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Contribution of traffic-originated nanoparticle emissions to regional and local aerosol levels

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1 Trimodal fit equation for emission factor particle size distribution

Emission factor particle size distribution (EFPSD) was fitted to a trimodal distribution with the equation

$$\frac{\mathrm{d}n}{\mathrm{d}\log D_\mathrm{p}} = \left.\frac{\mathrm{d}n}{\mathrm{d}\log D_\mathrm{p}}\right|_\mathrm{power\,law} + \left.\frac{\mathrm{d}n}{\mathrm{d}\log D_\mathrm{p}}\right|_\mathrm{nucleation} + \left.\frac{\mathrm{d}n}{\mathrm{d}\log D_\mathrm{p}}\right|_\mathrm{soot} \tag{S1}$$

where n denotes the emission factor of the particle number belonging to a mode. The first term is expressed with the equation (Olin et al., 2016)

$$\left.\frac{\mathrm{d}n}{\mathrm{d}\log D_\mathrm{p}}\right|_\mathrm{power\,law} = \begin{cases} n\left(\frac{D_\mathrm{p}}{D_2}\right)^\alpha \beta, & D_1 \le D_\mathrm{p} \le D_2 \\ 0, & \text{otherwise} \end{cases} \tag{S2}$$

where β is a scaling function

$$\beta\left(\alpha, \frac{D_1}{D_2}\right) = \begin{cases} \dfrac{\alpha \ln 10}{1 - \left(\frac{D_1}{D_2}\right)^\alpha}, & \alpha \neq 0 \\ \dfrac{-\ln 10}{\ln\left(\frac{D_1}{D_2}\right)}, & \alpha = 0 \end{cases} \tag{S3}$$

and α, D_1, and D_2 are the slope parameter and the diameters of the smallest and largest particles of the mode, respectively. The latter two terms are expressed with the equation

$$\left.\frac{\mathrm{d}n}{\mathrm{d}\log D_\mathrm{p}}\right|_\mathrm{nucleation/soot} = \frac{n \ln 10}{\sqrt{2\pi}\ln \mathrm{GSD}} \exp\left[-\frac{\ln^2(D_\mathrm{p}/\mathrm{CMD})}{2\ln^2 \mathrm{GSD}}\right] \tag{S4}$$

where CMD and GSD are the count median diameter and geometric standard deviation of the mode, respectively. The numerical values of the parameters from the fitting are shown in Table 1.

2 Description of the CFD-simulations used to determine chemical composition of the emitted particles

Chemical composition of particles emitted by road traffic was determined using the results from the CFD-simulations by Olin (2013). They consist of a situation where a Euro III bus is driving at a speed of 40 km/h with the engine power of 40 % of the maximum. The situation has been extracted from chasing experiments performed by Rönkkö et al. (2006).

As a Euro III-vehicle, the bus did not have a DPF and it used a diesel fuel having the sulfur content of 50 ppm, which has been the upper limit of the automotive fuel in the EU between years 2004 and 2009.

The simulations were performed using a commercial Ansys Fluent CFD-software with user-defined functions for aerosol dynamics modelling. The CFD-software simulates the flow field (momentum, velocities in 3 dimensions, temperature, and transport of gaseous species) of the bus driving situation including the exhaust flow from the tailpipe. The aerosol model simulates nucleation, condensation, coagulation, deposition, and diffusion of particles emitted directly from the tailpipe and of particles formed from the emitted gaseous precursors after releasing from the tailpipe. The CFD-software also handles the transport of aerosol and connects the aerosol model with temperature and with the concentrations of gaseous species.

