Human Exposure to Ultrafine Particles: Sources, Concentrations, Indoor-Outdoor Relationships, and Mitigation Techniques

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Lawrence Berkeley National Lab

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Importance of Ultrafines

- Outnumber all other particles
- High surface/volume ratio: more bioavailable
- Can penetrate human cells and may have a direct route to the brain (olfactory pathway)
- Toxicity > $PM_{2.5}$ on a mass basis
- A few studies show human health effects
  - Oxidative Stress–Induced DNA Damage (Bräuner et al., 2007)
  - Cardio-Respiratory Mortality (Stölzel et al., 2007)

UFP in Human Cell

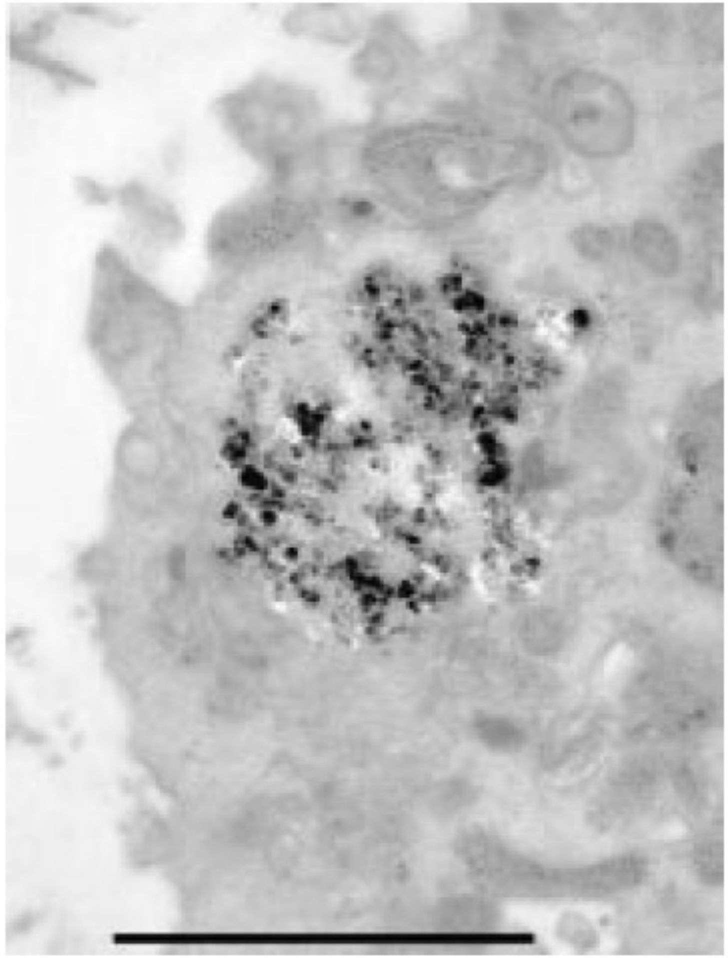


Figure 1 Elecronmicrograph showing carbonaceous ultrafine particles within a phagosome of an alveolar macrophage from a child aged 3 months. Bar = 1000 nm.

Outline of Talk

- Indoor Exposures (outdoor & indoor sources)
- Infiltration (UFP vs FP)
- Personal Exposure (e.g. driving)
- Mitigation (Filters and Exhaust Fans)

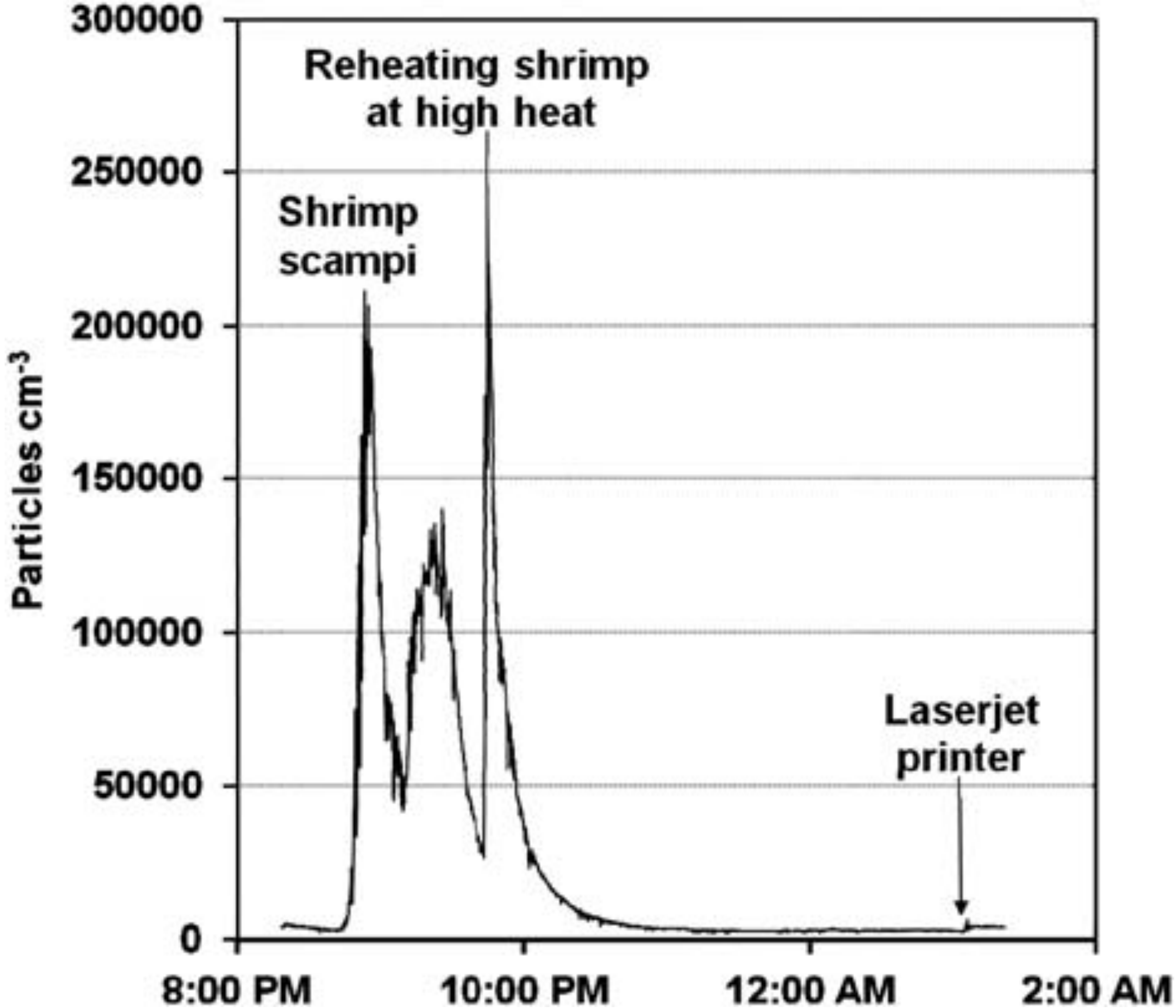
Three Groups of Studies

- Single Occupied Home (1996-2001)
- Test House at National Institute of Standards and Technology (2006-2011)
- Multihome Field Studies by Health Canada (2006-2011)

Single Occupied Home

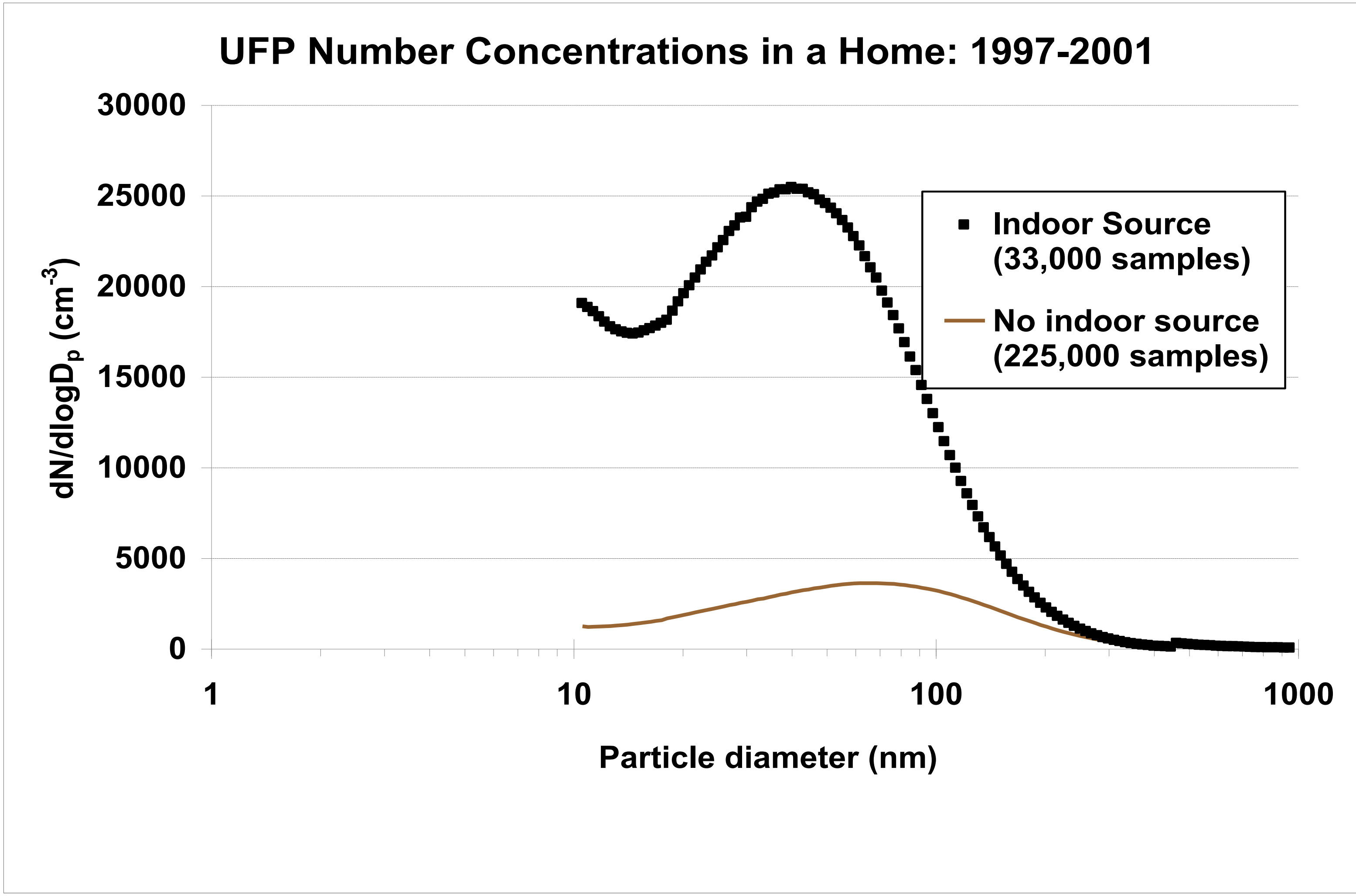
- 3-Story Town House (volume 380 m^3)
  - Gas stove, vented gas clothes dryer
  - Central forced air (fan almost always on)
- Methods
  - Scanning Mobility Particle Sizer (SMPS) with range 10-1000 nm
  - Automated SF6 injection with dedicated GC-ECD for air exchange rate measurements
  - Equipment in basement

Source: Wallace, L.A. and Howard-Reed, C.H. Continuous Monitoring of Ultrafine, Fine, and Coarse Particles in a Residence for 18 Months in 1999-2000. *J Air Waste Manage. Assoc*. **52**(7):828-844. 2002.



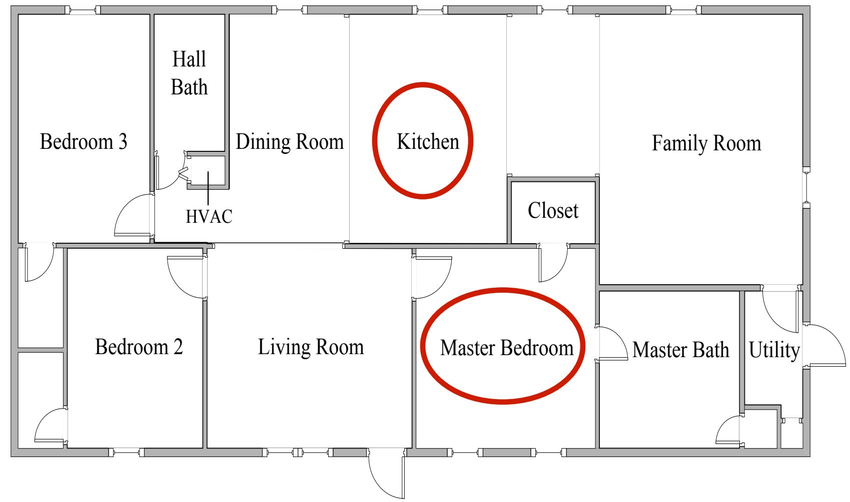
300000
250000
200000
150000
100000
50000
0
Particles cm-3
Reheating shrimp at high heat
Shrimp scampi
Laserjet printer
8:00 PM
10:00 PM
12:00 AM
2:00 AM

Single Home Study



(Source: Wallace et al., 2002)

Experiments in NIST manufactured test house



UFP monitoring with SMPS (2 nm - 100 nm)

- Electrostatic Classifier (TSI Model 3080)
- Nano-Differential Mobility Analyzer (TSI Model 3085)
- Water-based Condensation Particle Counter (CPC) (TSI Model 3786)
- Vacuum Pump and a critical orifice: aerosol flow rate from 0.6-1.5 Lpm
- A stronger neutralizer (TSI Model 3077A Krypton-85)

Focus of The NIST Research Effort

Most previous work on indoor sources has investigated UFPs from 10-100 nm

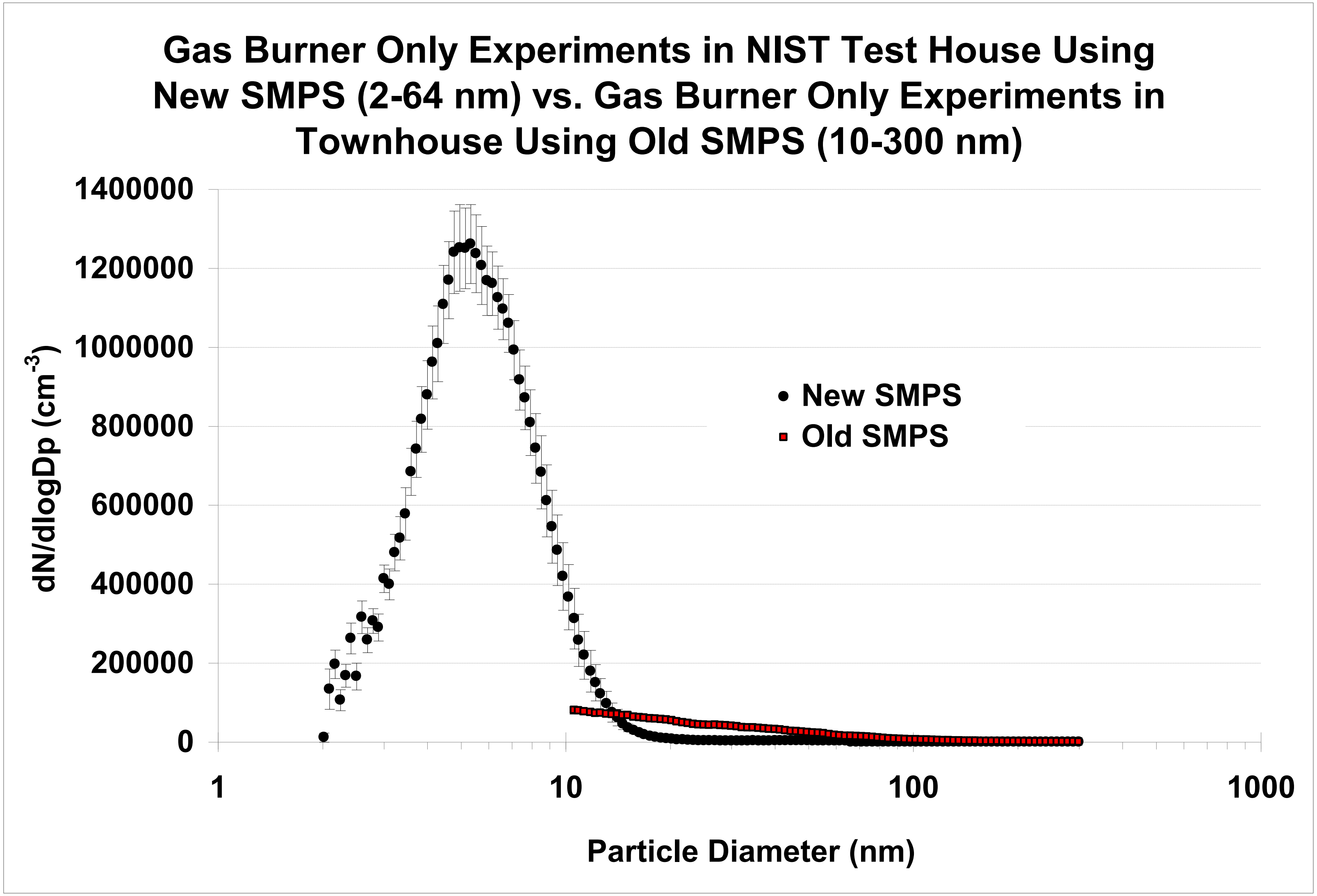
New technology allows measurements down to 2.5 nm

