Supplement of

Source apportionment of $PM_{2.5}$ in Shanghai based on hourly molecular organic markers and other source tracers

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Figure S1. Comparison of reconstructed and measured $PM_{2.5}$ mass for samples collected at Shanghai in 2018.

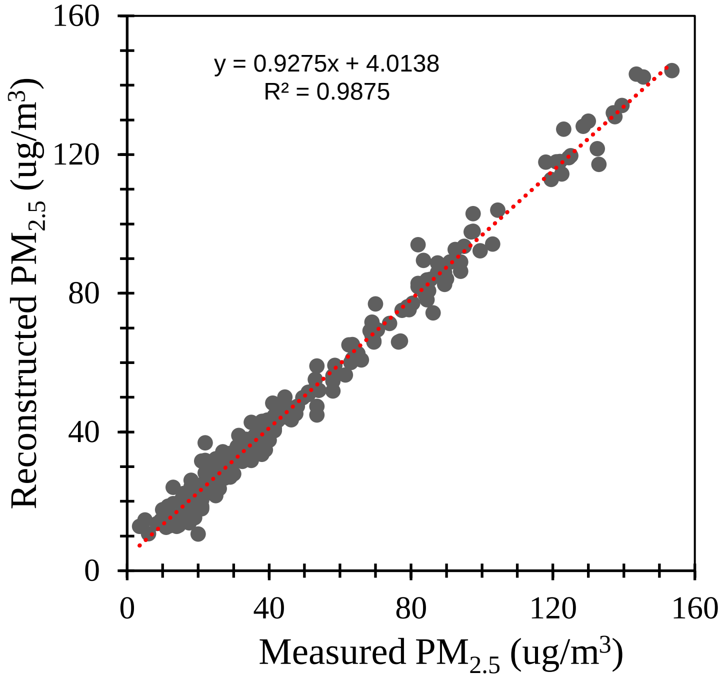


Figure S2. Change of Q/Q_{exp} value from 7 to 14 factors run for MM-PMF.

When the number of factors increases to a certain value, Q/Q_{exp} will change less dramatically. Q/Q_{exp} changed by 6.4% from the 10- to 11-factor model, less significant than the 11.4- 13.1% observed when the number of factors varied from 6 to 10, suggesting the factor number reaching ten was needed for explaining the input data. However, when examining factor profiles, the 11- factor solution provides the most reasonable source profiles by separating the vehicle exhaust from secondary nitrate factors. The 11-factor result of the vehicle emission source is a more accurate analysis of the composition of the atmospheric pollution source in Shanghai, China.

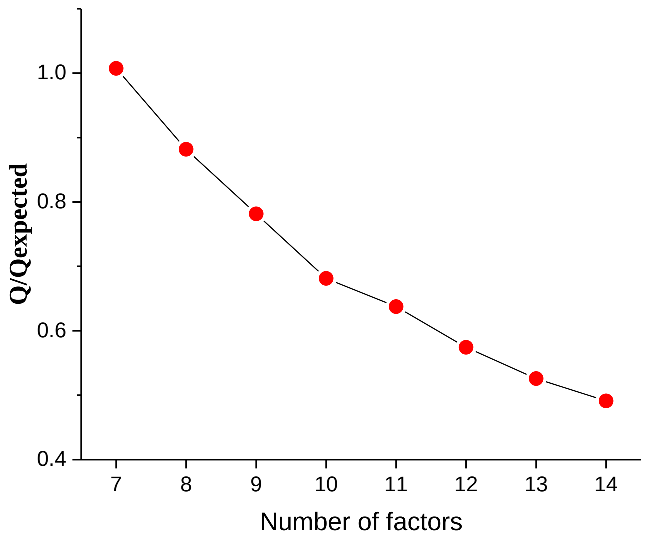


Figure S3. Change of Q/Q_{exp} value from 5 to 11 factors run for PMFt.