Particles were assumed to consist of sulfuric acid (H_2SO_4), water (H_2O), tetracosane ($C_{24}H_{50}$), and soot. Nucleation was modelled as binary H_2SO_4-H_2O nucleation and the nucleation rate was obtained from classical nucleation theory with a correction factor. Condensation modelling included condensation of emitted gases (H_2SO_4, H_2O, and $C_{24}H_{50}$) on emitted soot particles or on particles formed via nucleation. The concentrations of H_2SO_4 and H_2O in the raw exhaust were estimated from fuel and oil sulfur contents and engine parameters. Soot mode properties were obtained from the data measured 10 m behind the bus by chasing. The correction factor for nucleation rate was obtained inversely using measured nucleation mode concentration, i.e., by matching the simulated concentration with the measured one. Particle sizes, instead, were matched with the measured ones using a correction factor for the $C_{24}H_{50}$ concentration, representing the fraction of hydrocarbons which are able to condense on the particles in question.

According to the simulations, particles reached their final sizes (nucleation mode CMD$\sim$10 nm, soot mode CMD$\sim$60 nm) about 5 m behind the bus, but their compositions much earlier. The nucleation mode composition 10 m behind the bus was 11.4 % for H_2SO_4, 25.1 % for H_2O, and 63.5 % for $C_{24}H_{50}$. The soot mode composition was 6.2 % for H_2SO_4, 2.9 % for H_2O, 24.1 % for $C_{24}H_{50}$, and 66.8 % for soot. These (without H_2O) were used as the mass fractions of sulfate (SO_4), primary organic aerosol (POA), and black carbon (BC).

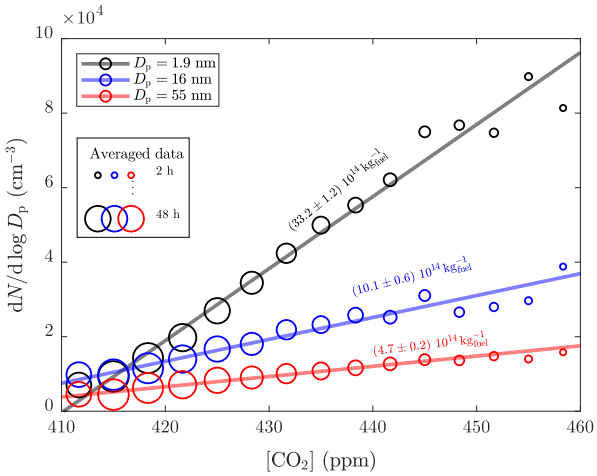

Figure S1. Examples of determining emission factors bin-by-bin for three different particle size bins, similarly to the method by Olin et al. (2020). Size-binned particle number concentration data ($\mathrm{d}N/\mathrm{d}\log D_\mathrm{p}$) are averaged within CO_2 concentration ($[CO_2]$) bins (circle diameters represent the amount of data used in the averaging). Linear fitting (using the circle diameters as weighting factor) is performed over the averaged data (separately for all 28 measured size bins). The slopes of the linear fits converted to kilograms of fuel combusted are marked in the figure.