To extend our knowledge of the 2.5-10 nm region

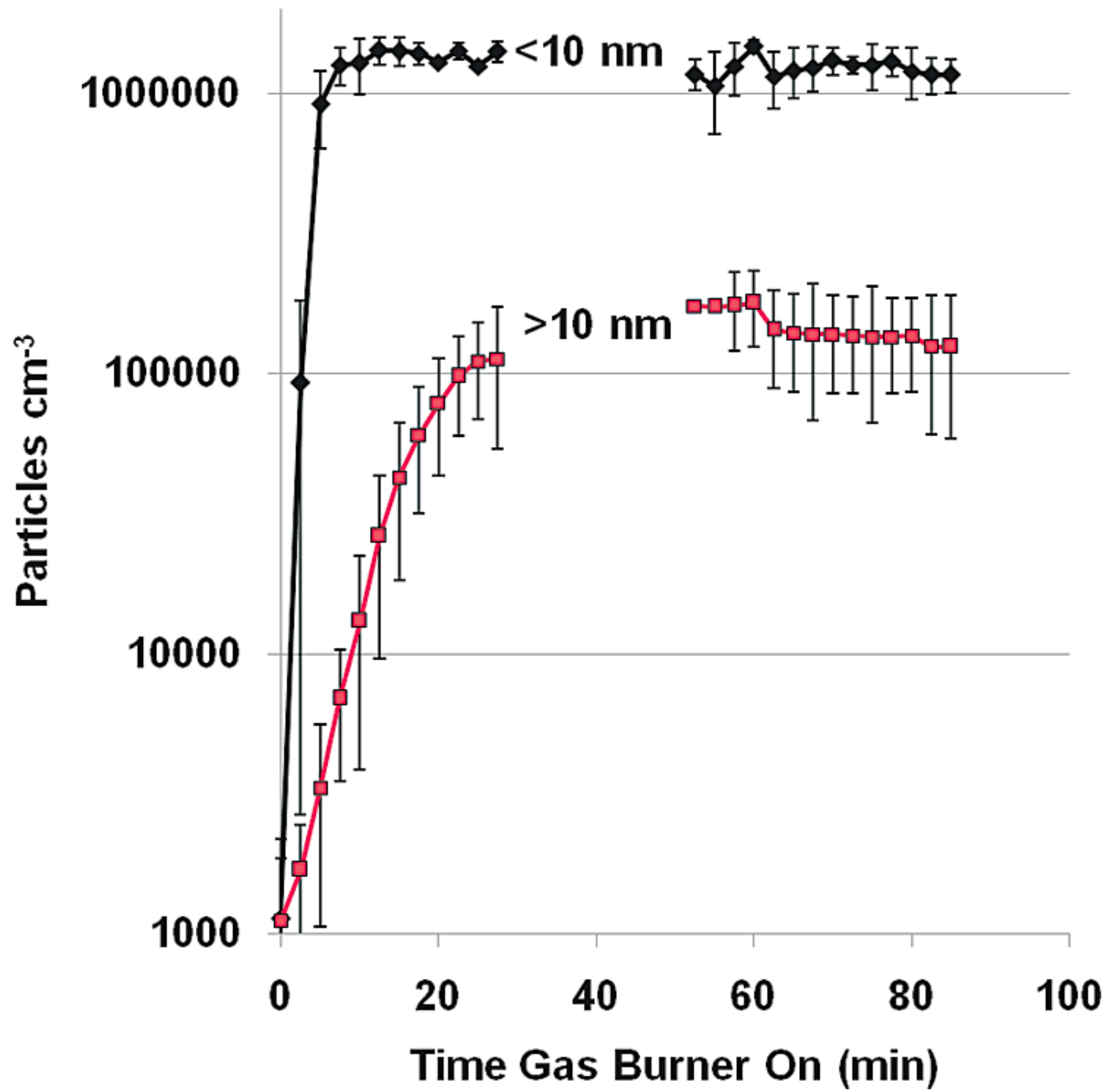
Source: Rim, D., Wallace, L., Persily A. (2010) Infiltration of outdoor ultrafine particles into a test house *Environ. Sci. Technol.* **2010,** *44,* 5908–5913

NIST Study: UFP from a Gas Stove

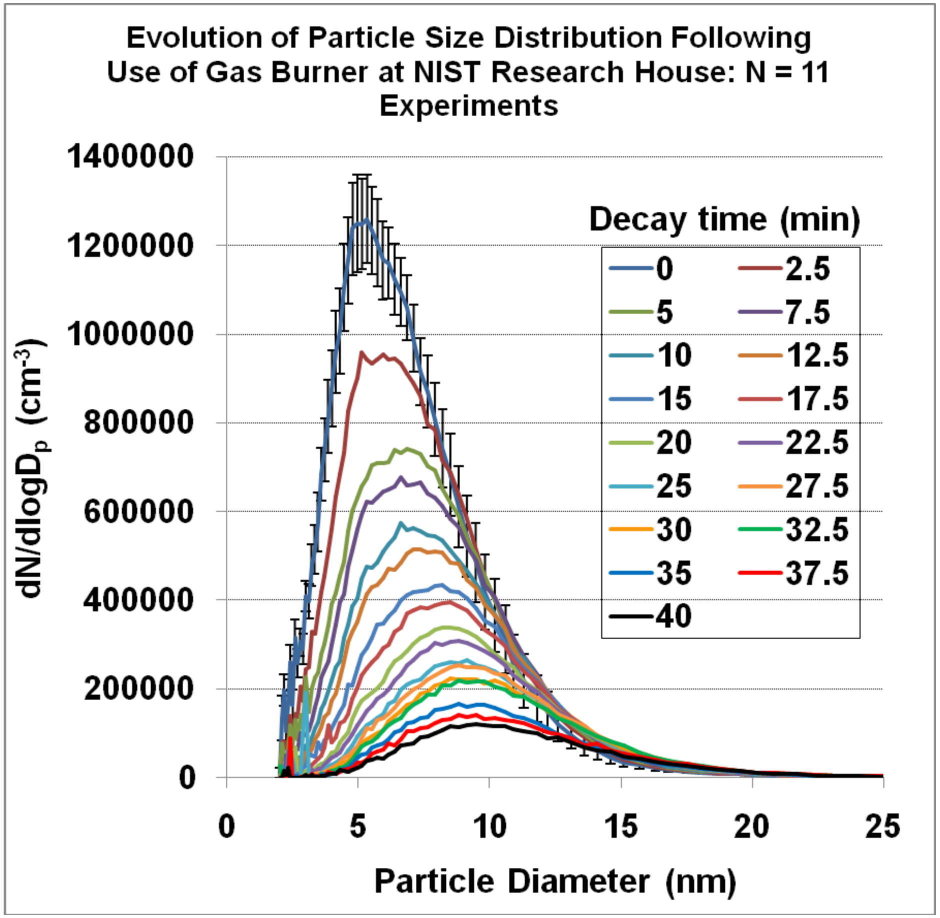
Old SMPS: >10 nm New SMPS: >2 nm



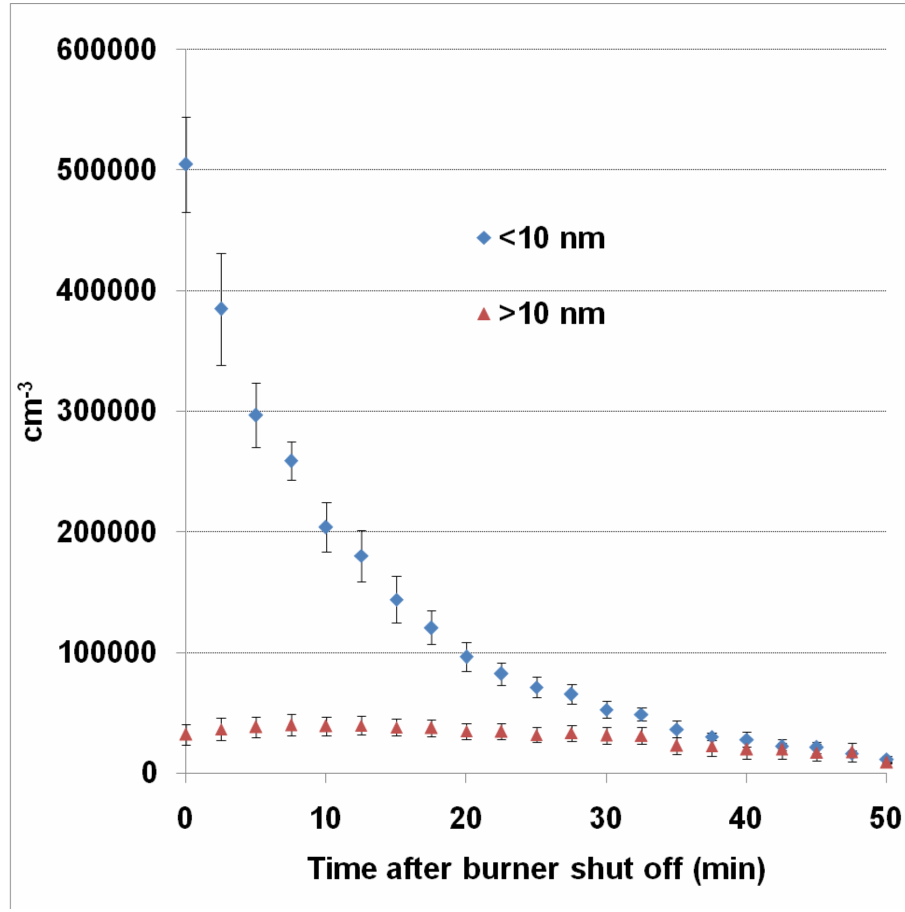
Approach to Steady State: 4 Gas Burners on High



Results------Gas Burner



Average size distribution of particles produced by a single gas burner on High setting (N = 11 experiments, 10 min each)



Particle concentrations during the decay period for particles > & < 10 nm

Two Kinds of Particles from Gas Burners

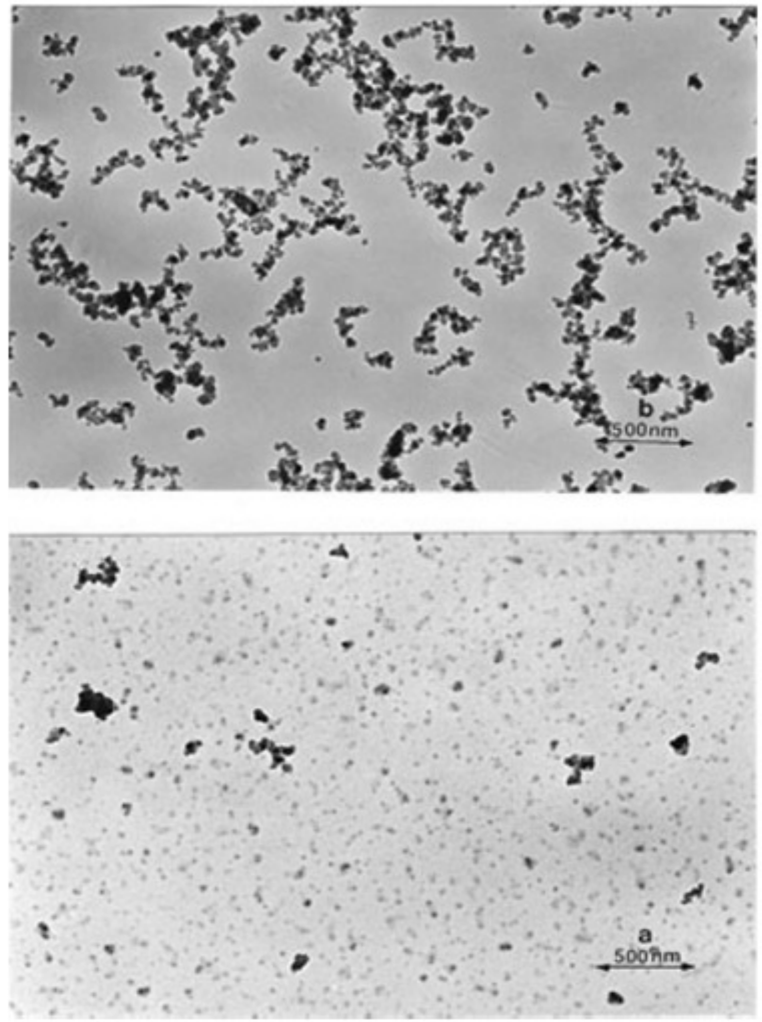
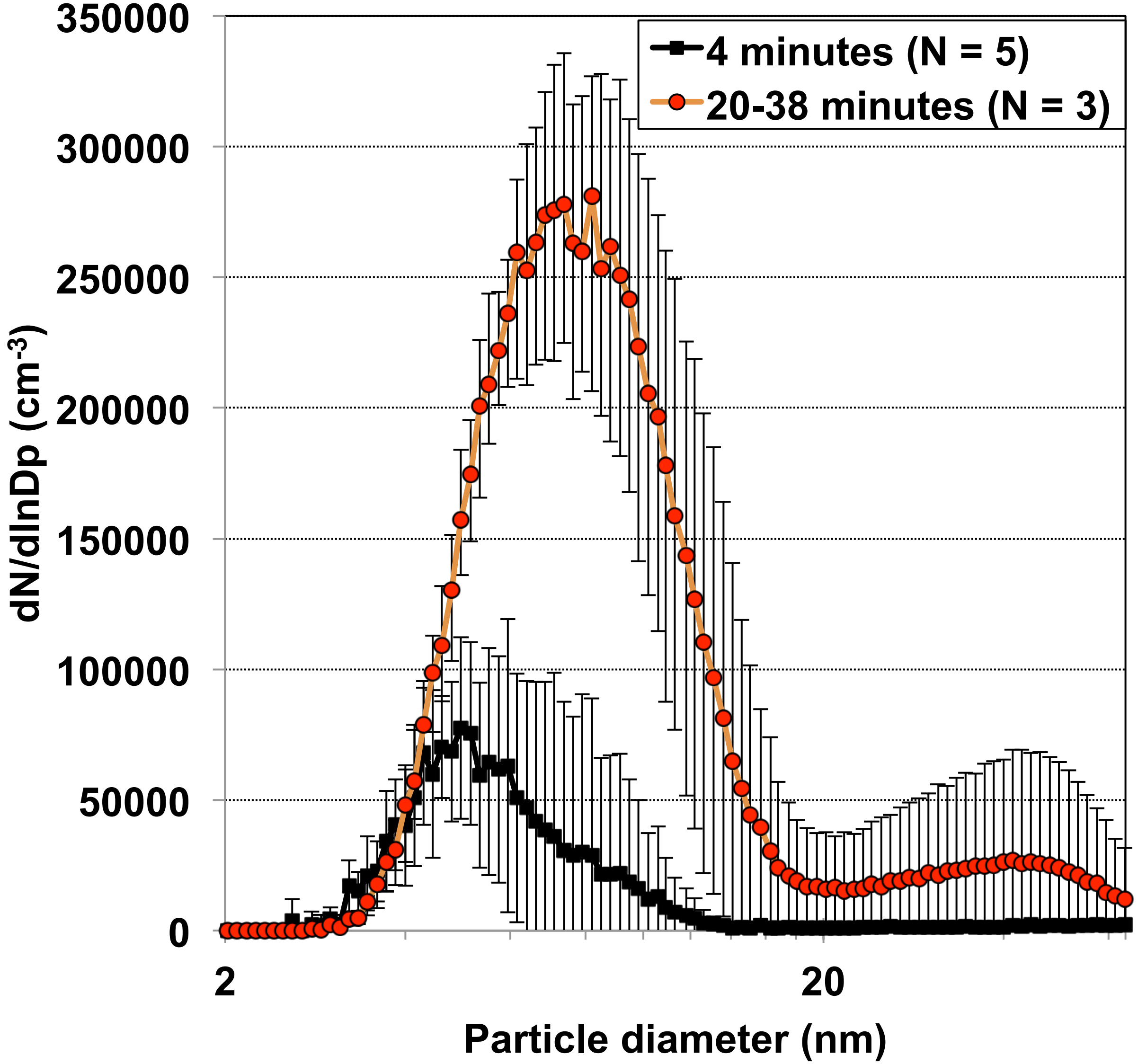
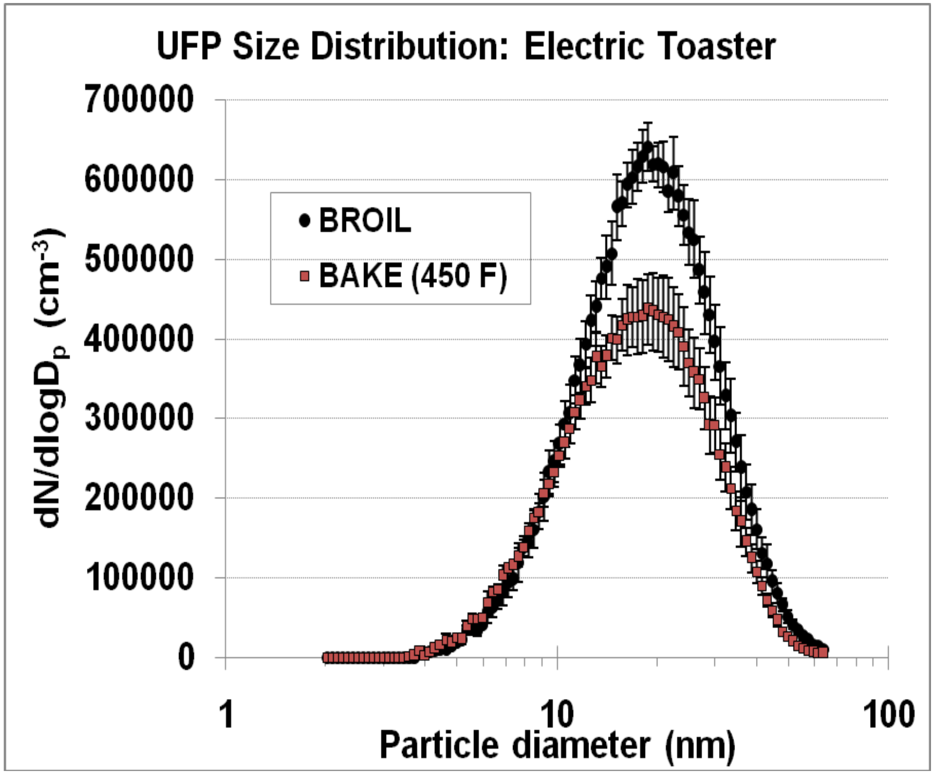


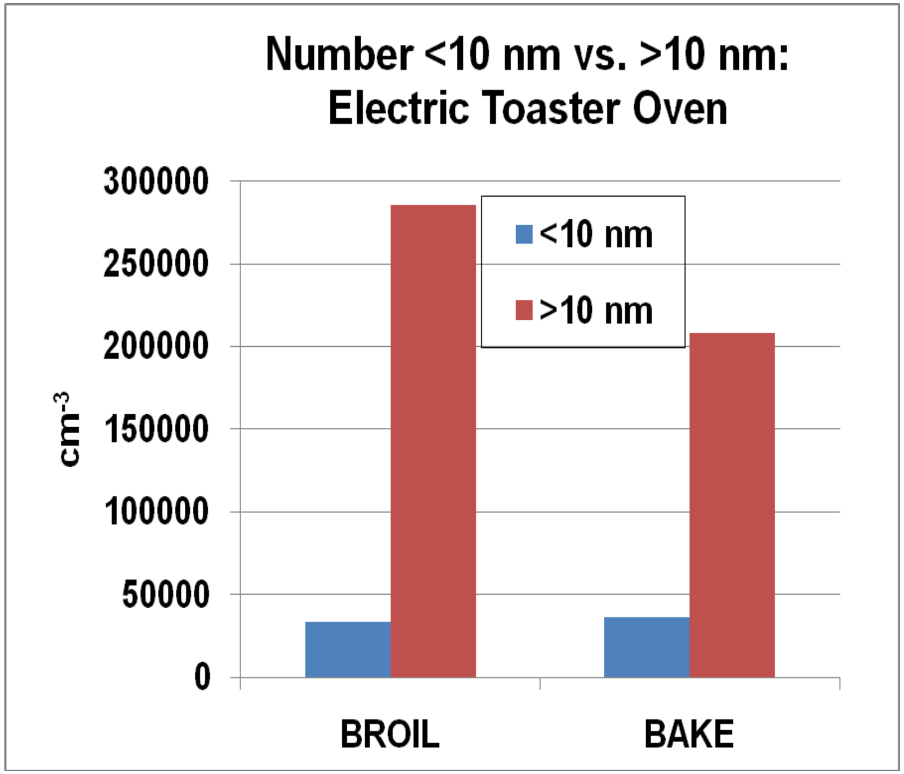
FIG. 1. Particles sampled From Axial Heights of (a) Z = 20 mm and (b) Z = 50 mm of the Laminar Ethene Diffusion Flame. The isolated translucent precursor particles dominate the lower axial region of the flame while opaque aggregates occupy the upper region of the flame.