When the number of factors increases to a certain value, Q/Q_{exp} will change less dramatically. Q/Q_{exp} changed by 10.3% from the 8- to 9-factor model, less significant than the 14.2- 25.0% observed when the number of factors varied from 5 to 8, suggesting the factor number reaching eight or nine was needed for explaining the input data. Increasing to 9 factors, Fe and Pb are no longer in the Industrial Emission or Coal combustion factor and forms an unexplainable factor. Finally choose the 8-factor result

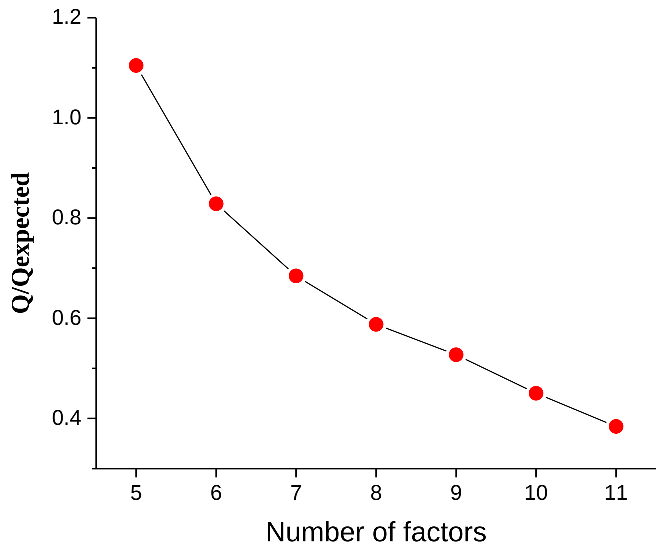


Figure S4. Air mass trajectories during observation.

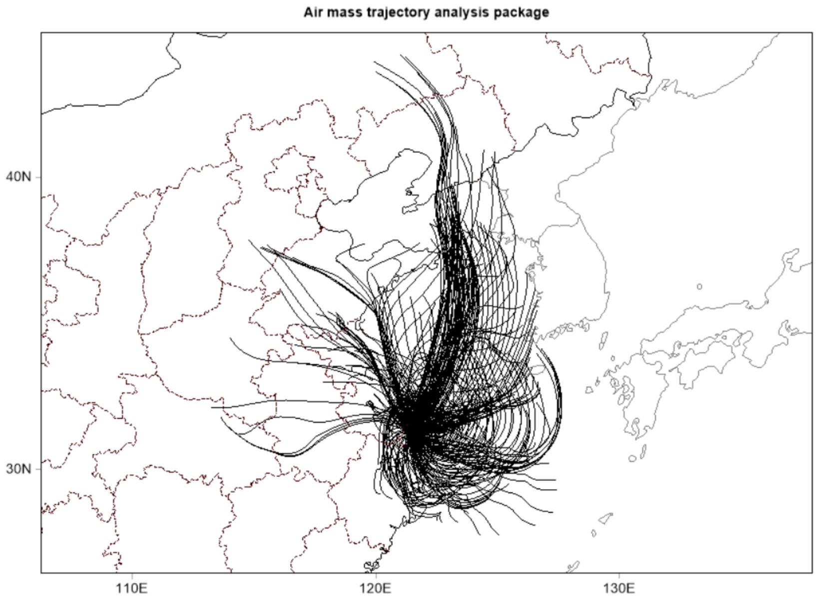
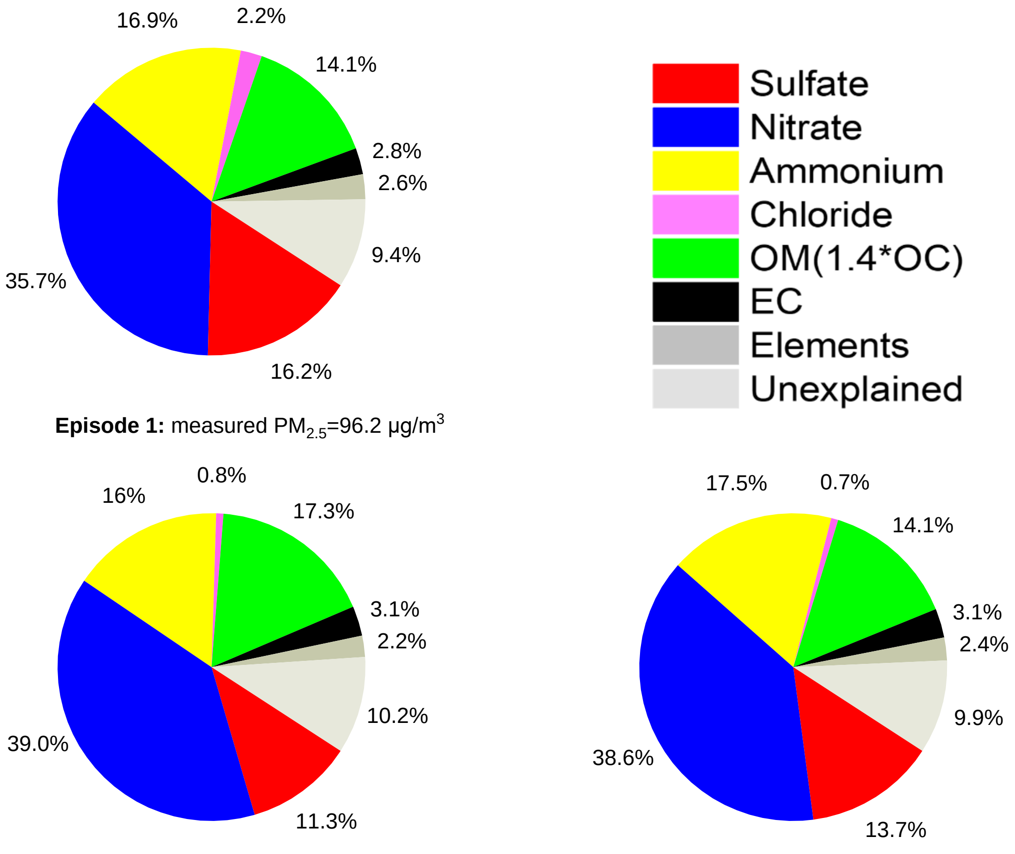