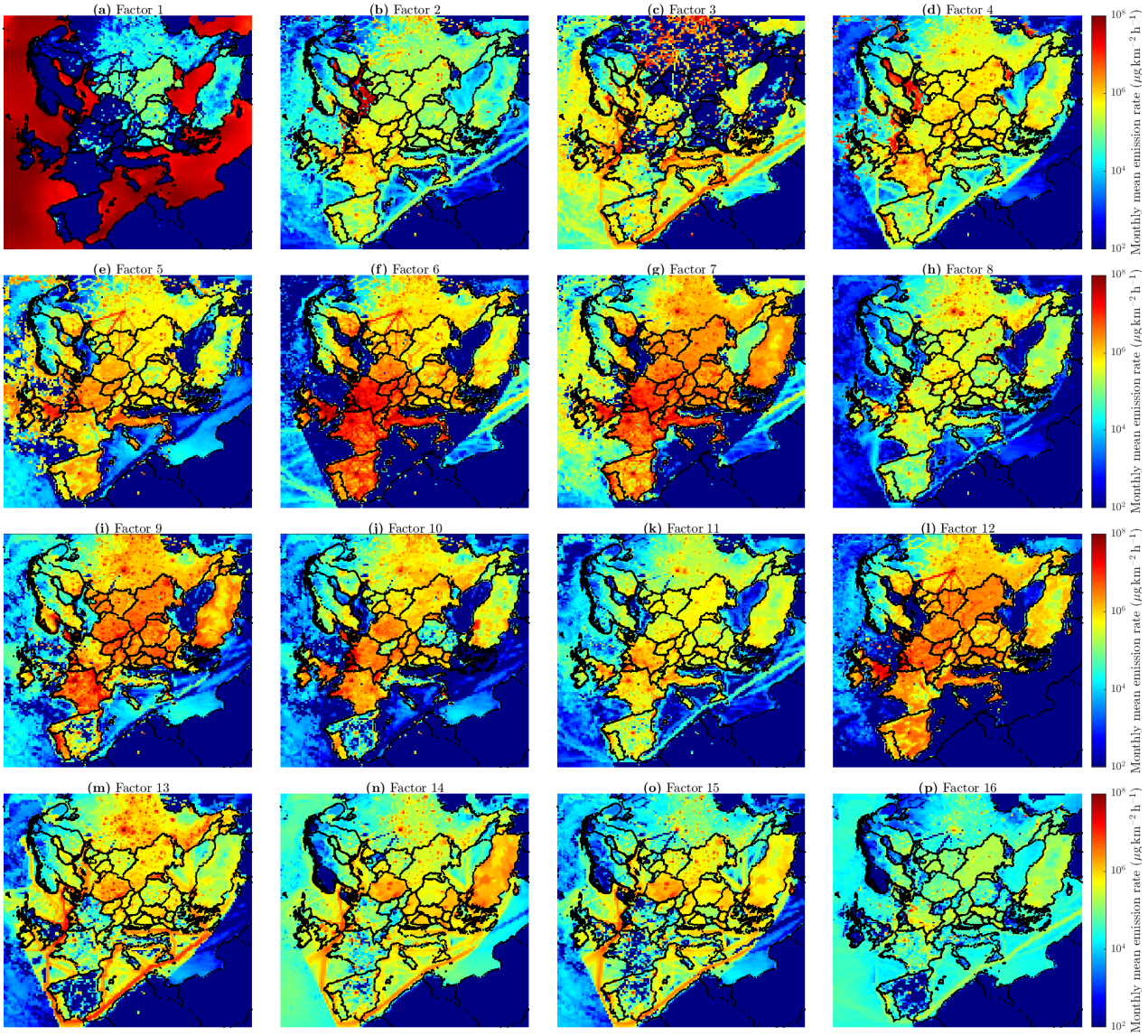

Figure S2. Monthly means of the particle mass emission rates of all PMF factors (**a**–**p**).