UFP from Electric Stovetop Coil



Electric Toaster Oven





Average size distribution of particles produced by electric toaster oven (N = 6 experiments)

Particle concentrations during the peak period for particles > & < 10 nm

Cooking conclusions

- Both gas and electric stoves produce copious UFP. Processes include
  - Combustion
  - Heating element emission
- Stovetops produce mostly UFP < 10 nm
- Ovens produce mostly UFP > 10 nm
- Emission rates ≈ 10^{12}/minute
- Composition of electric stove UFP?

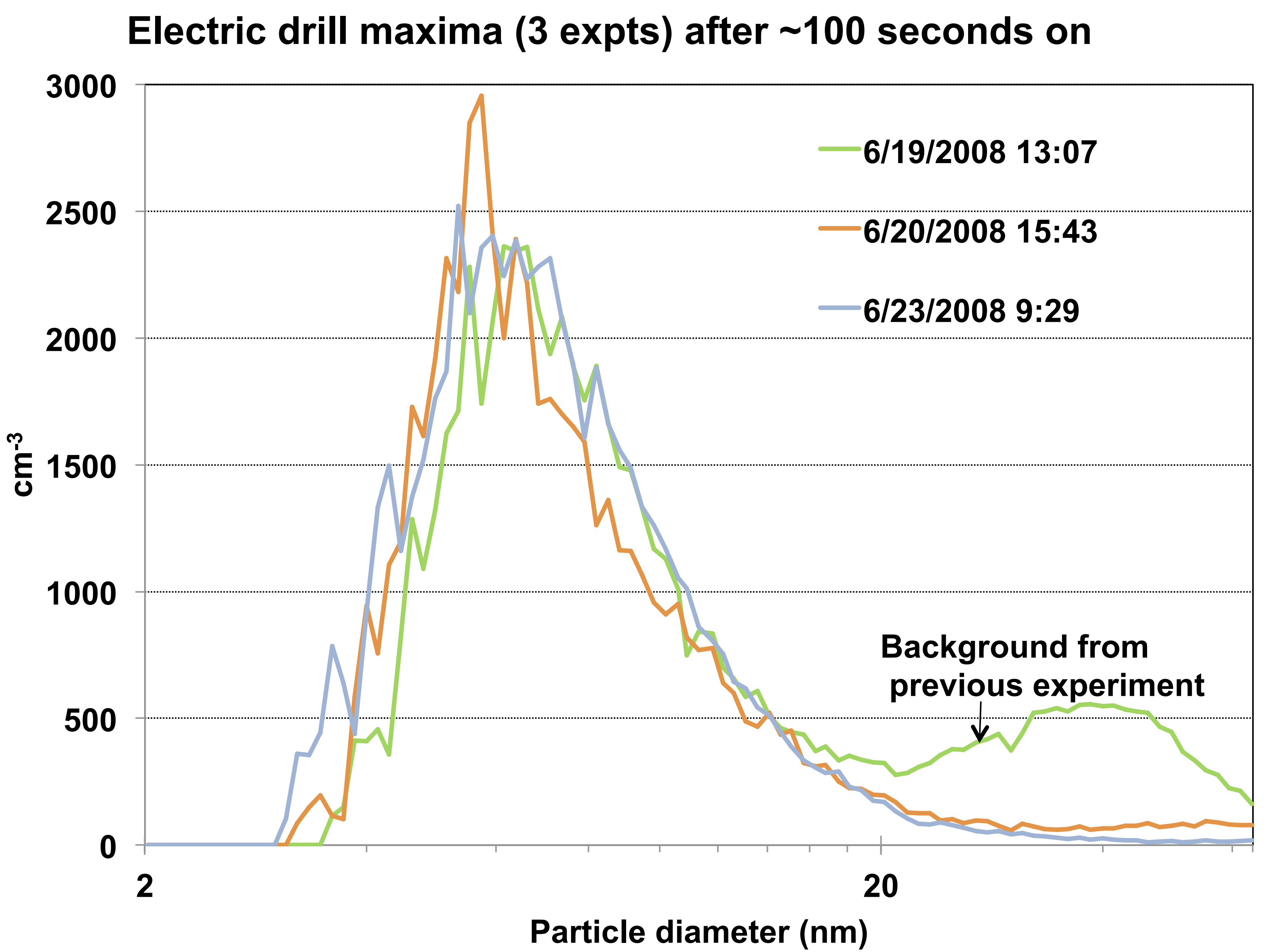
Ultrafines from Electric Motors

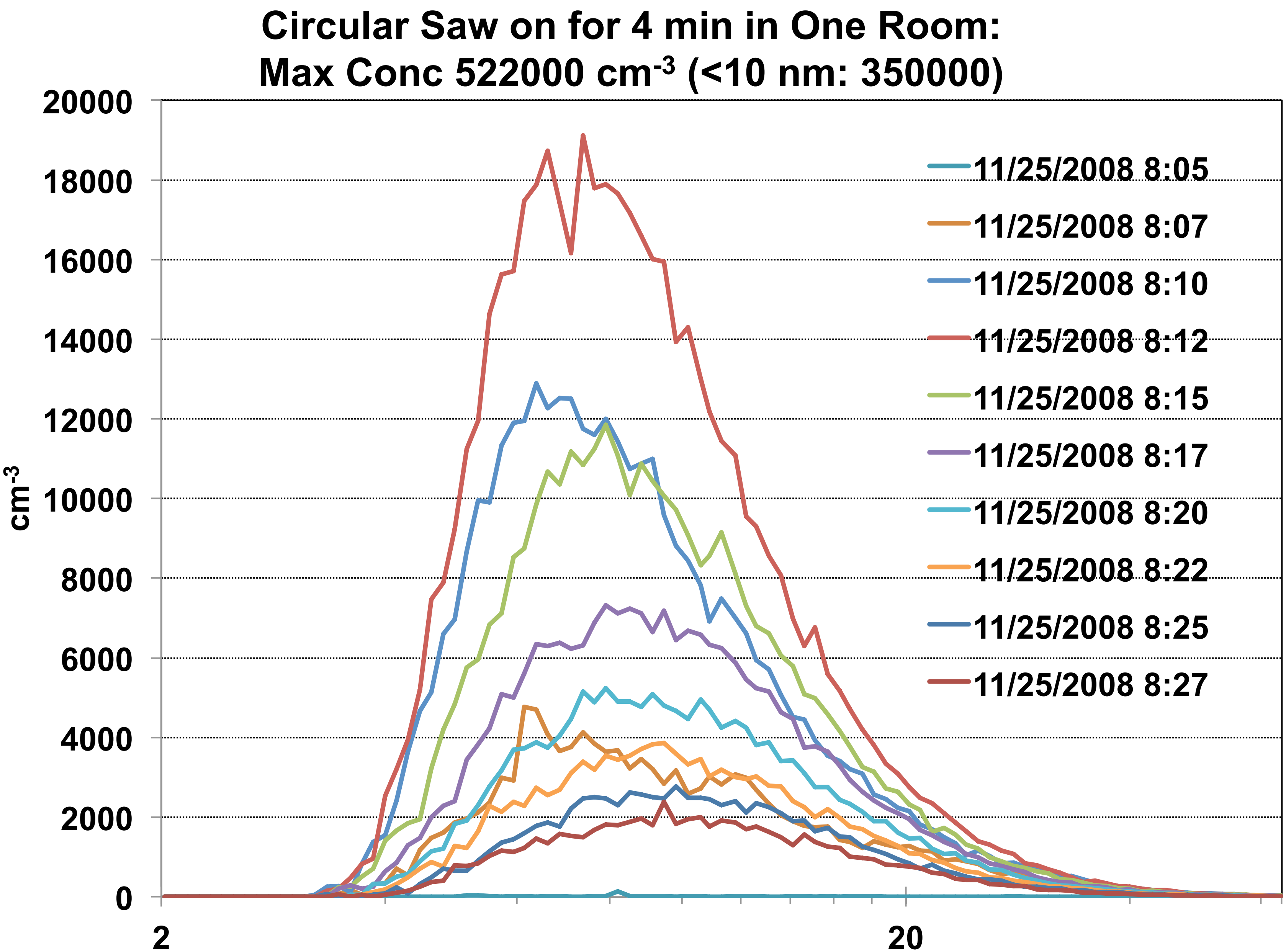
- Probably produced by spark generation between graphite brushes and copper commutator
- Copper a main constituent (40-50% by mass); remainder probably mostly carbon

  – Szymczak, Wilfried, Menzel, Norbert, and Keck, Lothar. Emission of ultrafine copper particles by universal motors controlled by phase angle. *J Aerosol Sci* 38(5), 520-531. 2007.

Experimental Protocol

- Two scenarios
  - Source and SMPS in same room (volume 30 m^3), door closed
  - Different rooms, door open (total volume >300 m^3)
- Power tools on 2 minutes, off 1, on 2
- Vacuum cleaner on 20 minutes

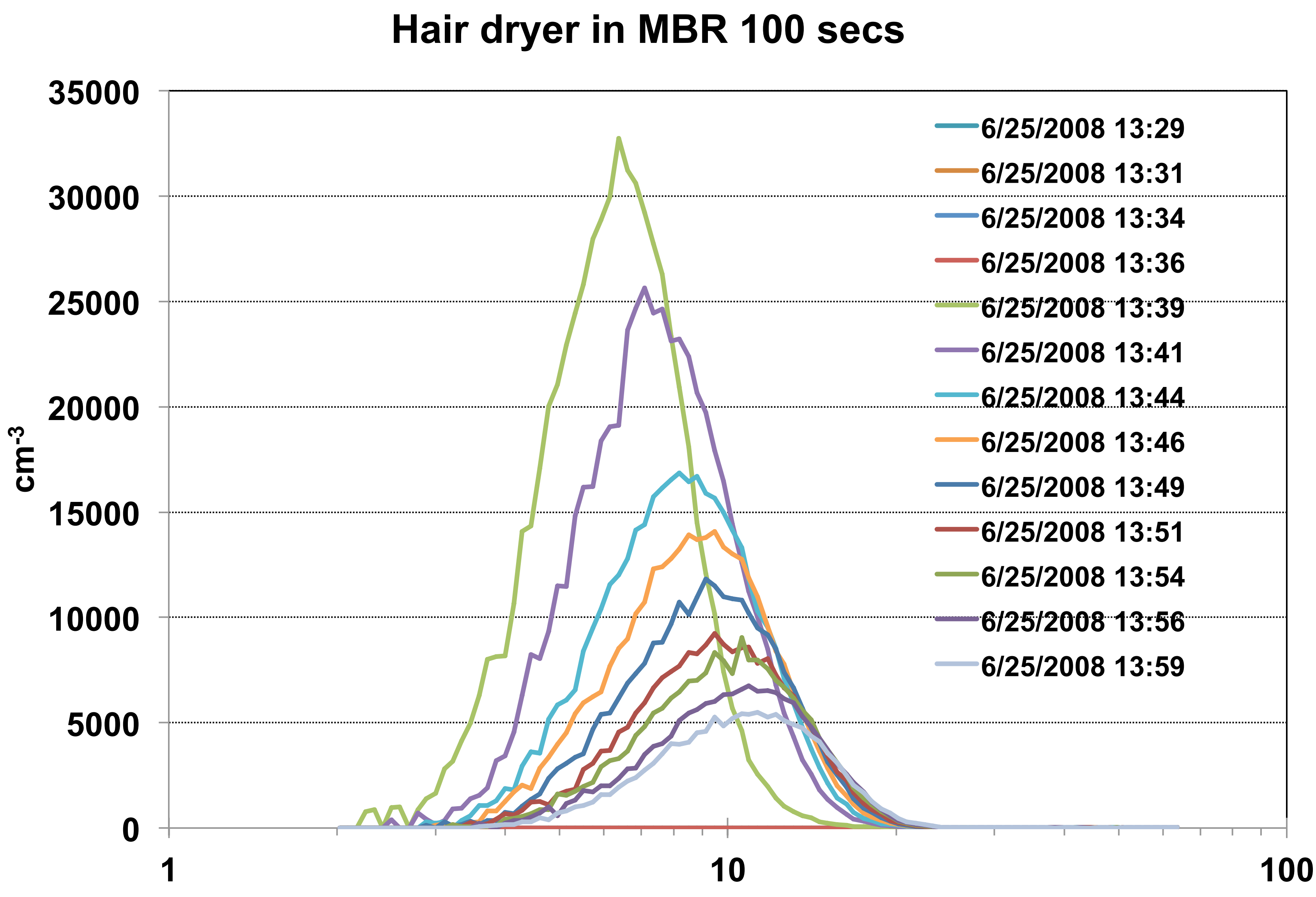

Electric drill maxima (3 expts) after ~100 seconds on
3000
2500
2000
1500
1000
500
0
cm-3
2
20
Particle diameter (nm)
6/19/2008 13:07
6/20/2008 15:43
6/23/2008 9:29
Background from previous experiment



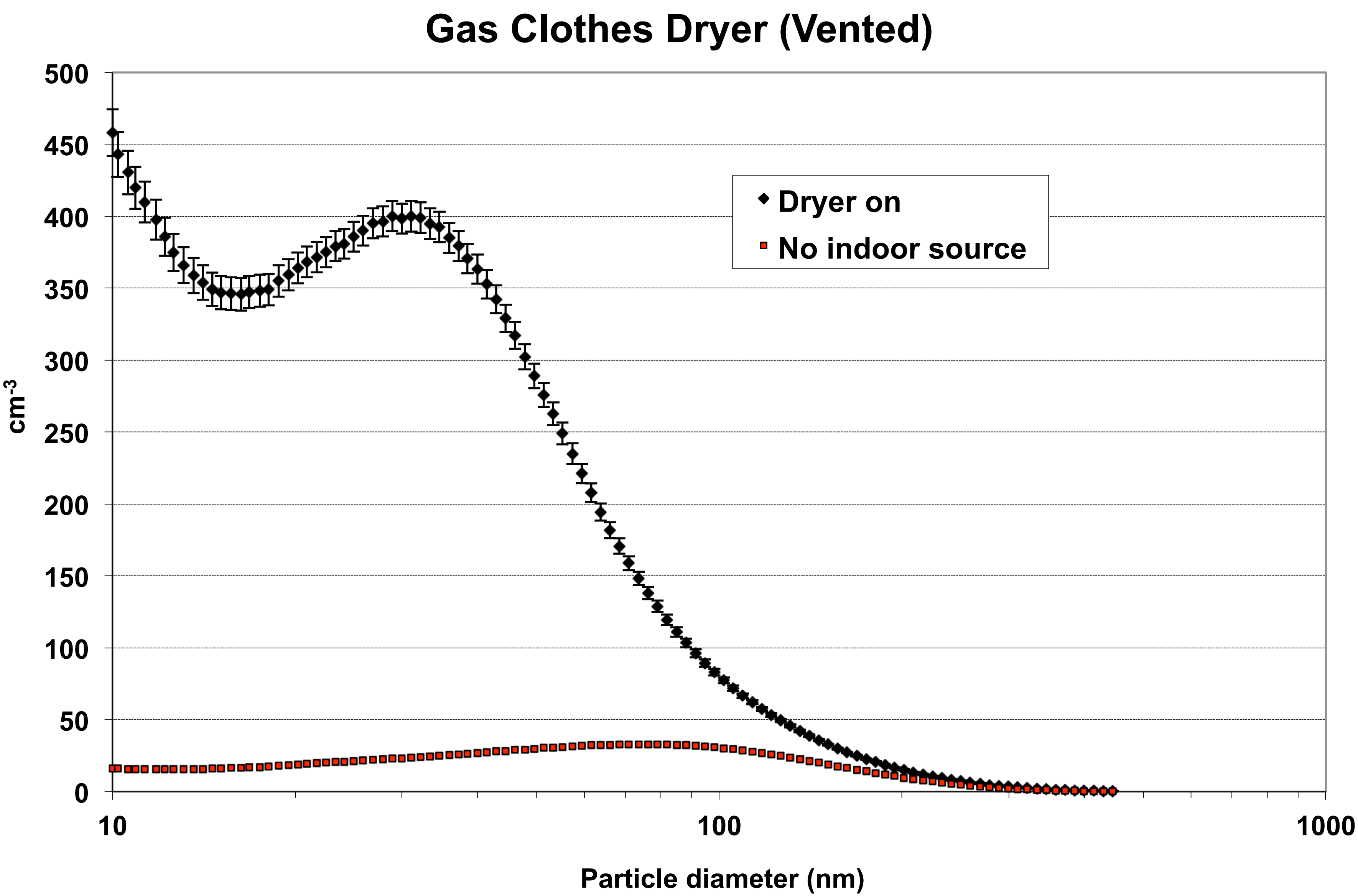
Circular Saw on for 4 min in One Room:
Max Conc 522000 cm^{-3} (<10 nm: 350000)
cm^{-3}
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0
2
20
11/25/2008 8:05
11/25/2008 8:07
11/25/2008 8:10
11/25/2008 8:12
11/25/2008 8:15
11/25/2008 8:17
11/25/2008 8:20
11/25/2008 8:22
11/25/2008 8:25
11/25/2008 8:27