Figure S5. The average concentration of $PM_{2.5}$ and the percentage of components represented by each episode.



Episode 1: measured $PM_{2.5}$=96.2 μg/m^3

Episode 2: measured $PM_{2.5}$=79.8 μg/m^3

Episode 3: measured $PM_{2.5}$=109.1 μg/m^3

Figure S6. Resolved factor profiles (left) and factor contributions (right) in eight-factor solution in PMFt.

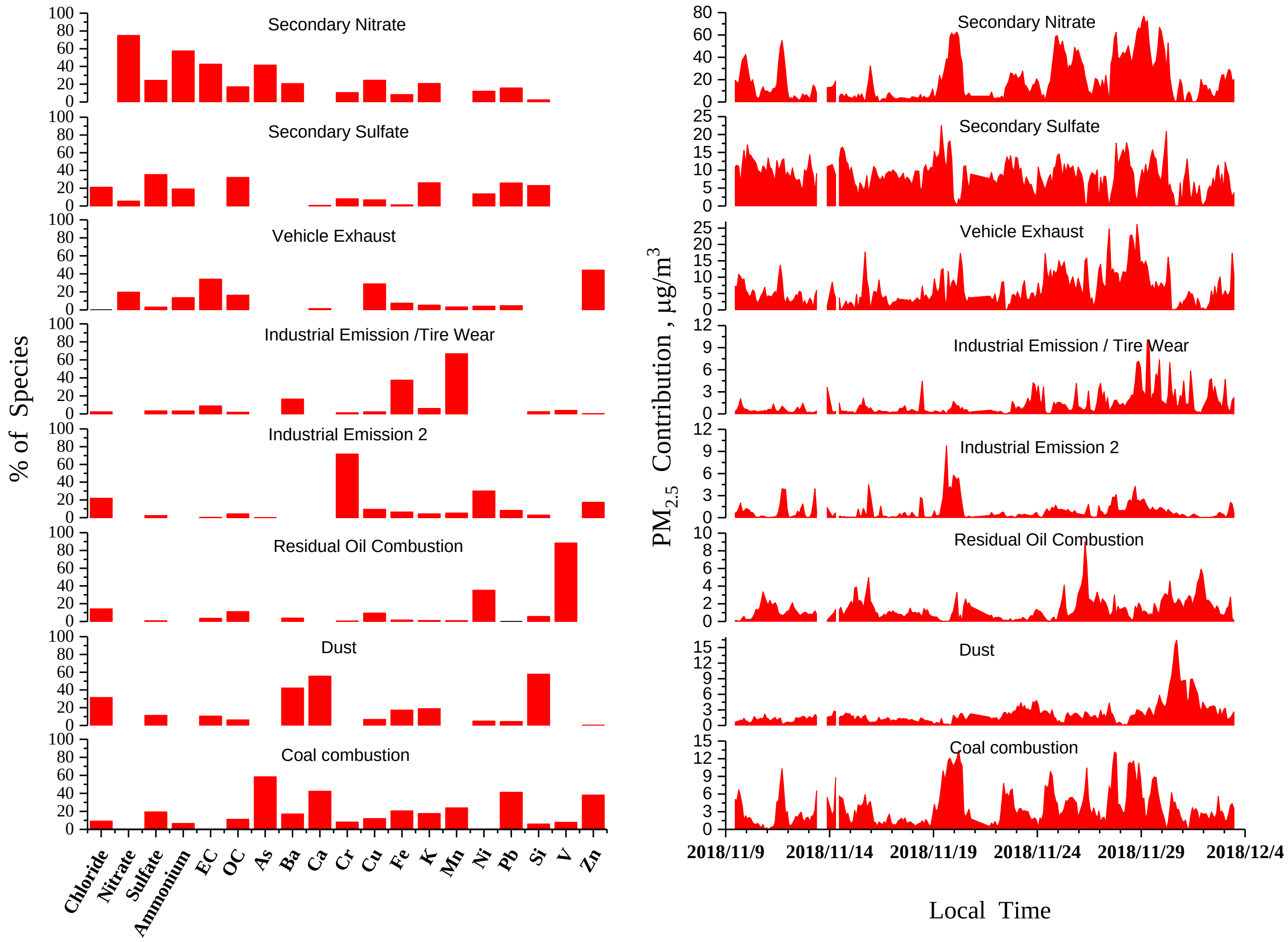


Table S1. Summary of error estimation diagnostics from BS, DISP and BS-DISP for MM-PMF.

BS Mapping(R⩾0.6)		Secondary Nitrate	Secondary Sulfate	Vehicle Exhaust	Industrial / Tire Wear	Industrial Emission 2	Residual Oil Combustion	Dust	Coal combustion	Biomass Burning	Cooking	SOA	Unmapped
Secondary Nitrate		97	1	0	0	0	0	0	1	1	0	0	0
Secondary Sulfate		5	90	0	1	0	0	0	1	1	0	1	1
Vehicle Exhaust		20	1	61	0	0	0	1	3	5	0	2	7
Industrial / Tire Wear		0	0	0	100	0	0	0	0	0	0	0	0
Industrial Emission 2		0	0	0	0	100	0	0	0	0	0	0	0