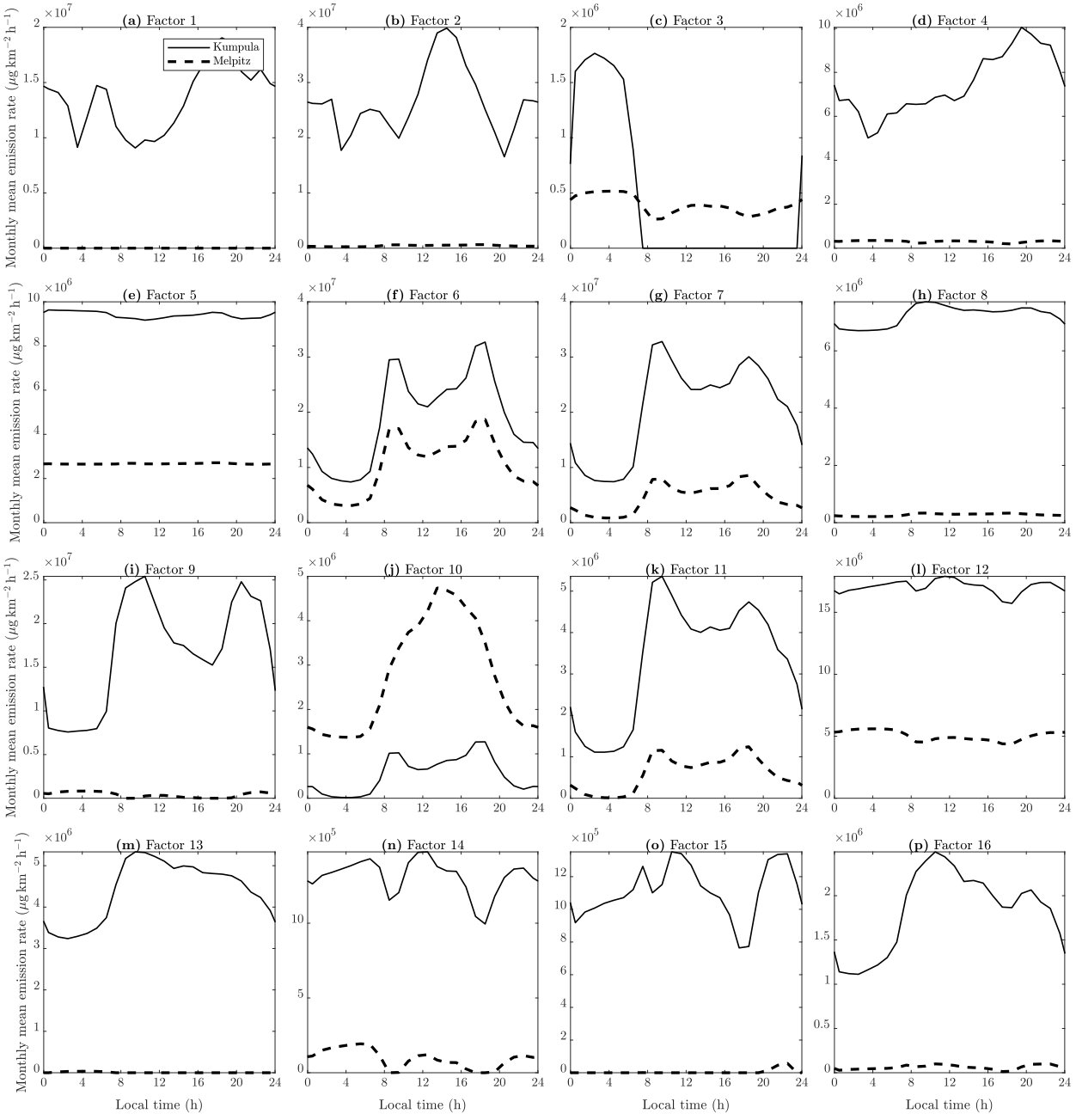

Figure S3. Monthly mean diurnal variations of the particle mass emission rates in Kumpula/Mäkelänkatu, Finland, and Melpitz, Germany, of all PMF factors (**a**–**p**).

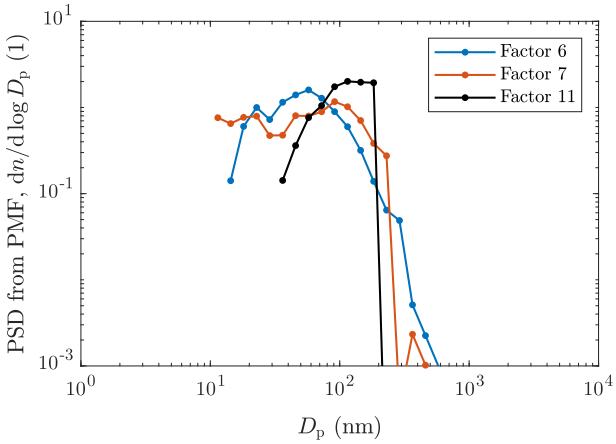

Figure S4. Particle size distributions obtained from PMF factors 6, 7, and 11.

