Online Supporting Information (SI)

Reduced Ultrafine Particle Concentration in Urban Air: Changes in Nucleation and Anthropogenic Emissions

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Table S1: Instrumentation details for 2016-17 (CACES) measurements

Measurement	Instrument	Data time resolution
Particle size distribution, 10-300 nm	TSI 3081 DMA/TSI 3772 CPC	3.3 min per scan
$PM_{2.5}$ mass	TEOM	Hourly average
Ozone	Teledyne O_3 analyzer, Model T400	1 min
SO_2	Teledyne SO_2 analyzer, Model T100	1 min
NO, NO_2	NO_2/NO/NOx Monitor, Model 405 nm, 2B Technologies	1 min
CO	Teledyne CO analyzer, Model T300U	1 min
Black carbon	MAAP, Model 5012, Thermo Scientific	1 min
Temperature, Relative Humidity, Wind speed	Met station, Pittsburgh Allegheny County Airport PA	Hourly average

Table S2: Summary of month-wise nucleation frequency observed during 2001-02

Season	Number of nucleation event days					Nucleation frequency (% day)					
						This study analysis			Stainer et al. (2004b) analysis		
	Sampling Month	Operational days	Regional nucl. days	Short-lived nucl. days	Total nucl. days	Regional nucl.	Short-lived nucl.	Total nucl.	Regional nucl.	Short-lived nucl	Total nucl.
Winter	Dec/2001	28	1	4	5	4	14	18	5	14	19
	Jan/2002	31	9	2	11	29	6	35	31	3	33
	Feb/2002	25	5	10	15	20	40	60	16	45	61
Spring	Mar/2002	26	8	5	13	30	19	50	34	21	55
	Apr/2002	27	13	6	19	48	22	70	49	27	76
	May/2002	24	11	2	13	46	8	54	39	11	50
Summer	June/2002	21	11	3	14	52	14	66	50	14	64
	July/2001	31	9	7	16	29	23	52	29	23	52
	Aug/2001	26	3	8	11	12	31	43	9	35	45
Fall	Sept/2001	28	5	10	15	18	36	54	12	48	60
	Oct/2001	31	15	4	19	41	13	54	51	11	62
	Nov/2001	29	10	4	14	34	14	48	38	16	54
Overall		327	100	65	165	30	20	50	31	22	53
Overall (excluding April and May)		276	76	57	133	28	21	48			

Table S3: Summary of month-wise nucleation frequency observed during 2016-17

Season	Number of nucleation event days					Nucleation frequency (% day)		
	Sampling Month	Operational days	Regional nucl. days	Short-lived nucl. days	Total nucl. days	Regional nucl.	Short-lived nucl.	Total nucl.
Winter	Dec/2016	30	0	6	6	0	20	20
	Jan/2017	29	0	4	4	0	14	14
	Feb/2017	27	1	3	4	4	11	15
Spring	Mar/2017	22	6	3	9	27	14	41
	Apr/2017	2	1	0	1	50	0	50
	May/2017	7	0	2	3	0	29	29
Summer	June/2017	15	1	5	6	7	33	40
	July/2017	24	2	6	8	8	25	33
	Aug/2017	31	2	6	8	6	19	25
Fall	Sept/2016	24	8	7	15	33	29	62
	Oct/2016	30	4	2	6	13	7	20
	Nov/2016	29	2	1	3	7	3	10
Overall		270	27	45	73	10	17	27
Overall (excluding April and May)		261	26	43	69	10	16	26

Table S4: Summary of durations of nucleation events during 2001-02 and 2016-17

Seasons	Event duration (hours)[a]	
	2001-02	2016-17
Winter	5.8±1.5	5.9± 2.3
Spring	5.9±2.1	7.4±2.6
Summer	6.8±1.9	4.8±1.2
Fall	5.3±2.0	7.2±2.5
Overall	6.0±1.9	6.4±2.4