Peak Concentrations, Modes, and Estimated Emission Rates

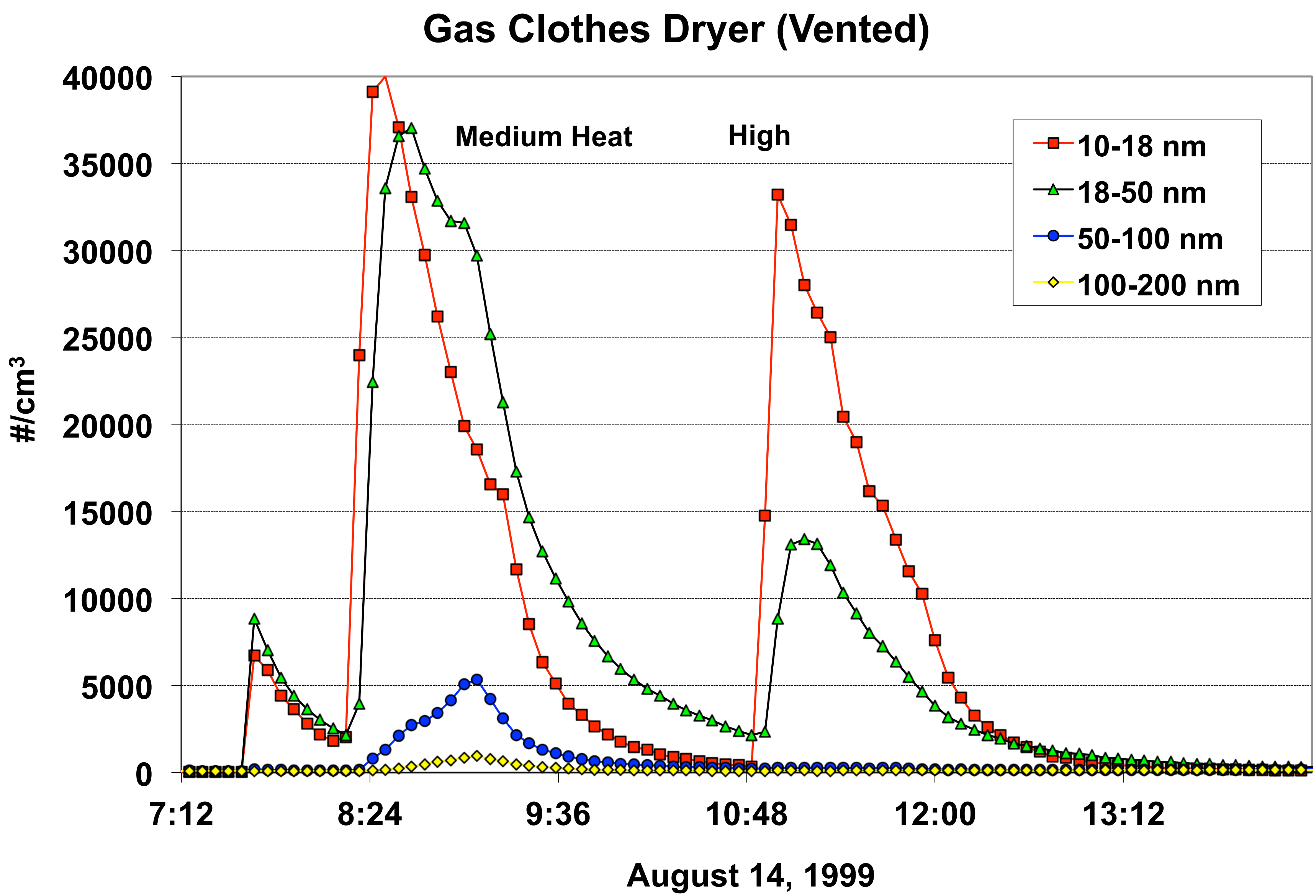
Power Tool	Time on (minutes)	Location	N ≤10 nm (000 cm^{-3})	N>10 nm (000 cm^{-3})	Mode (nm)	Emission rate (10^{12} min^{-1})
circular saw	4	Same room	701	553	8.3	9.4
		Different room	59	62	8.8	11.5
jig saw	4	Same room	430	200	7	4.7
		Different room	16	12	7.8	2.7
belt sander	4	Same room	235	87	6	2.4
		Different room	13	7	6.6	1.9
reciprocating saw	4	Same room	88	50	7.1	1.0
		Different room	3	2	6.4	0.5
drill	10	Same room	56	21	7.2	0.2
		Different room	7	2	6.9	0.3
vacuum cleaner	20	Same room, door closed	28	22	8.4	0.1
		Same room, door open	9	8	7.6	0.3
hedge clippers	4	Same room	0	0	N/A	0
compressor	5	Same room	0	0	N/A	0
pump	20	Same room	0	0	N/A	0
shaver	10	Same room	0	0	N/A	0



Hair dryer in MBR 100 secs
cm-3
35000
30000
25000
20000
15000
10000
5000
0
1
10
100
6/25/2008 13:29
6/25/2008 13:31
6/25/2008 13:34
6/25/2008 13:36
6/25/2008 13:39
6/25/2008 13:41
6/25/2008 13:44
6/25/2008 13:46
6/25/2008 13:49
6/25/2008 13:51
6/25/2008 13:54
6/25/2008 13:56
6/25/2008 13:59



Source: **Wallace, L.A. (2005). Ultrafine particles from a vented gas clothes dryer. *Atmos Environ* 39:5777-5786.**


Gas Clothes Dryer (Vented)
40000
35000
30000
25000
20000
15000
10000
5000
0
#/cm3
Medium Heat
High
10-18 nm
18-50 nm
50-100 nm
100-200 nm
7:12
8:24
9:36
10:48
12:00
13:12
August 14, 1999

Indoor UFP Sources : Status

Combustion

Source of UFPs	*Test data*
Gas burners	Lots
Gas oven	Lots
Gas clothes dryer	Some
Gas hot water heater	No
Candles	Some
Incense	Some
In-vehicle exposure to traffic	Some

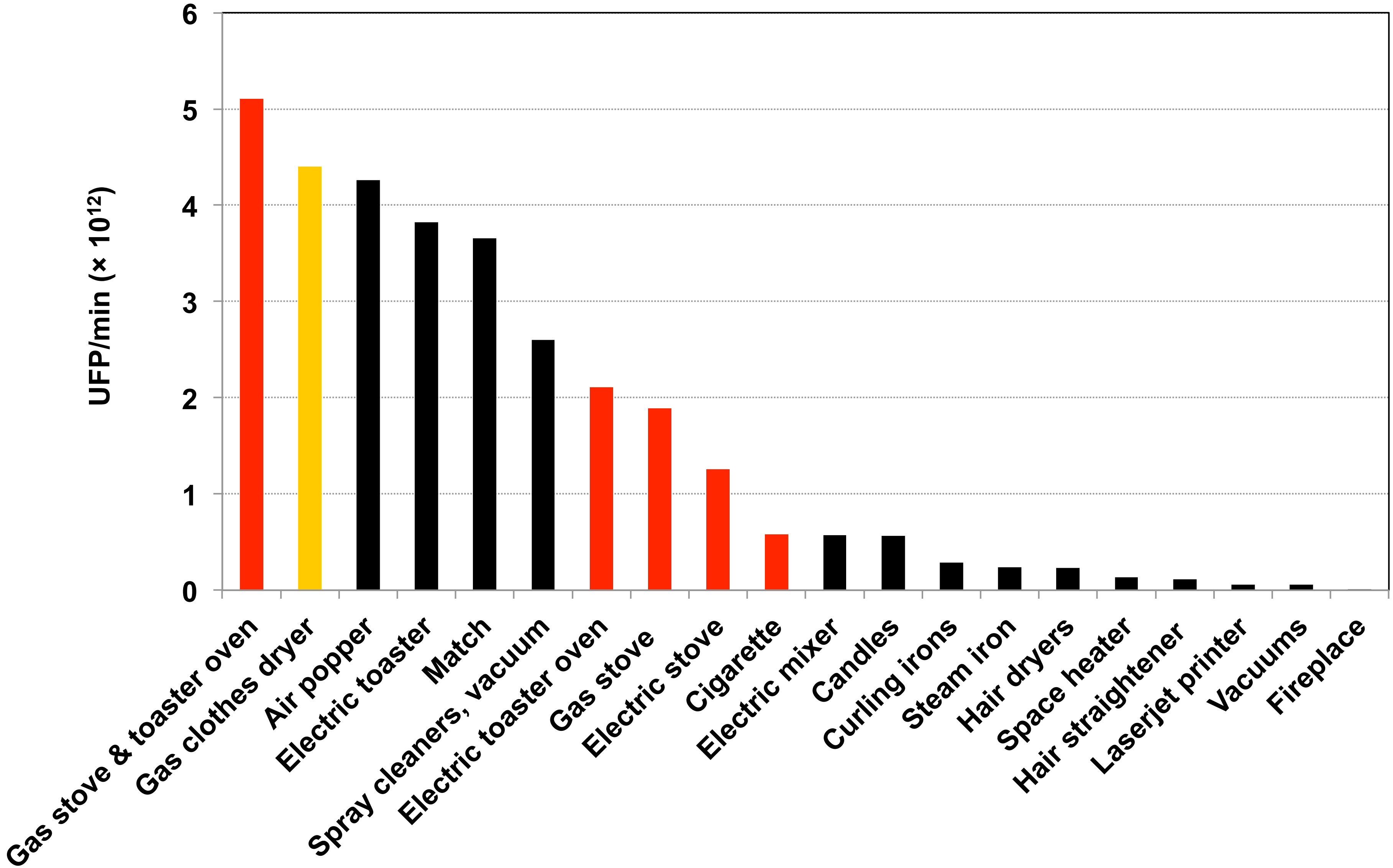
Electric Motors

Source of UFPs	*Test data*
Vacuum Cleaner	Some
Washing machine	No
Electric mixer/blender	Some
Power Tools	Some

Electric Heating Elements

Source of UFPs	*Test data*
Electric stove	Lots
Toaster	Lots
Toaster oven	Lots
Electric space heater	Some (no emitters yet)
Electric blanket	Some (no emitters)
Hair dryer	Some
Curling iron	No
Hair straightener	Some
Steam iron	Some
Air popper	Some
Hot plate	No
Coffeemaker	No

Mean Emission Rates of Household Appliances

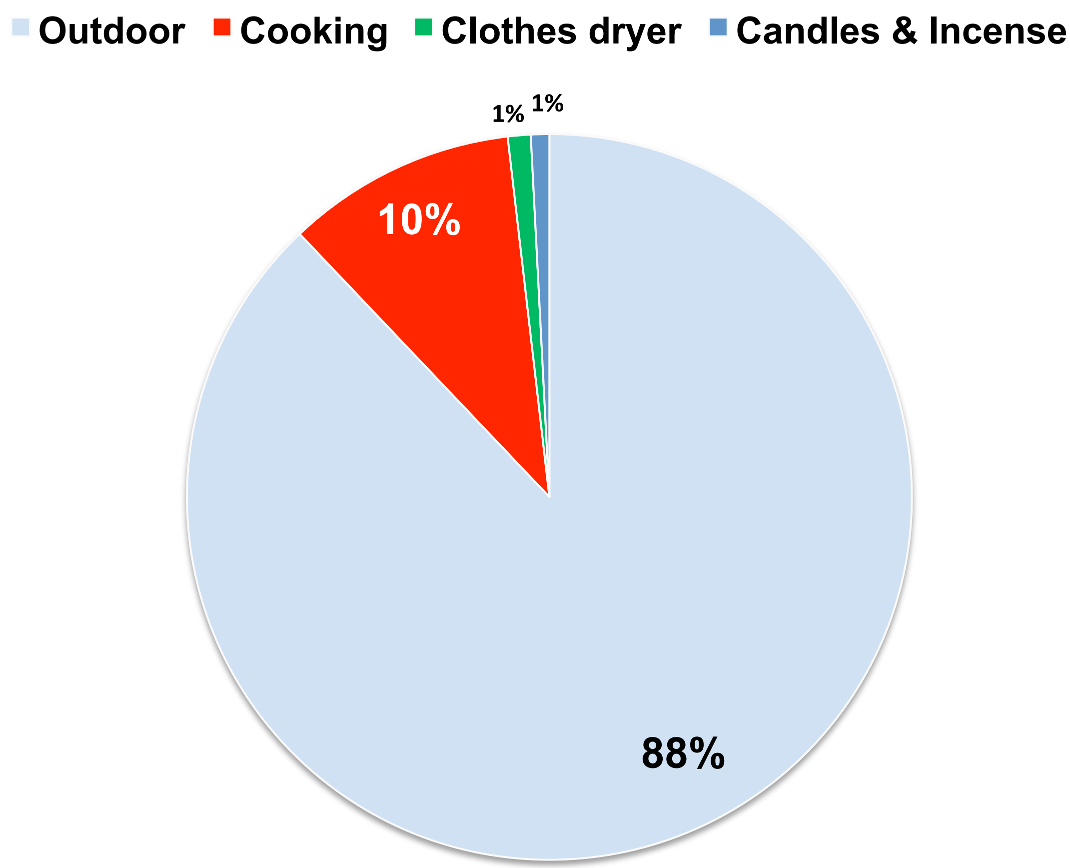


Wallace, LA and Ott, WR. (2010). Personal exposure to ultrafine particles. *J Expos Sci & Environ Epidemiol* 21:20-30.

Processes Producing UFP

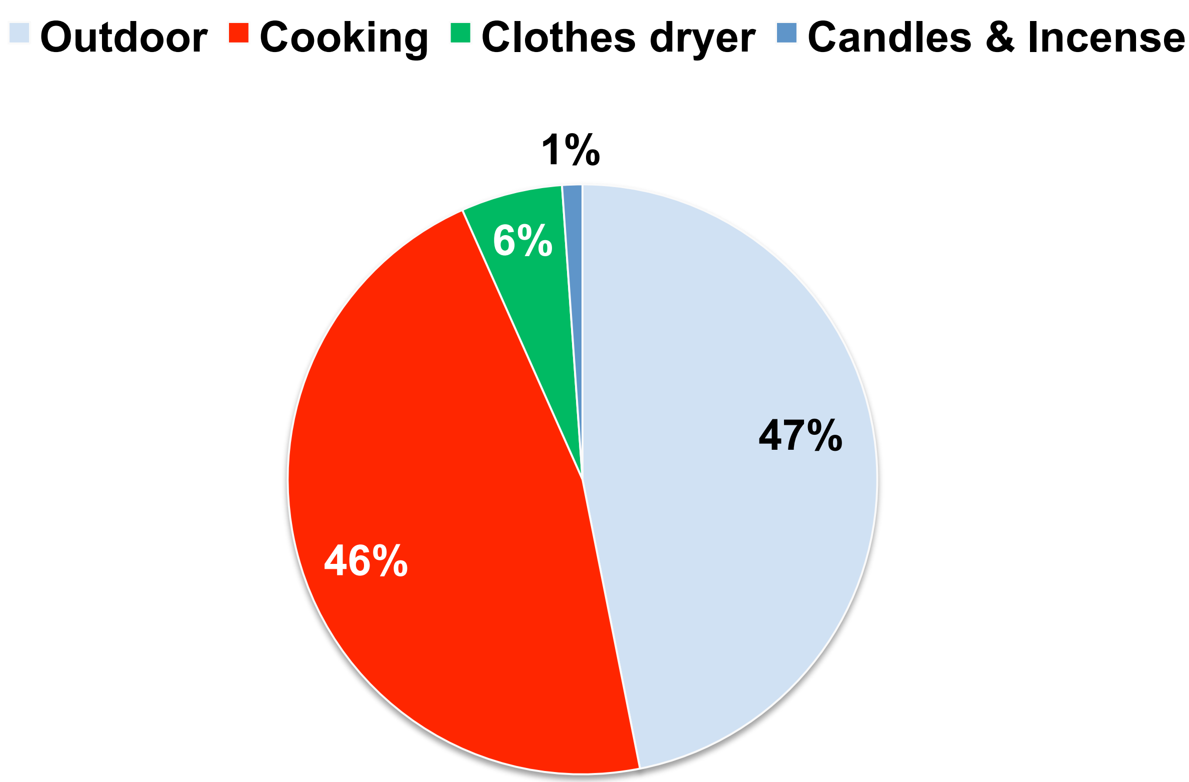
- Combustion (gas stoves, gas clothes dryers, cigarettes, candles)
- Heating elements (electric stoves, toaster ovens, hair dryers)
- Electric motors (power tools, vacuum cleaner)
- Laserjet printers?

Percent of Time Affected by Major UFP Sources



Source: Wallace, L.A. and Howard-Reed, C.H. Continuous Monitoring of Ultrafine, Fine, and Coarse Particles in a Residence for 18 Months in 1999-2000. *J Air Waste Manage. Assoc*. **52**(7):828-844. 2002.

Percent of Indoor Exposure to Major Sources of UFP



Source: Wallace, L.A. and Howard-Reed, C.H. Continuous Monitoring of Ultrafine, Fine, and Coarse Particles in a Residence for 18 Months in 1999-2000. *J Air Waste Manage. Assoc*. **52**(7):828-844. 2002.

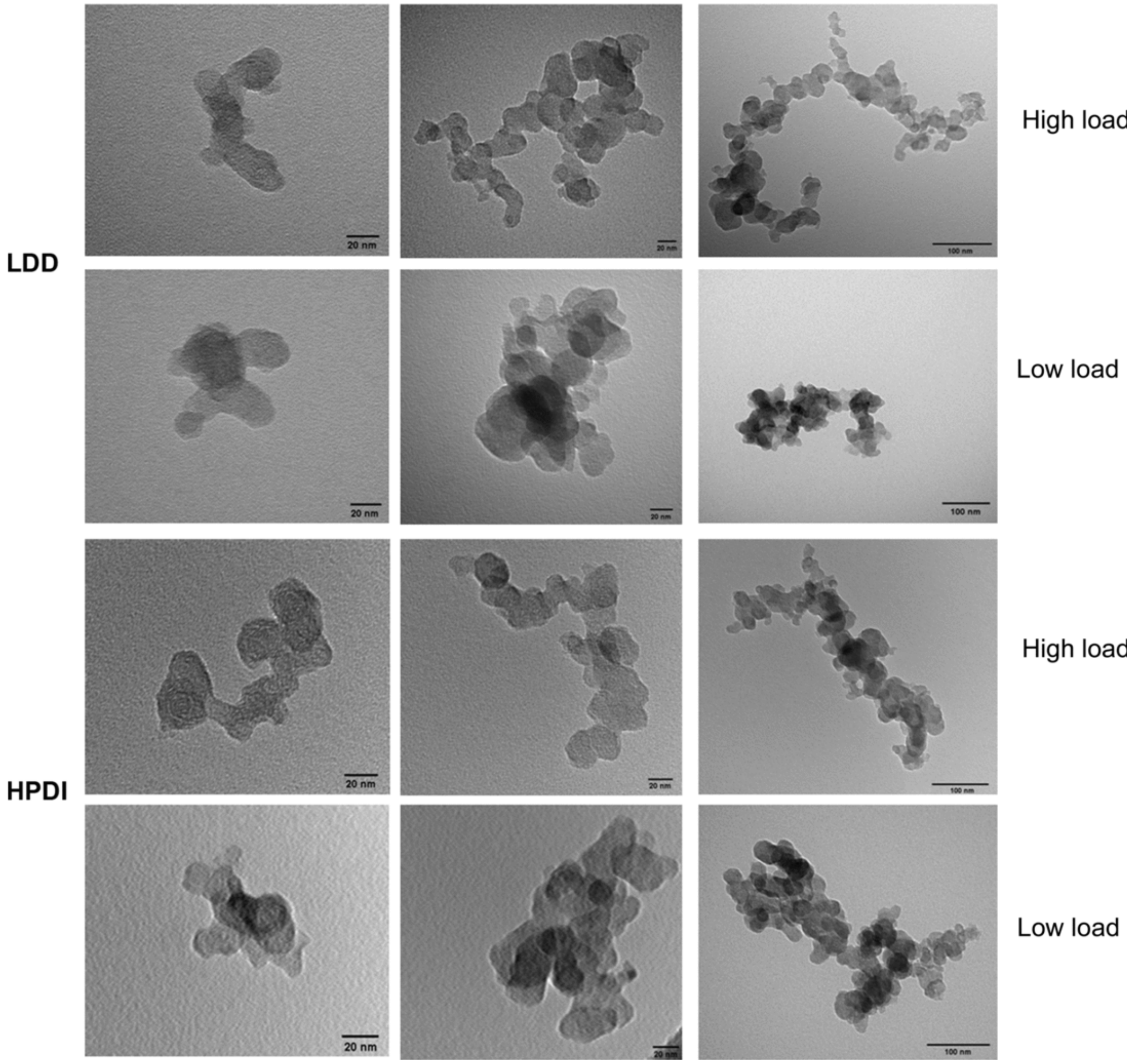
Conclusions

- Indoor UFP sources are at least as important as outdoor infiltration
- Cooking (gas or electric) appears to be the major indoor source
- Other high emitters
  - Vented gas clothes dryer
  - Appliances with heating elements (hair dryers)
  - Power tools

Major outdoor sources of UFP:

- **Emissions from vehicles**
- **Regional atmospheric nucleation events**

➢ **Typical outdoor concentrations may range from 5000-30,000 particles/cc; with occasional spikes to 50,000-100,000 particles/cc caused by nucleation bursts (H_2SO_4, ammonia, water vapor + sunlight)**



Source: Soewono, A. and Rogak, S. (2011) Morphology and Raman Spectra of Engine-Emitted Particulates, *Aerosol Science & Technology* **45**: 1206-1216.