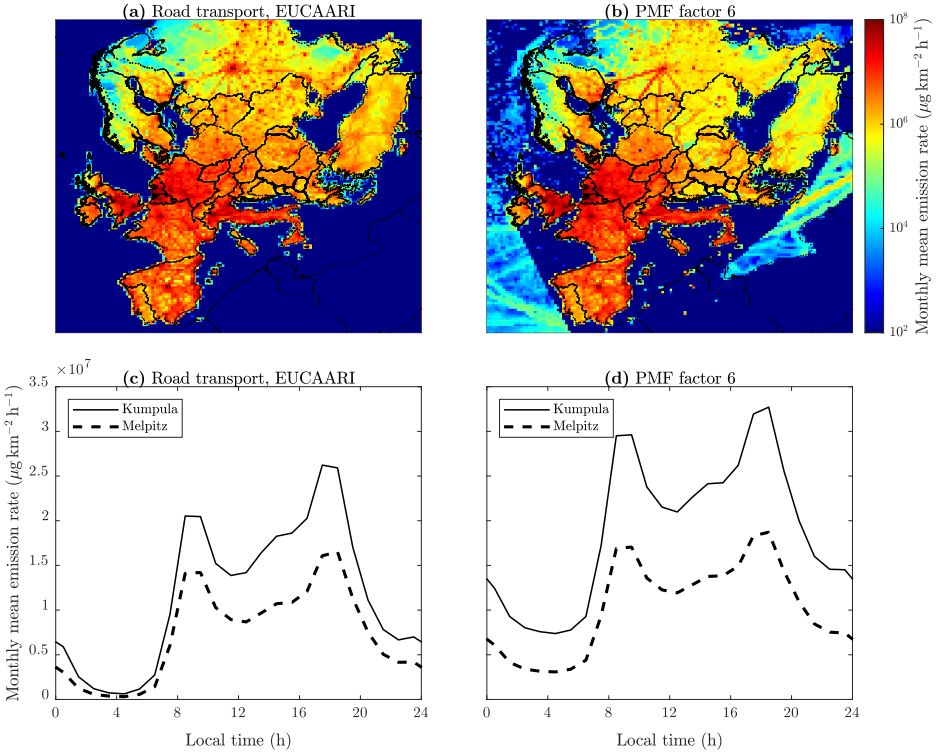

Figure S5. Monthly means of the particle mass emission rates (**a**,**c**) from the road transport-related source in the original EUCAARI inventory and (**b**,**d**) from PMF factor 6 (**a**,**b**) as maps and (**c**,**d**) as diurnal variations in Kumpula/Mäkelänkatu, Finland, and Melpitz, Germany.