Residual Oil Combustion		0	0	0	0	0	100	0	0	0	0	0	0
Dust		1	1	0	1	0	0	94	1	2	0	0	0
Coal combustion		5	0	0	0	0	0	0	93	1	1	0	0
Biomass Burning		0	0	0	0	0	0	0	2	95	0	0	3
Cooking		0	0	0	0	0	0	0	0	0	95	2	3
SOA		6	1	0	0	0	0	0	1	0	0	90	2
DISP Diagnostics		Error Code: 0					Largest Decrease in Q: -16.3 (-0.7%)						
Factor Swaps	dQ^{max}=4	0	0	0	0	0	0	0	0	0	0	0	
	dQ^{max}=8	0	0	0	0	0	0	0	0	0	0	0	
	dQ^{max}=15	0	0	0	0	0	0	0	0	0	0	0	
	dQ^{max}=25	0	0	0	0	0	0	0	0	0	0	0	
BS-DISP Diagnostics			# of runs accepted: 49 out of 101					Largest Decrease in Q: -10.5 (-0.4%)					
# of Decreases in Q: 0				# of Swaps in Best Fit: 32					# of Swaps in DISP: 20				
Factor Swaps	dQ^{max}=0.5	12	1	1	9	26	2	2	10	20	6	15	
	dQ^{max}=1	12	2	1	11	26	2	2	11	20	6	15	
	dQ^{max}=2	13	2	2	13	30	2	4	12	22	6	15	
	dQ^{max}=4	18	3	4	16	35	2	4	17	23	6	15	

Table S2. Summary of error estimation diagnostics from BS, DISP and BS-DISP for PMFt.

