019, 2021; Bergmann et al., 2019; Brahney et al., 2020; Evangeliou et al., 2020; Materić et al., 2021; Revell et al., 2021; Wetherbee et al., 2019; Xiao et al., 2023; Zhang et al., 2019), the possibility for the long-range atmospheric transport of abraded black TWPs raises the question: Do fibrous and non-fibrous TWPs travel similar distances given possibly different aerodynamic behavior according to sizes, shapes, and perhaps relative densities? The amounts of these atmospheric MP particulates, nonetheless, will likely increase with increased vehicular use while attempts continue to lower emissions from widely recognized BC sources, such as residential combustion and industry-transportation exhaust. Such counteracting influences represent challenges to mitigate the potential RF effects produced by road-tire-wear substances. In addition, more information about the toxicity of TWPs would address increasing concern about their health effects on organisms (Allen, S. et al., 2022; Boisseaux et al., 2024; Kole et al., 2017; Leslie et al., 2022; Li et al., 2023; Materić et al., 2021; Prata et al., 2020; Rogge et al., 1993; Shen et al., 2019; Sherman et al., 2024; Smith et al., 2018; Vethaak & Legler, 2021; Wik & Dave, 2009; Wright & Kelly, 2017). In this respect, the presence of toxic organic compounds associated with road-tires underscores the importance of assessing the potential toxicity of far-traveled road-tire-wear particles on terrestrial biota as documented for those particles transported into water bodies in local runoff (Boisseaux et al., 2024; Greer et al., 2023; Tian et al., 2021).

8. Summary and Conclusions

The presence of a black substance that coated a few translucent microplastic fibers collected from snow in the Colorado Rocky Mountains led to the suspicion that tire-derived fibers were coated by black-tire matter. This

possibility was strengthened by the presence of identical and similar, black-coated particles in road-surface and shredded tire samples. Road tires are black because of the presence of carbon black, a graphitic tire additive that homogeneously permeates tire polymers and other additives and that may thus be very similar in its spectral properties to forms of black carbon, such as soot. The elemental compositions of the black-matter coatings, determined by SEM-EDS, were consistent with compositions of road tires and asphaltic roads. Results from two-dimensional gas chromatography provided evidence that road-tire matter was present in six snow samples by revealing that (a) the snow samples contained many specific organic compounds found in tires and (b) the number of these tire-related organic compounds constituted about half of all organic compounds in each sample.

The interpreted presence of black-tire matter in Colorado Rocky Mountain snow led to an assessment of the amounts of atmospherically transported black-tire-wear particles across areas beyond the Colorado Rocky Mountains. The distribution of masses of particles produced by road-tire wear was estimated by multiplying measured amounts of eroded tire-per-distance traveled by vehicular distances over different scenarios of time and space. Radiative forcing by carbon black-infused particulates produced by annual road-tire wear may augment the RF effects of atmospheric black carbon by about 10%–30%. As such, the spectral behavior (absorption, extinction, and scattering efficiency) of micro- and nano-size tire fragments, thoroughly blackened by graphitic, light absorbing carbon black, might be considered a relevant component for modeling the spectral albedo of snow.

Conflict of Interest

Authors declare no real or perceived financial conflicts of interests or other affiliations that may be construed as having a conflict of interest with respect to the results of this paper.

Data Availability Statement

Data, observations, and metadata supporting the conclusions can be obtained in Supporting Information S1 and in the ScienceBase database, https://doi.org/10.5066/P13HIOO8 (Goldstein et al., 2024; published 22 July 2024). Metadata include identification information, data quality information, spatial reference information, entity and attribute information, distribution information, and metadata reference information. For location information, the Geodetic Model used the Horizontal Datum WGS84 and Ellipsoid GRS_1980.

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