[a]The reported values are mean ± standard deviation

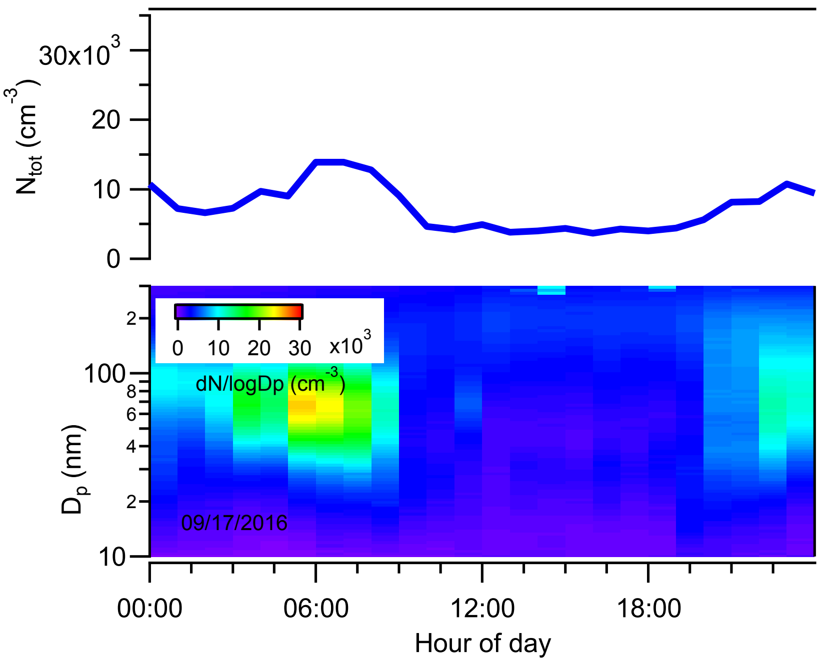

Figure S1: An example of particle size distributions (bottom) and integrated particle number concentrations (top) time series on a typical non-nucleation day.

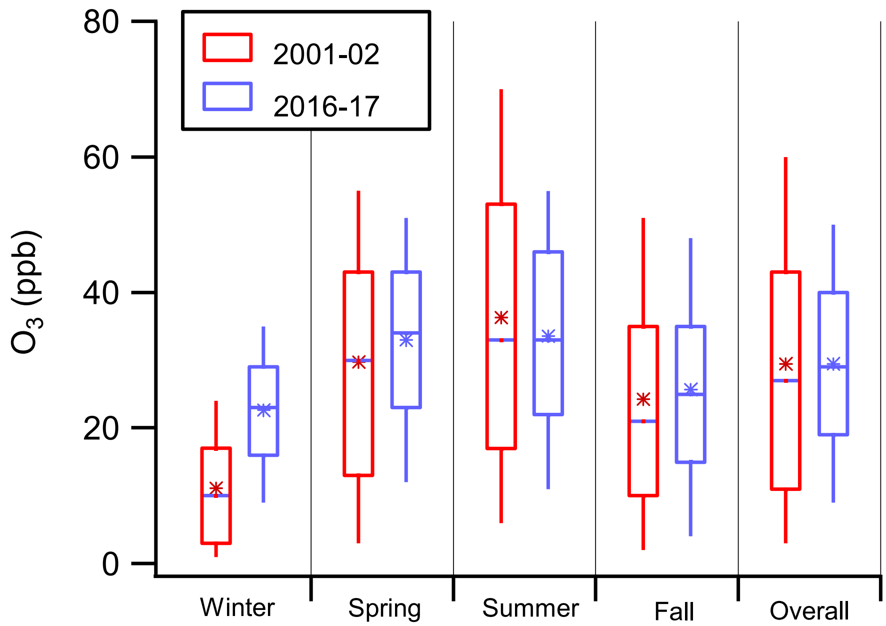

Figure S2: The observed seasonal changes in ambient O3 concentrations between 2001-02 and 2016-17. The box shows data interquartile range, the line inside the box is the median, the length of the whiskers covers 95% of the data, and the star represents the mean.