Mean particle number concentrations measured for 1-2 years in world cities.

City	Ultrafine (3-100 nm)	Total	Percent ultrafine	Reference
Beijing	24,900	32,800	76	Wu et al, 2008
Helsinki	13,420	-------		Hussein et al, 2004
Alkmaar	18,300 (10-100 nm)	25,800	>71	Ruuskanen et al, 2001
Erfurt	15,700 (10-100 nm)	18,000	88	Wichmann et al, 2000
Leipzig	19,263	21,377	90	Wehner and Wiedensohler, 2003
Pittsburgh	19,800	21,988	90	Stanier et al., 2004
Atlanta	19,046	20,736	92	Woo et al., 2001
Rochester	7,310 (10-100 nm)	8,190	89	Jeong et al., 2004
Toronto	Not reported (>7 nm)	28,010		Jeong et al., 2006
Fresno		15,300		Watson et al., 2006

(Source: Modified from Table 2, Wu et al, 2008).

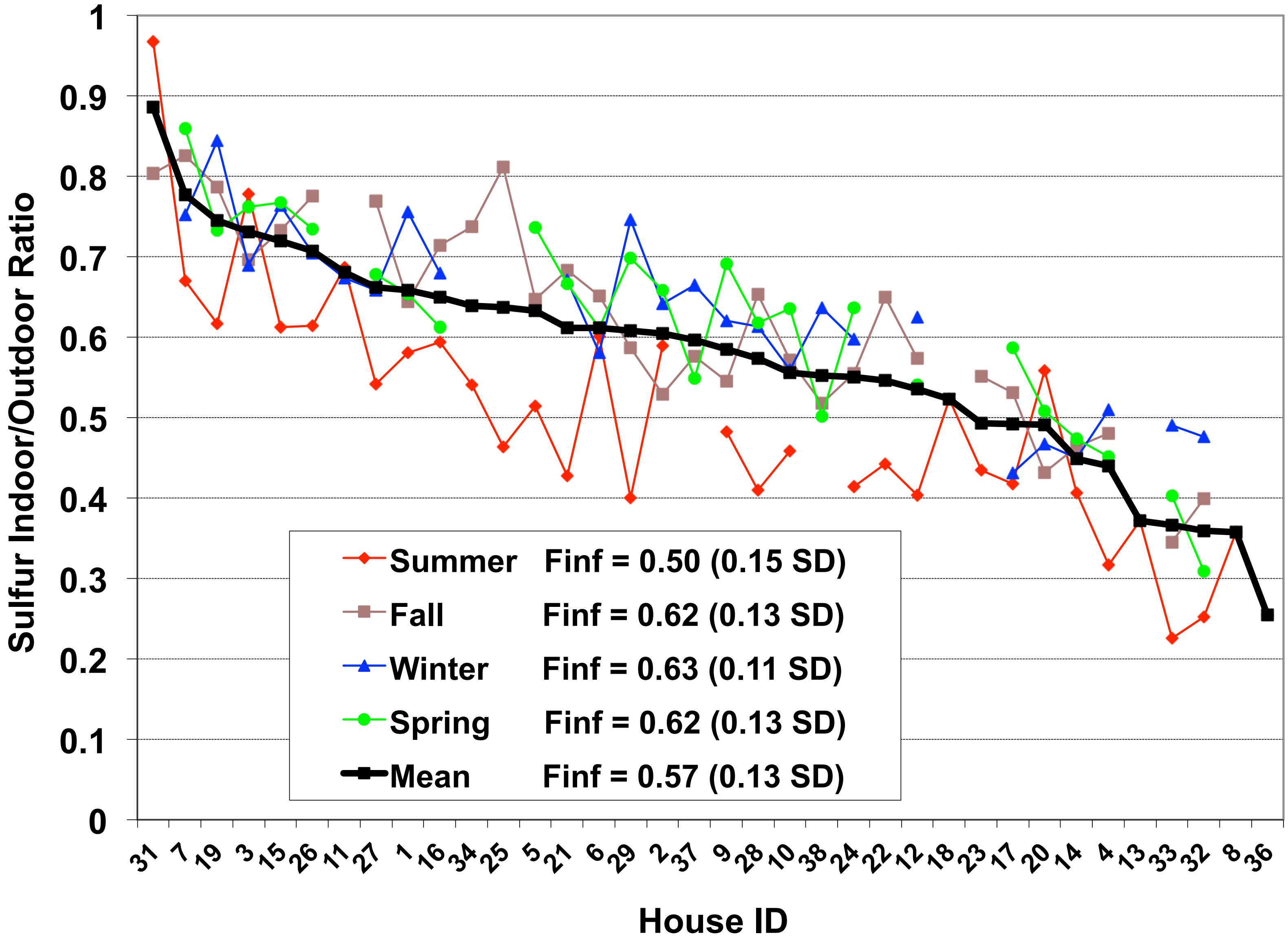
The Effect of Outdoor Sources on Indoor Exposure

Depends Heavily on the Infiltration Factor

Infiltration Factor

- Determines the fraction of outdoor air particles entering the home
- Since we are home 16 h/day, this is a major influence on our exposure to ambient PM
- Varies according to particle size
- Depends on house quality (number & size of cracks, presence of a vapor barrier, etc.), behaviors (open windows, air conditioner use)

Infiltration Factor for $PM_{2.5}$ in 37 Homes Measured Over 4 Seasons



Source: Wallace et al., 2003

Infiltration Factor (fine particles)

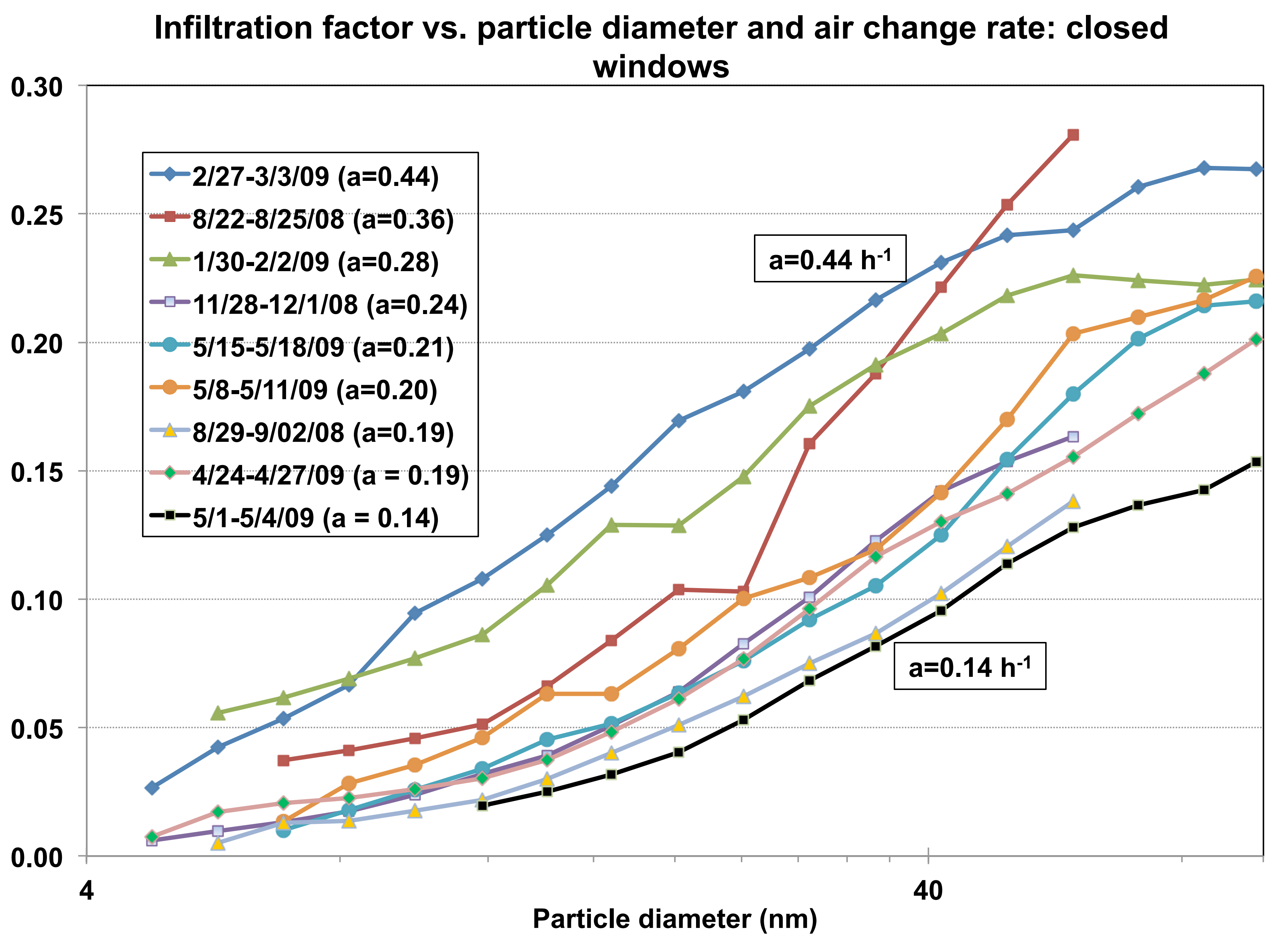
- Epi studies generally do not consider F_{inf}
- They assume exposure = outdoor concentration (or some constant fraction)
- But we see that homes can maintain widely different F_{inf} (factor of 3) over all seasons
- So indoor home exposure varies by a factor up to 3 also within a single geographic area
- Therefore much exposure misclassification!

The infiltration factor

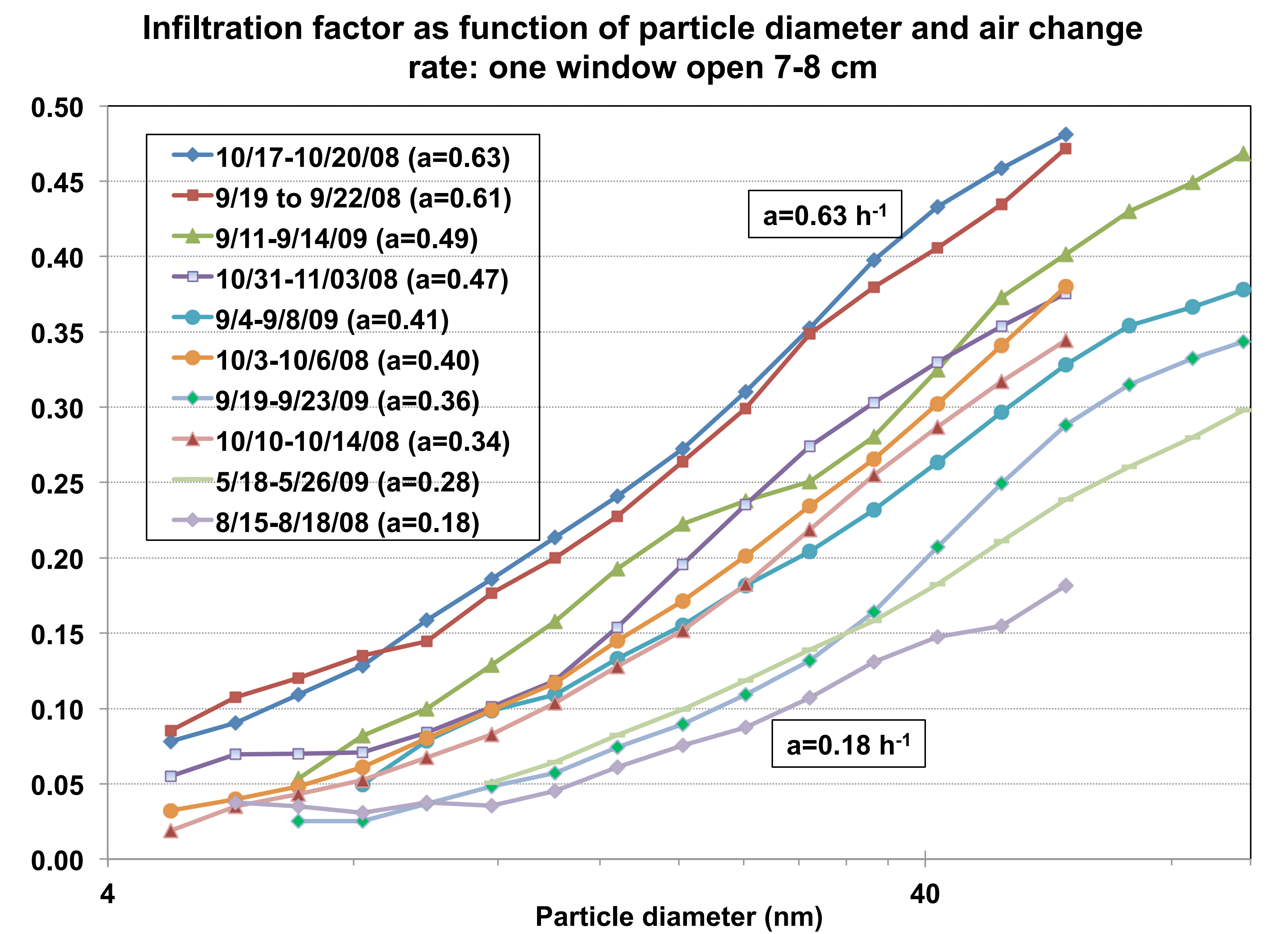
$$F_{inf} = \frac{Pa}{a + k}$$

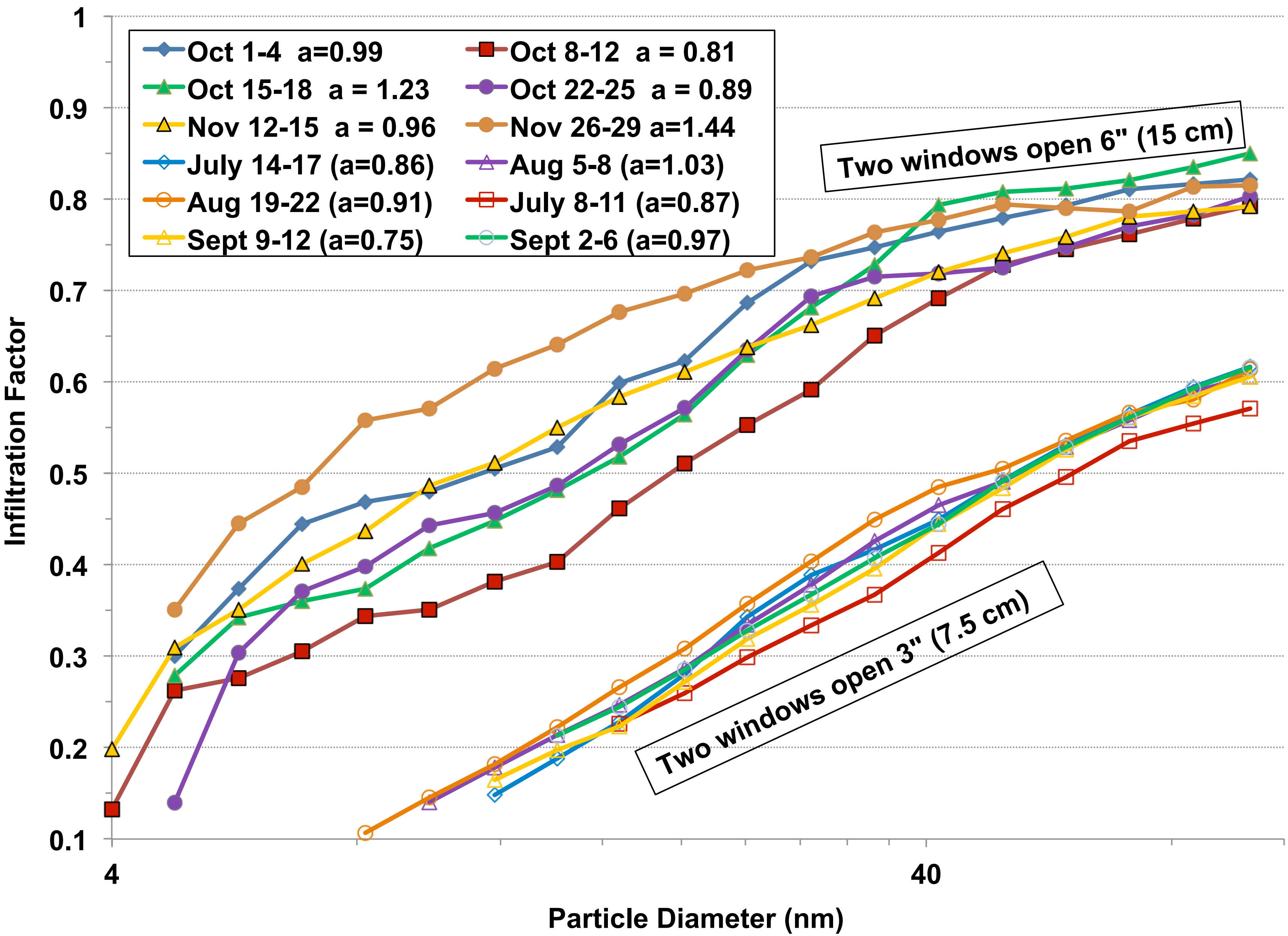
Measuring F_{inf} for UFP

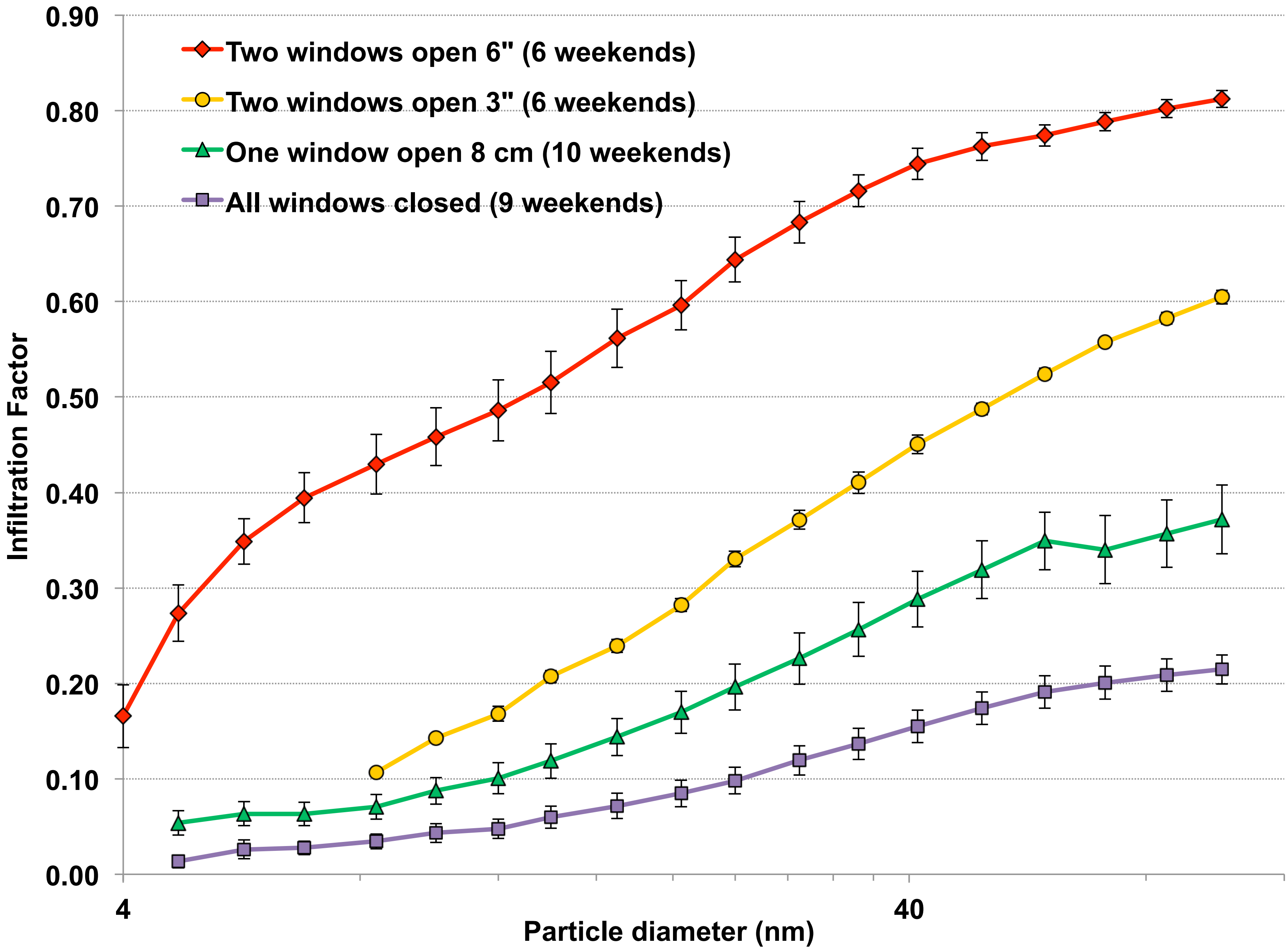
- Uninhabited house (e.g. NIST test house)
  - Indoor-outdoor sampling over 3-day periods (weekends) with no indoor sources
  - Air exchange measured by recurrent injection SF6
  - Solve nonlinear recursive relation (Switzer & Ott, 2001) for P and k to estimate F_{inf}
  - Check using C_{in}/C_{out} ratio