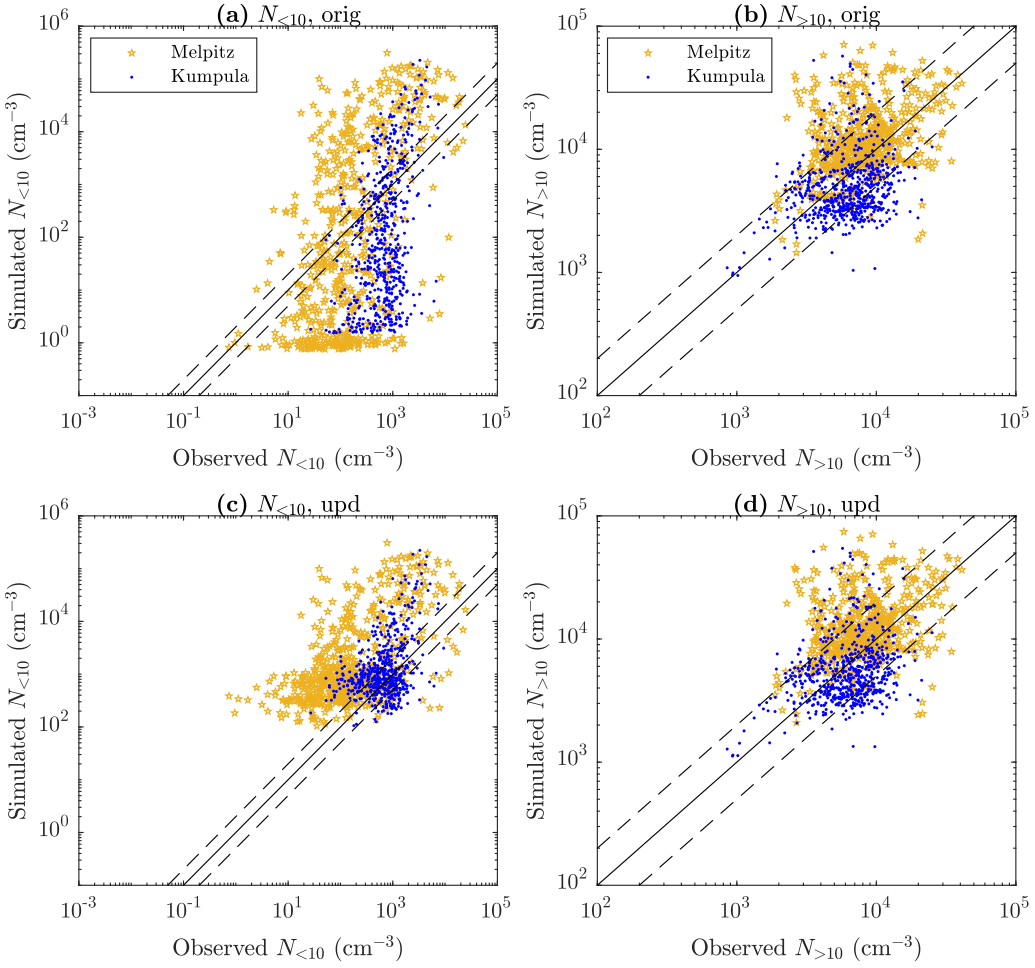

Figure S6. Simulated versus observed number concentrations of particles (**a**, **c**) smaller than 10 nm ($N_{<10}$) and (**b**, **d**) larger than 10 nm ($N_{>10}$) at the stations with the highest traffic influences (Melpitz and Kumpula) with (**a**, **b**) the original and (**c**, **d**) updated emission inventory. All data correspond to hourly means for May 2008. The solid diagonal lines represent 1:1 lines and the dashed ones 1:2 and 2:1 lines.

Table S1. Statistics for grid cell-separated ratios of monthly means of concentrations on the surface-level simulated with the updated and with the original emission inventory. The bold values highlight the particle size ranges experiencing the greatest effects due to updating the inventory.

	$\frac{N_{\mathrm{NCA}}^{\mathrm{upd}}}{N_{\mathrm{NCA}}^{\mathrm{orig}}}$	$\frac{N_{<10}^{\mathrm{upd}}}{N_{<10}^{\mathrm{orig}}}$	$\frac{N_{7-20}^{\mathrm{upd}}}{N_{7-20}^{\mathrm{orig}}}$	$\frac{N_{<23}^{\mathrm{upd}}}{N_{<23}^{\mathrm{orig}}}$	$\frac{N_{<100}^{\mathrm{upd}}}{N_{<100}^{\mathrm{orig}}}$	$\frac{N_{\mathrm{tot}}^{\mathrm{upd}}}{N_{\mathrm{tot}}^{\mathrm{orig}}}$
Mean	**1.113**	1.060	1.019	1.029	1.011	1.009
Population-weighted mean	1.100	1.053	**1.096**	1.034	1.023	1.022
Median	0.9996	1.001	**1.007**	1.0004	1.001	1.002
Population-weighted median	1.004	1.004	**1.043**	1.006	1.006	1.006

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