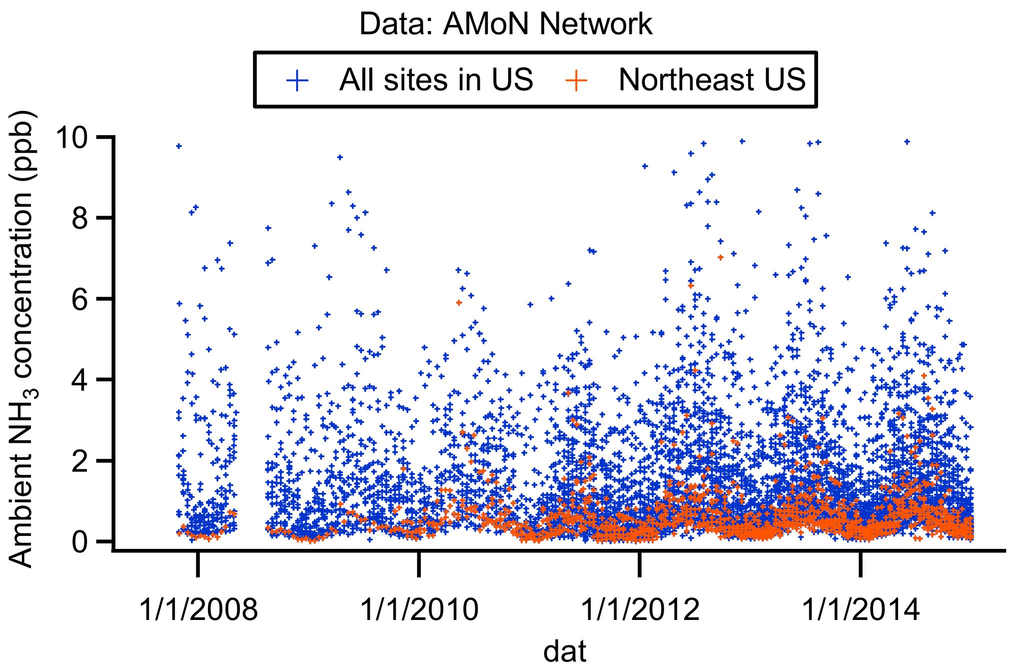

Figure S3: Long term trend in ambient NH_3 concentration in the US. Data are shown from AMoN Network (http://nadp.sws.uiuc.edu/data/AMoN)

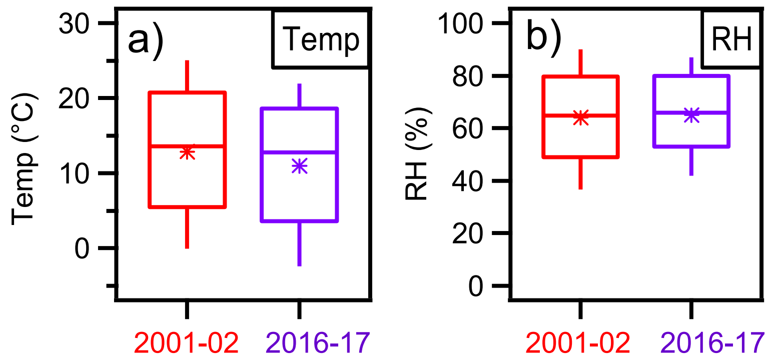

Figure S4: (a) Temperature and (b) relative humidity (RH) in 2001-02 and 2016-17. The box shows data interquartile range, the line inside the box is the median, the length of the whiskers covers 95% of the data, and the star represents the mean.