Infiltration factor vs. particle diameter and air change rate: closed windows
0.30
0.25
0.20
0.15
0.10
0.05
0.00
4
40
Particle diameter (nm)
2/27-3/3/09 (a=0.44)
8/22-8/25/08 (a=0.36)
1/30-2/2/09 (a=0.28)
11/28-12/1/08 (a=0.24)
5/15-5/18/09 (a=0.21)
5/8-5/11/09 (a=0.20)
8/29-9/02/08 (a=0.19)
4/24-4/27/09 (a = 0.19)
5/1-5/4/09 (a = 0.14)
a=0.44 h-1
a=0.14 h-1


Infiltration factor as function of particle diameter and air change rate: one window open 7-8 cm
0.50
0.45
0.40
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
4
40
Particle diameter (nm)
10/17-10/20/08 (a=0.63)
9/19 to 9/22/08 (a=0.61)
9/11-9/14/09 (a=0.49)
10/31-11/03/08 (a=0.47)
9/4-9/8/09 (a=0.41)
10/3-10/6/08 (a=0.40)
9/19-9/23/09 (a=0.36)
10/10-10/14/08 (a=0.34)
5/18-5/26/09 (a=0.28)
8/15-8/18/08 (a=0.18)
a=0.63 h-1
a=0.18 h-1


Oct 1-4 a=0.99
Oct 8-12 a = 0.81
Oct 15-18 a = 1.23
Oct 22-25 a = 0.89
Nov 12-15 a = 0.96
Nov 26-29 a=1.44
July 14-17 (a=0.86)
Aug 5-8 (a=1.03)
Aug 19-22 (a=0.91)
July 8-11 (a=0.87)
Sept 9-12 (a=0.75)
Sept 2-6 (a=0.97)
Two windows open 6" (15 cm)
Two windows open 3" (7.5 cm)
Infiltration Factor
Particle Diameter (nm)
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
4
40

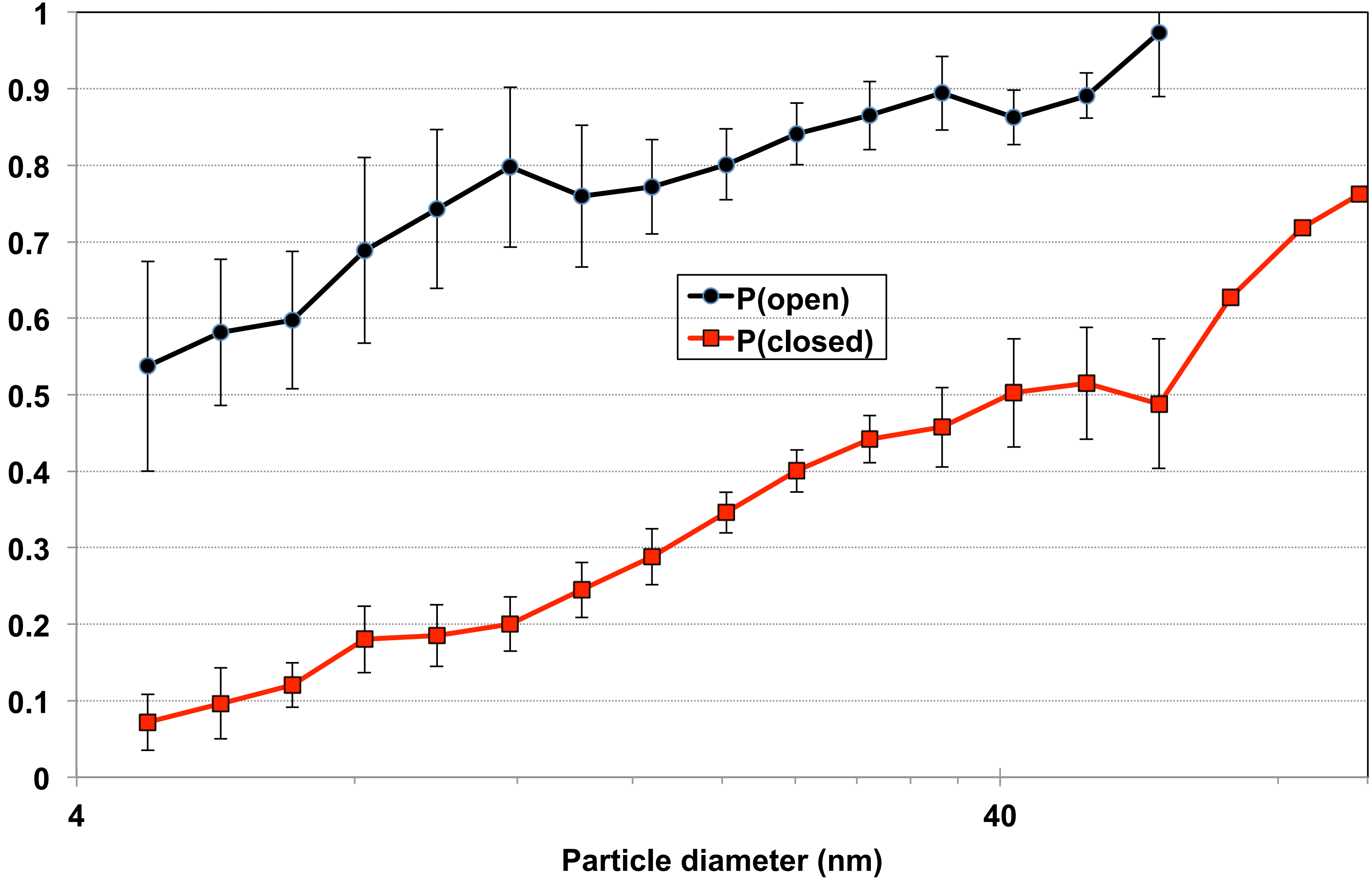


Two windows open 6" (6 weekends)
Two windows open 3" (6 weekends)
One window open 8 cm (10 weekends)
All windows closed (9 weekends)
0.90
0.80
0.70
0.60
0.50
0.40
0.30
0.20
0.10
0.00
Infiltration Factor
4
40
Particle diameter (nm)

Penetration coefficient *P*

- Depends on house construction, occupant behavior, particle diameter
- Well-built house ($P<<1$)
  - Few cracks, well-sealed windows and doors
- Closed windows ($P \leq 1$)
- Ultrafine particles ($P<<1$): lost in cracks (Brownian motion)
- Hard to measure!

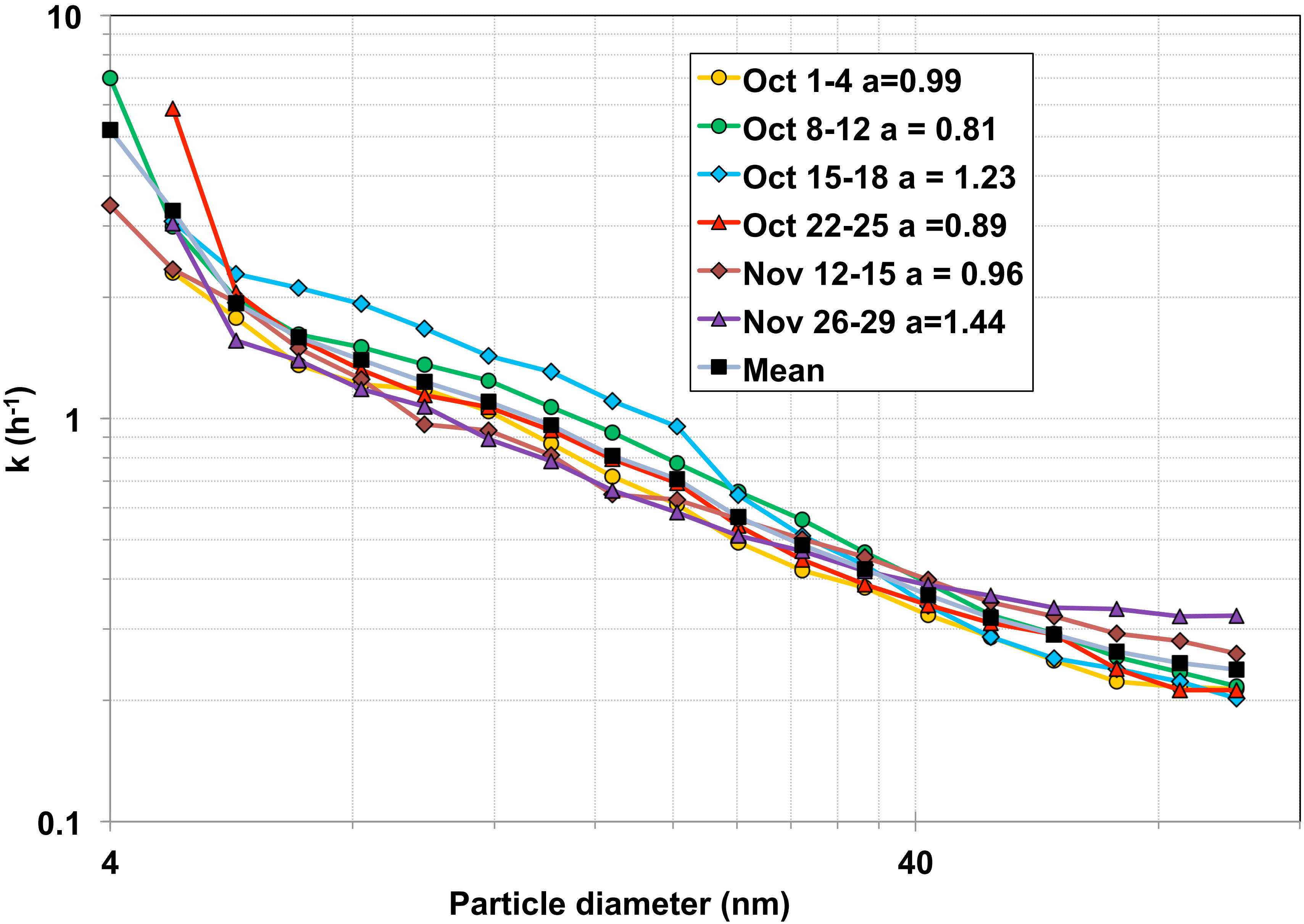
Penetration Coefficient: Closed Windows vs. One Window Open 7.5 cm (3 inches)



Source: Rim, D., Wallace, L., Persily A. (2010) Infiltration of outdoor ultrafine particles into a test house *Environ. Sci. Technol.* **2010,** *44,* 5908–5913

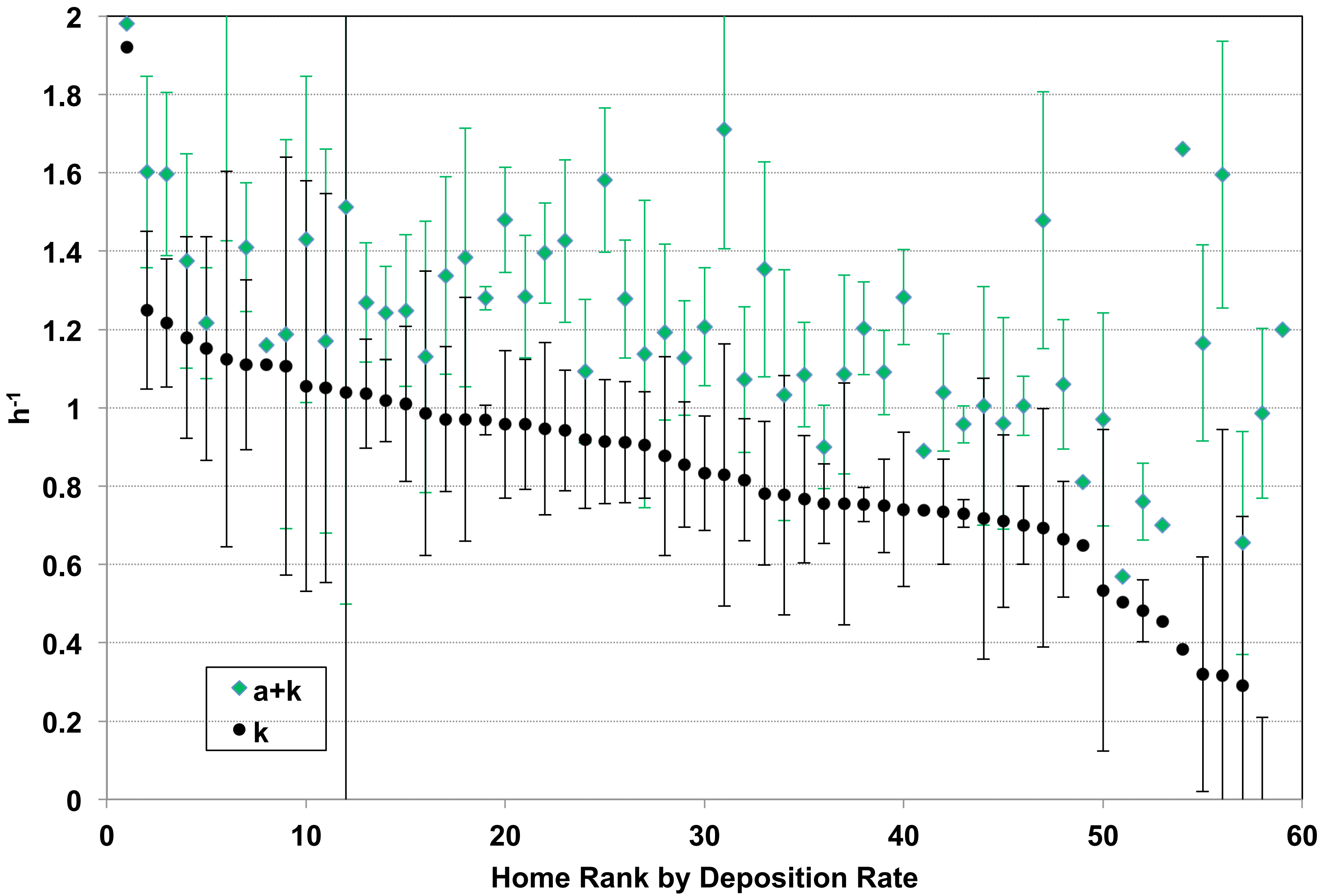
Deposition *k*

- Depends on:
- Surfaces: smooth (*k* small); rough (*k* large)
- Indoor air motion: still (*k* small); fast (k large)
- Filtration: mechanical, electrostatic
- Particle size: governed by Brownian diffusion



Oct 1-4 a=0.99
Oct 8-12 a = 0.81
Oct 15-18 a = 1.23
Oct 22-25 a =0.89
Nov 12-15 a = 0.96
Nov 26-29 a=1.44
Mean
10
1
0.1
k (h-1)
4
40
Particle diameter (nm)

UFP Deposition (*k*) and Decay (*a*+*k*) Rates



Source: Unpublished data—please do not cite or quote

Health Canada Studies

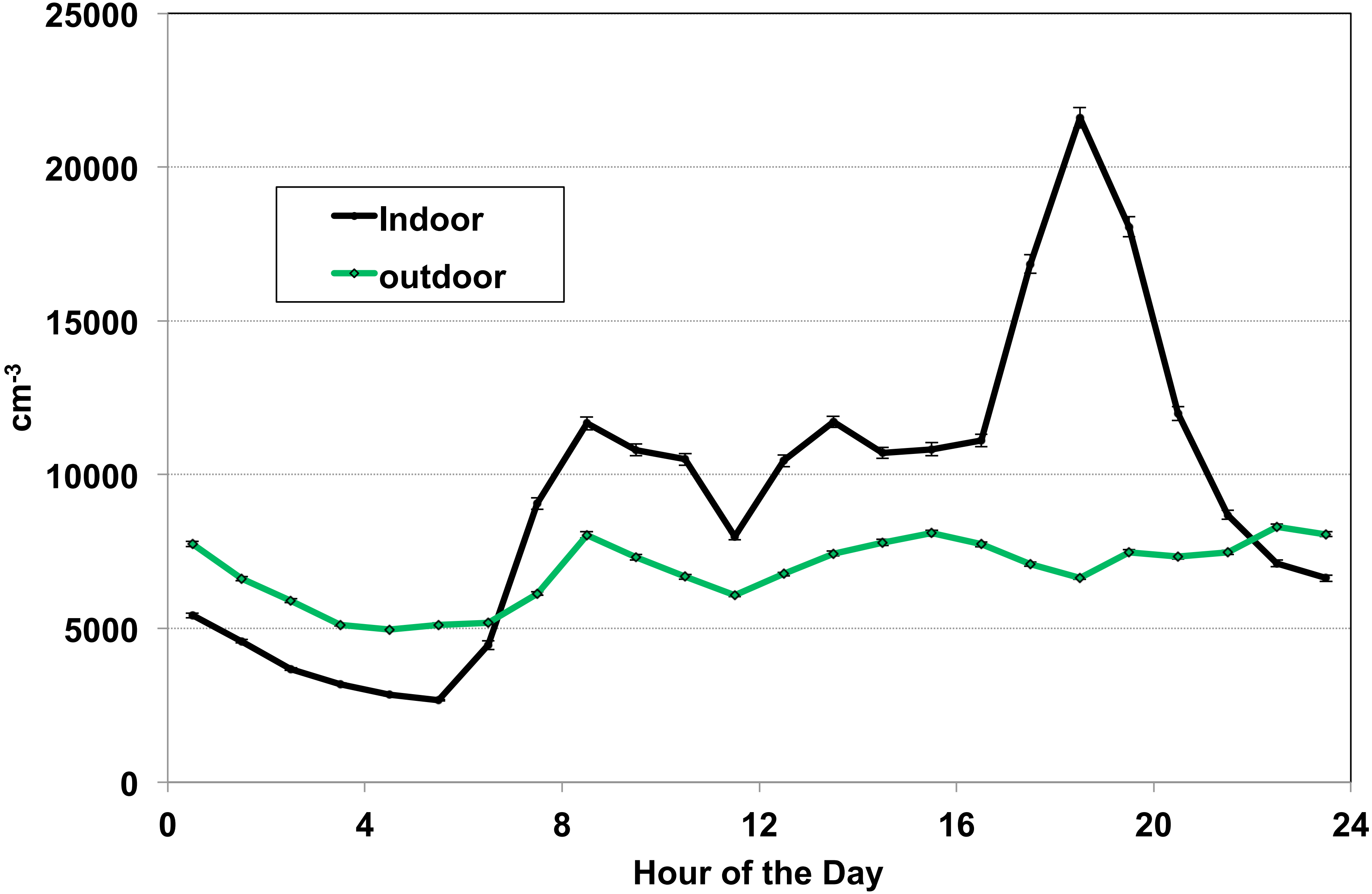
- Multihome studies in several Canadian cities
  - 50-100 homes, 2-4 seasons, one week per season
- Methods
  - TSI P-Trak (20-1000 nm) operating for 10-30 minutes each hour
  - Daily air exchange by perfluorotracer (PFT)
  - Questionnaires to determine sources

Source:

Wheeler, A.J., Wallace, L.A., Kearney, J., Van Ryswyk, K, You, H., Kulka, R., Brook, J.R., and Xu, X. (2011) Personal, indoor, and outdoor concentrations of fine and ultrafine particles using continuous monitors in multiple residences. *Aerosol Science and Technology* 45: 1078-1089.