BS Mapping(R⩾0.6)	Secondary Nitrate	Secondary Sulfate	Vehicle Exhaust	Industrial /Tire Wear	Industrial Emission 2	Residual Oil Combustion	Dust	Coal combustion	Unmapped
Secondary Nitrate	100	0	0	0	0	0	0	0	0
Secondary Sulfate	1	93	0	1	0	0	1	1	3
Vehicle Exhaust	11	0	73	5	0	0	0	3	8
Industrial / Tire Wear	0	0	0	100	0	0	0	0	0
Industrial Emission 2	0	0	0	0	100	0	0	0	0
Residual Oil Combustion	0	0	0	0	0	100	0	0	0
Dust	0	0	1	1	0	0	97	1	0
Coal combustion	2	1	0	1	0	0	0	96	0
DISP Diagnostics	Error Code: 0				Largest Decrease in Q: -17.6 (-1.3%)				
Factor Swaps dQ^{max}=4	0	0	0	0	0	0	0	0	
Factor Swaps dQ^{max}=8	0	0	0	0	0	0	0	0	
Factor Swaps dQ^{max}=15	0	0	0	0	0	0	0	0	
Factor Swaps dQ^{max}=25	0	0	0	0	0	0	0	0	
BS-DISP Diagnostics	# of runs accepted: 52 out of 101				Largest Decrease in Q: -2.3 (-0.2%)				
# of Decreases in Q: 0		# of Swaps in Best Fit: 30				# of Swaps in DISP: 19			
Factor Swaps dQ^{max}=0.5	0	18	5	23	36	22	7	22	
Factor Swaps dQ^{max}=1	0	19	9	24	40	22	8	23	
Factor Swaps dQ^{max}=2	0	22	14	25	48	22	9	25	
Factor Swaps dQ^{max}=4	1	25	18	27	60	22	10	31	

Table S3. The MM-PMF input data statistics for the modelled chemical compounds and base model diagnostics obtained for the 11-factor solution.

Species	R^2	Intercept	Slope
Chloride	0.39	0.29	0.42
Nitrate	0.86	0.86	0.84
Sulfate	0.73	1.04	0.85
Ammonium	0.88	0.43	0.91
EC	0.86	-0.02	0.95
OC	0.86	-0.11	0.99
As	0.98	0.01	0.99
Ba	0.97	-0.01	0.99
Ca	0.95	0.01	0.93
Cr	0.95	0.01	0.92
Cu	0.88	0.01	0.86
Fe	0.42	0.22	0.32
K	0.95	0.01	0.99
Mn	0.98	0.01	0.97
Ni	0.91	0.01	0.85
Pb	0.54	0.02	0.42
Si	0.98	0.04	0.89
V	0.98	0.01	0.97
Zn	0.81	0.02	0.74
PAHs252	0.93	0.01	0.79
PAHs276	0.92	0.01	0.82
$C_{6\sim8}$DICAs	0.78	0.02	0.65
OHBAs	0.64	0.01	0.49
SFAs	0.81	0.02	0.62
phthalic acid	0.51	0.01	0.38
Mannosan	0.91	0.01	0.83
Levoglucosan	0.88	0.01	0.93
α-pinT	0.72	-0.01	0.88
DHOPA	0.62	0.01	0.66
C_9-acids	0.84	0.02	0.82

Table S4. Pearson correlation (R) of MM-PMF factor contributions with corresponding tracer species, meteorological parameters (wind speed [WS], temperature [T], and relative humidity [RH]), and gas concentrations (SO_2, CO, and NOx).

PMF Factor	Secondary Nitrate	Secondary Sulfate	Vehicle Exhaust	Industrial Emission /Tire Wear	Industrial Emission 2	Residual Oil Combustion	Dust	Coal combustion	Biomass Burning	Cooking	SOA
WS(m/s)	-0.52	0.16	-0.12	-0.34	-0.06	-0.08	-0.10	-0.23	-0.21	-0.15	-0.39
T(℃)	-0.18	-0.22	-0.09	0.08	-0.18	0.39	0.26	-0.08	-0.18	-0.12	-0.29
RH(%)	0.14	-0.13	-0.18	-0.01	0.05	0.19	-0.26	-0.23	0.19	0.33	0.05
SO_2	0.59	0.32	0.46	0.52	0.51	-0.05	-0.18	0.59	0.58	0.01	0.67
CO	0.79	0.36	0.58	0.35	0.59	-0.03	-0.17	0.56	0.69	0.11	0.77
NOx	0.54	-0.19	0.67	0.49	0.31	0.30	0.10	0.33	0.50	0.15	0.43

Table S5. The average concentration of $PM_{2.5}$ and the percentage of components under different air mass clusters.