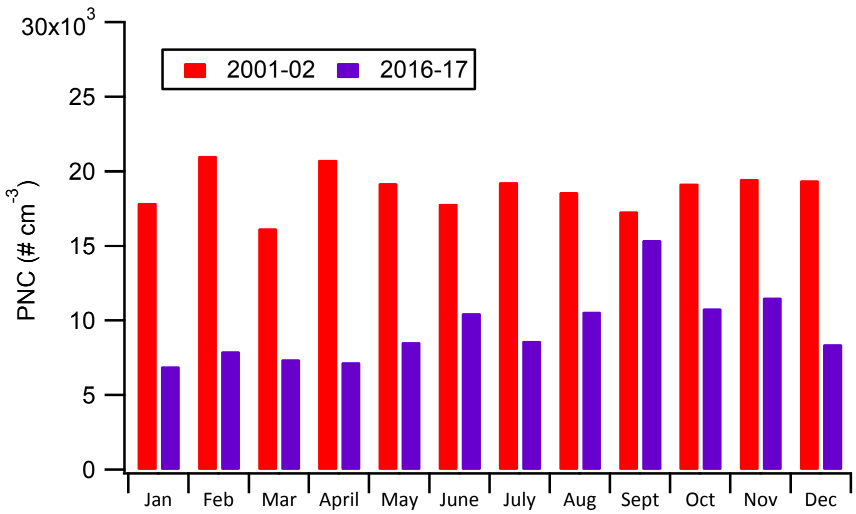

Figure S5: Monthly average particle number concentrations (PNC) measured in 2001-02 and 2016-17. Here the PNC is the integrated particle number concentrations over 10-300 nm. We used it as a metric for characterizing ambient UFP concentrations.