Kearney, J., Wallace, L., MacNeill, M., Xu, X., VanRyswyk, K., You, H., Kulka, R., Wheeler, A.J. (2011) Residential indoor and outdoor ultrafine particles in Windsor, ON. Accepted by *Atmos Environ*, October, 2010.

Diurnal Variation of Indoor & Ambient UFP



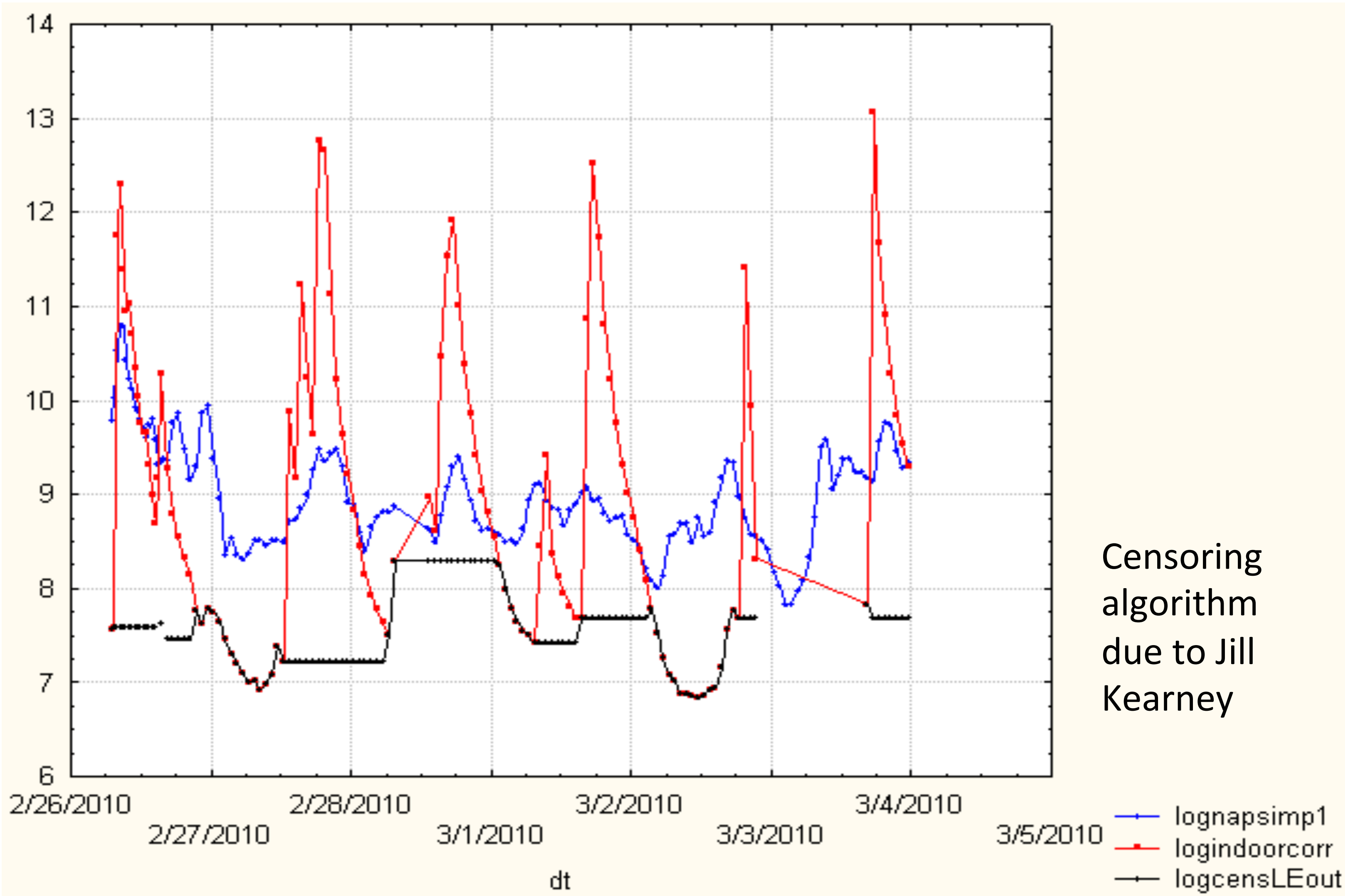
Source: Unpublished data for 50 homes—please do not cite or quote

Calculating the Infiltration Factor F_{inf}

- **Adapt censoring algorithm (Kearney et al., 2010) to detect indoor peaks**
- **Employ recursive mass-balance model to estimate infiltration factors for homes**
- **Calculate outdoor and indoor-generated contributions to total indoor UFP levels**
- **Relate indoor levels to sources (e.g., cooking)**

Source: Kearney, J., Wallace, L., MacNeill, M., Xu, X., VanRyswyk, K., You, H., Kulka, R., Wheeler, A.J. (2010) Residential indoor and outdoor ultrafine particles in Windsor, ON. *Atmos Environment*, ***In Press, Corrected Proof***, *Available online 25 November 2010*. doi:10.1016/j.atmosenv.2010.11.002.

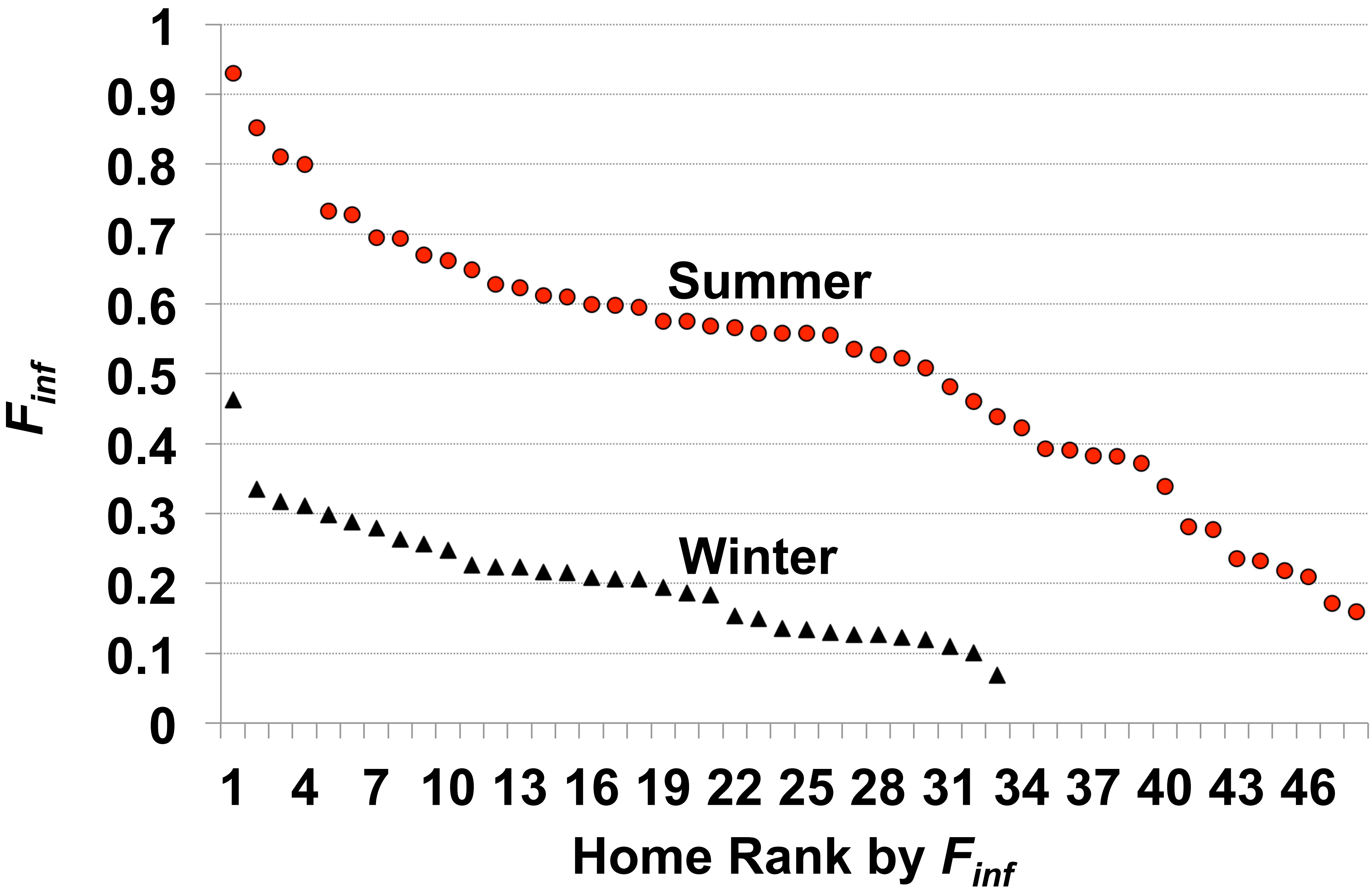
Indoor, Outdoor, & Censored Indoor UFP: Dinnertime Peaks



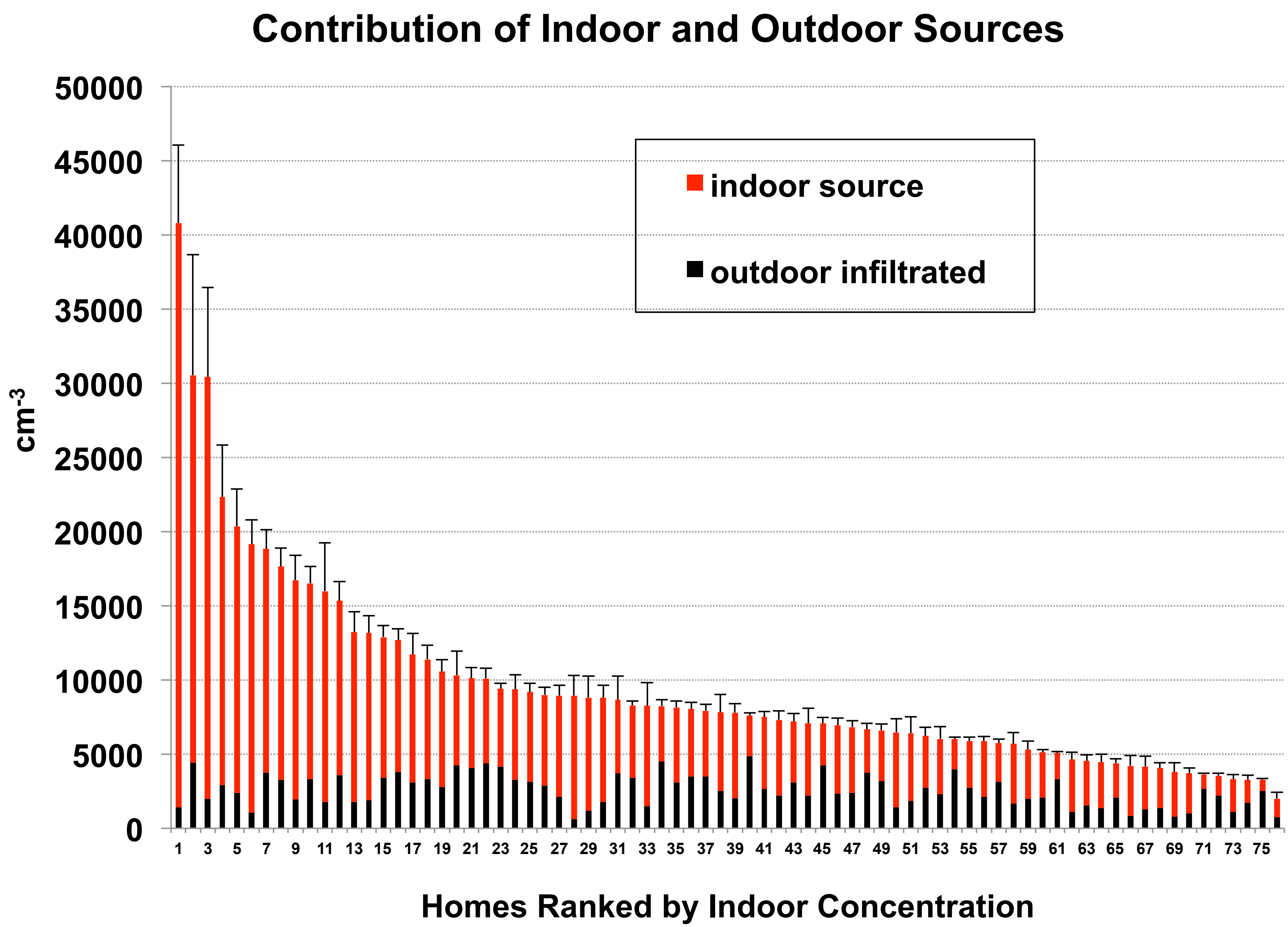
Censoring algorithm due to Jill Kearney

Source: Unpublished data—please do not cite or quote

Seasonal Dependence of UFP Infiltration Factor

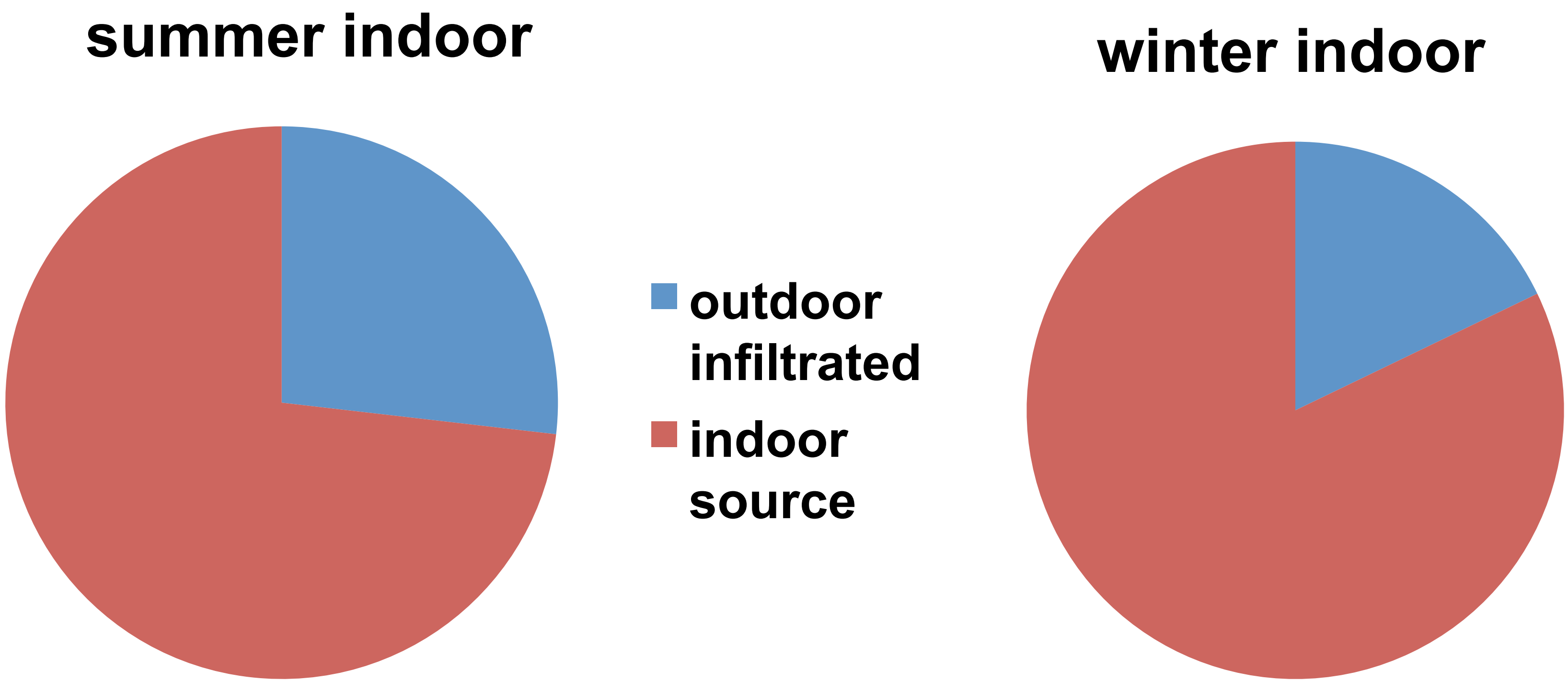


Source: Unpublished data—please do not cite or quote



Source: Unpublished data—please do not cite or quote

Average Contributions from Indoor and Outdoor Sources to Indoor UFP Levels in 74 Canadian Homes