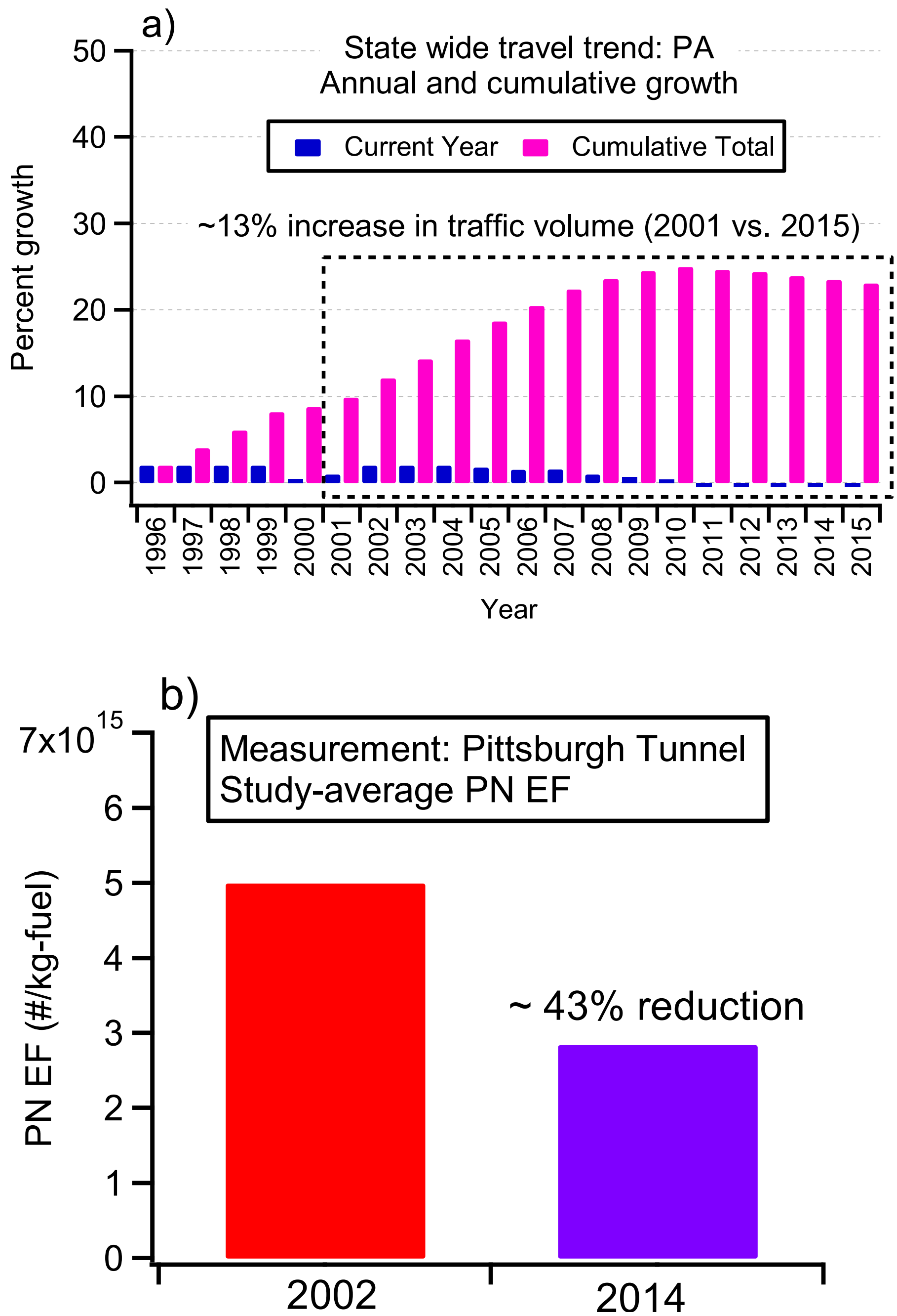

Figure S6: Changes in traffic activity and traffic PN emissions over the last few years. a) Statewide traffic trends in Pennsylvania (PA). Figure shows yearly and cumulative changes in traffic volume from 1996 to 2015. Data are adopted from: 2015 Pennsylvania Traffic Data (http://www.penndot.gov/2015 Traffic Information Report). b) Study-average PN emission factors measured in traffic tunnel in Pittsburgh in 2002 and 2014.

Estimation of condensation Sink (CS)

Condensation Sink (CS) are estimated from measured size distributions (10-300 nm).

$$CS = 2\pi D \sum F(d_{p,i}) d_{p,i} N_i \qquad \text{(S-1)}$$

Where, D is the diffusion coefficient, F is the Fuchs and Sutugin transitional correction factor [1], N_i is the number concentration of particles in size bin of $d_{p,i.}$
A value of D of 7e-6 m^2 s^{-1} was assumed [2]. The F was estimated using an accommodation coefficient (α) of 1.

Estimation of formation rate (FR)
We determine the formation rate (FR) using an empirical approach similar to Hamed et al.[3]. In this approach, we defined the FR as of the rate of change of the particle number concentrations between the nucleation event start time and event peak time.

$$FR = \frac{N_{tot,event\ peak} - N_{tot,event\ start}}{t_{event\ peak} - t_{event\ start}} \qquad \text{(S-2)}$$

Where, $N_{tot,\ event\ start}$ is the total particle number concentration (10-300 nm) during event start time ($t_{event\ start}$). $N_{tot,\ event\ peak}$ is the total particle number concentration when the particle number concentrations reached a peak during the event ($t_{event\ peak}$).

References:

(1) Fuchs, N. A.; Sutugin, A. G. *Highly Dispersed Aerosols.*; Ann Arbor Science, Ann Arbor, MI., 1970.
(2) Saleh, R.; Khlystov, A.; Shihadeh, A. Determination of Evaporation Coefficients of Ambient and Laboratory-Generated Semivolatile Organic Aerosols from Phase Equilibration Kinetics in a Thermodenuder. *Aerosol Sci. Technol.* **2012**, *46* (1), 22–30.
(3) Hamed, A.; Joutsensaari, J.; Mikkonen, S.; Sogacheva, L.; Dal Maso, M.; Kulmala, M.; Cavalli, F.; Fuzzi, S.; Facchini, M. C.; Decesari, S.; et al. Nucleation and Growth of New Particles in Po Valley, Italy. *Atmos Chem Phys* **2007**, *7* (2), 355–376.