Source: Unpublished data—please do not cite or quote

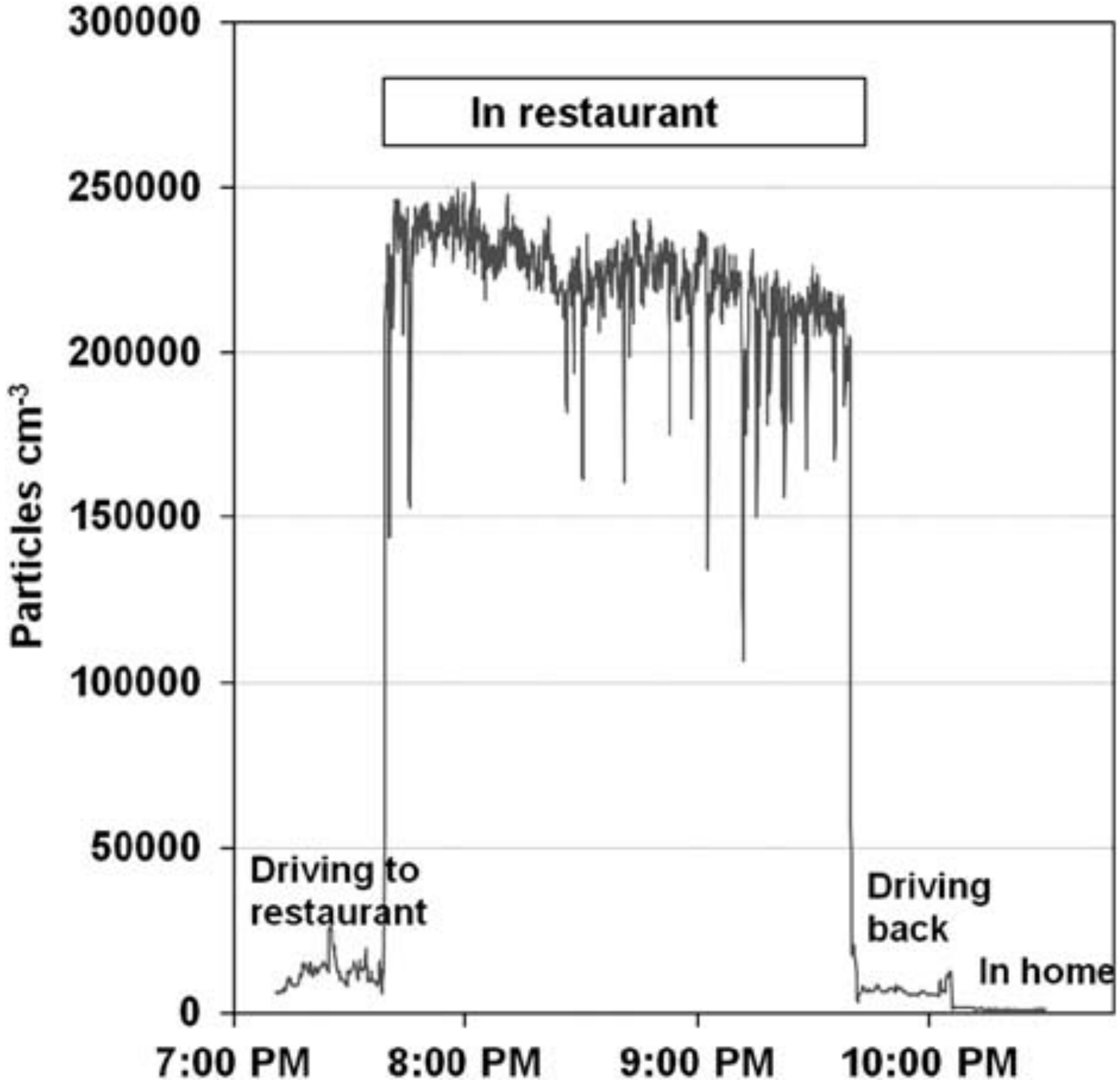
Personal Exposure to UFP

- Indoors (85% of the time)
  - Indoor sources
  - Outdoor infiltration
- Outdoors (6%)
- In vehicles (7%)
- In restaurants (1.8%)

In-Vehicle Exposures

- 29,000-33,000 cm^{-3} (California, East Coast, Montreal)
- Perhaps 3X outdoor concentrations
- 6% of time in vehicles X 3 = 18% of total exposure

Wallace, LA and Ott, WR. (2010). Personal exposure to ultrafine particles.
J Expos Sci & Environ Epidemiol 21:20-30.



Wallace, LA and Ott, WR. (2010). Personal exposure to ultrafine particles. *J Expos Sci & Environ Epidemiol* 21:20-30.

Exposures to UFP in Restaurants

No.	Description	Patron Count	Duration (min)	Volume (m^3)	Mean ($\times 10^3$ cm^{-3})	SD ($\times 10^3$ cm^{-3})	Outdoor Background ($\times 10^3$ cm^{-3})
1	Mexican Restaurant	16	27	174	28	16	7.2
2	Sports Tavern	14	37	639	126	46	10
3	Mexican Restaurant	3	30	211	10	3.2	5.4
4	American Restaurant	75	75	611	22	11	2.1
5	Sports Tavern	15	29	548	88	13	2.2
6	Thai Restaurant	6	67	302	166	43	12
7	Italian Restaurant	25	88	385	25	6.3	10
8	Hotel Restaurant 1	77	69	810	98	23	10
9	Hotel Restaurant	68	35	810	97	33	8.5
10	Hotel Restaurant	23	44	810	58	18	14
11	Seafood Restaurant	53	103	652	110	44	4.3
12	Sandwich-Pizza-Bakery	6	77	--	61	35	9.5
13	Elegant Indian Restaurant	58	69	790	192	39	9.3
14	Seaside Beach Restaurant	112	90	1,422	77	30	7.3
15	Family Tavern	34	128	448	109	51	18
16	Italian Restaurant	41	76	284	37	6.8	3.7
17	Steak House	124	73	610	16	6.3	3.6
18	Sicilian Restaurant	24	60	490	69	4.0	7.6
19	Indian Restaurant	16	75	360	222	27	3.9
20	Seafood Grill	80	104	960	37	7.2	6
21	Chinese restaurant	60	75	1,200	203	43	12
22	French restaurant	30	60	132	228	11	6.3
Mean		43.6	67.8	602.3	94.5	23.5	7.9
SD		34.4	26.5	333.2	68.9	16.0	4.0

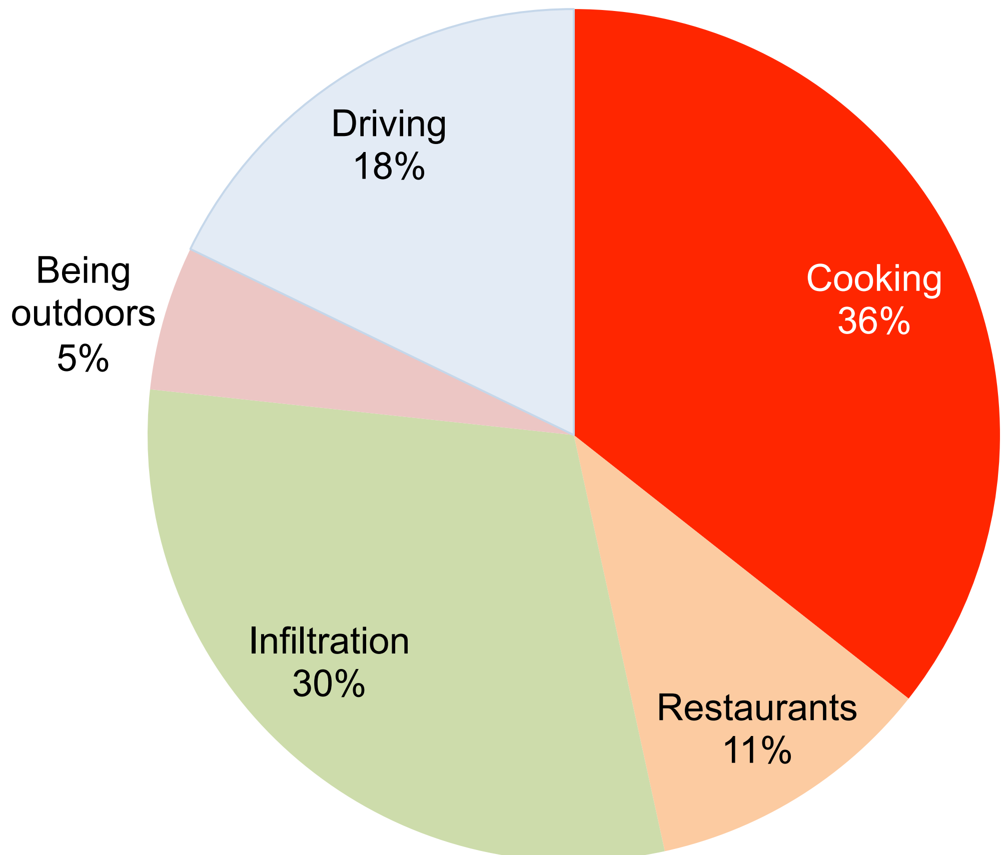
Source: Wallace and Ott, 2010

Estimated UFP Exposure from Major Sources

Location/Activity	Daily duration (h)	Conc (cm^{-3})	Exposure ($h\text{-}cm^{-3}$)	%
Outdoors	1	10000	10000	6
Indoors, no sources	19.4	2700	52000	30
In vehicles	1	30000	30000	17
In restaurants	0.2	90000	18000	10
Indoors, sources	2.4	26000	62400	36
Total	24	7200	172600	100

Source: Wallace and Ott, 2010

Personal UFP Exposure



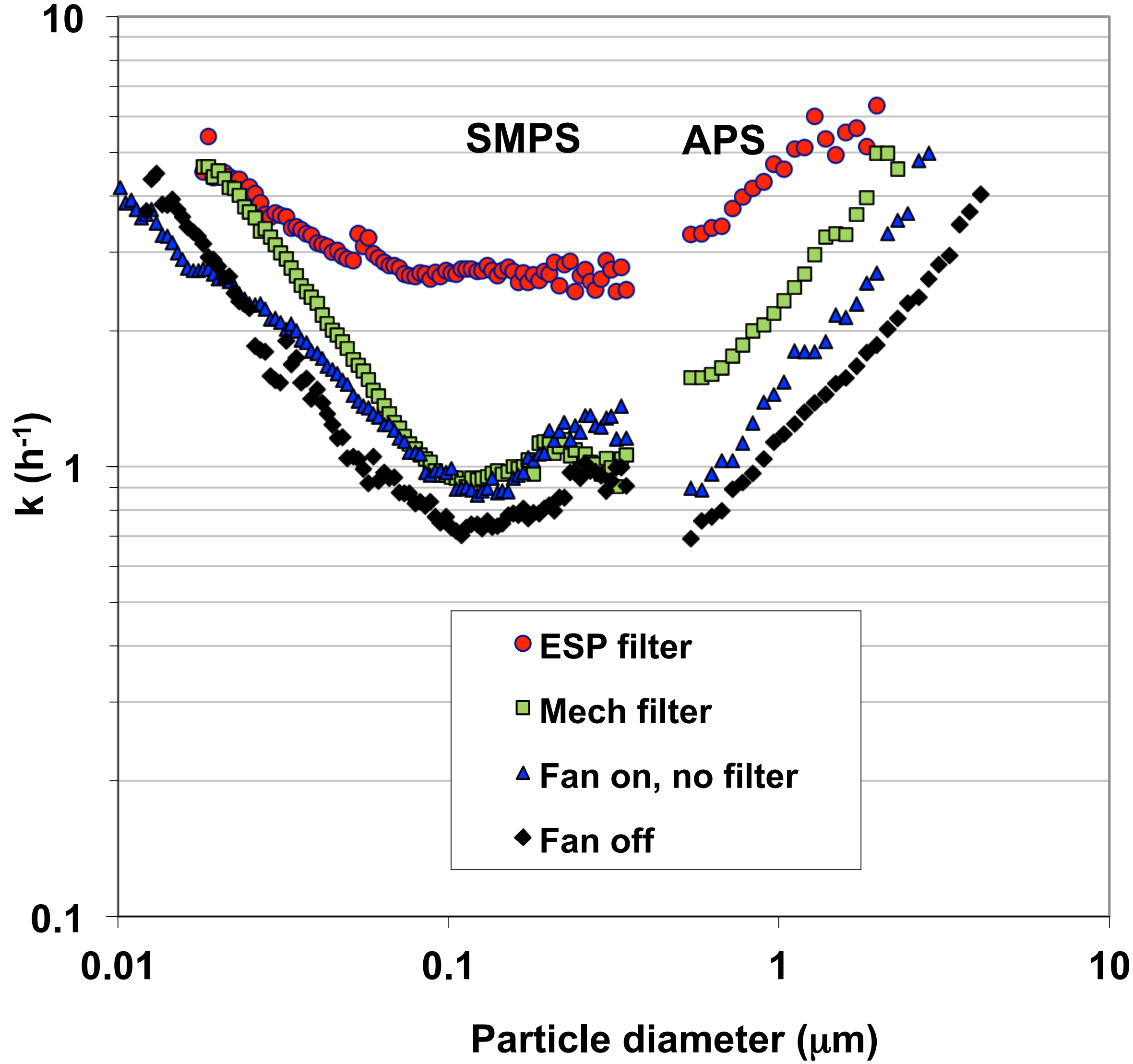
Wallace, LA and Ott, WR. (2010). Personal exposure to ultrafine particles.
J Expos Sci & Environ Epidemiol 21:20-30.

Conclusions: Exposure to UFP

- Less Outdoor Infiltration than FP (25% vs 50%)
- Important Indoor Sources:
  - Cooking (Gas or Electric)
  - Restaurants
- Indoor Contributions Often > Outdoor
- Driving May Contribute 15-20%

Reducing Exposure

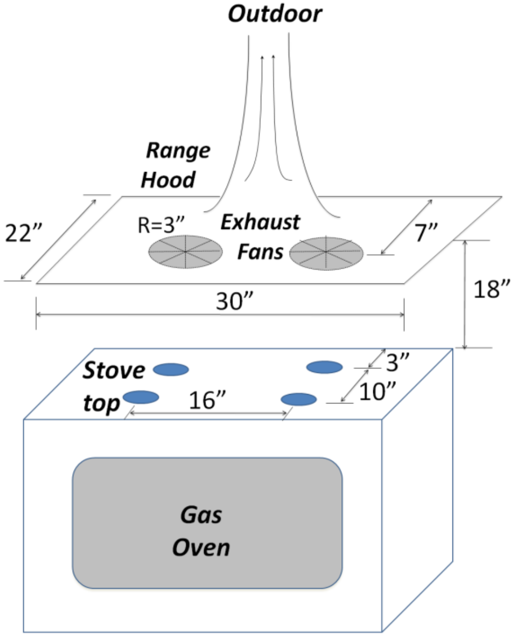
- Air cleaners
  - In-duct Electrostatic Precipitators—best for the larger UFP
  - Membrane filters—may be better for smaller UFP
  - Best to use ESP backed up by membrane filter?
- Kitchen exhaust fans
  - Unclear how efficiently they capture UFP



Source: Wallace, L.A. and Howard-Reed, C.H. Continuous Monitoring of Ultrafine, Fine, and Coarse Particles in a Residence for 18 Months in 1999-2000. *J Air Waste Manage. Assoc.* **52**(7):828-844. 2002.

METHODS

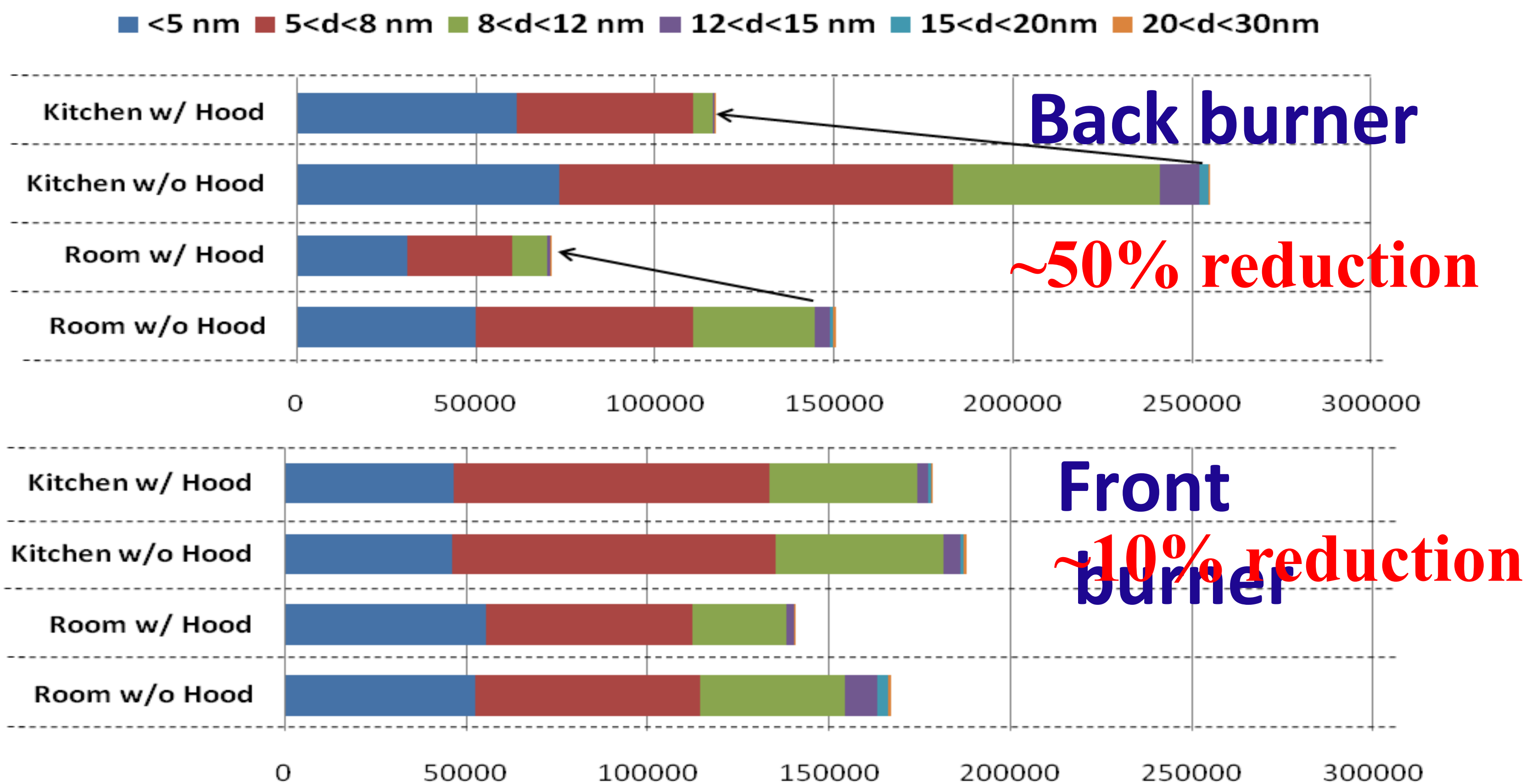
Calculating the efficiency of kitchen exhaust fans



Experimental settings

- ➢Range hood flow rate: 100 m^3/h - 370 m^3/h
- ➢ Front burner vs. back burner
- ➢ Gas oven temperature: 350 F – 450 F
- ➢ Central mixing fan on

Effect of range hood - Size resolved peak concentration (#/cm^3)



Conclusions

- Two Mitigation Methods Suggested
  - Use of Kitchen Fan During and After Cooking
    - Design and Noise Issues
  - Use of In-Duct High-Quality Air Filters (ESP + membrane + activated charcoal for ozone reduction)
    - Cost and Maintenance Issues

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