Component	Cluster-1		Cluster-2		Cluster-3		Cluster-4	
	Concentration, ug/m^3	Percentage, %	Concentration, ug/m^3	Percentage, %	Concentration, ug/m^3	Percentage, %	Concentration, ug/m^3	Percentage, %
SO_4^{2-}	10.1	16.8%	9.6	14.5%	5.1	15.2%	4.6	23.2%
NO_3^-	20.7	34.5%	24.1	35.5%	8.4	25.0%	3.1	15.7%
NH4+	10.1	16.8%	11.3	16.6%	4.6	13.6%	2.7	13.5%
Cl^-	1.1	1.7%	0.8	1.2%	0.8	2.4%	0.6	3.2%
OM (1.4*OC)	9.5	15.8%	11.2	16.4%	8.4	24.9%	6.1	30.3%
EC	1.6	2.7%	2.3	3.4%	1.4	4.1%	0.6	3.0%
Elements	1.6	2.6%	2.2	3.2%	1.2	3.5%	1.2	5.8%
Unexplained	5.4	9.1%	6.3	9.2%	3.8	11.4%	1.1	5.3%
$PM_{2.5}$	60.0	/	64.4	/	32.9	/	19.8	/

Cluster 1 and 4 with longer trajectory length, representing long range transport air masses; clusters 2 and 3 with shorter trajectory length, indicating local or msdian range transport air masses. Cluster 1 represents the air mass transport from Hebei, Shandong and Henan Provinces in the central region, and the average concentration of $PM_{2.5}$ reached 60.0 μg/m3. Cluster 2 represents the local transport in Shanghai and neighbouring areas. The average $PM_{2.5}$ concentration was the highest among the four clusters, which was 64.4 μg/m3. Cluster 3 is a shorter distance transport of marine air masses in the eastern sea area of Shanghai, with a low $PM_{2.5}$ average of 32.9 μg/m^3. Cluster 4 represents long-distance air mass transport from the northern seas of China and the north-eastern regions of Liaoning province and Inner Mongolia. The $PM_{2.5}$ concentration was the lowest among the four clusters, only 19.8 μg/m^3.

Table S6. The PMFt input data statistics for the modelled chemical compounds and base model diagnostics obtained for the 8-factor solution.

Species	R^2	Intercept	Slope
Chloride	0.18	0.41	0.37
Nitrate	0.84	1.49	0.79
Sulfate	0.78	1.22	0.78
Ammonium	0.84	0.59	0.85
EC	0.85	-0.04	0.94
OC	0.84	0.22	0.93
As	0.98	-0.01	1.01
Ba	0.98	0	1.02
Ca	0.89	0.01	0.88
Cr	0.94	0.01	0.91
Cu	0.84	0.01	0.81
Fe	0.99	0.03	0.91
K	0.95	0.01	0.96
Mn	0.98	0.01	0.97
Ni	0.91	0.01	0.83
Pb	0.98	0	0.98
Si	0.98	0.04	0.88
V	0.99	0	0.99
Zn	0.81	0.01	0.75

Table S7: Concentrations(μg/m³) of reconstructed $PM_{2.5}$ and OC from MM-PMF and PMFt.

Factor	MM-PMF		PMFt	
	$PM_{2.5}$	OC	$PM_{2.5}$	OC
SOA	6.71	0.25		
Secondary Nitrate	15.25	1.31	19.71	1.09
Secondary Sulfate	7.10	1.36	8.82	2.05
Vehicle Exhaust	5.13	1.05	6.43	1.04
Industrial Emission / Tire Wear	1.36	0.17	1.32	0.17
Industrial Emission 2	0.46	0.05	0.91	0.21
Residual Oil Combustion	0.82	0.69	1.40	0.83
Dust	2.27	0.09	2.26	0.24
Coal combustion	3.07	0.46	3.75	0.71
Biomass Burning	2.24	0.37		
Cooking	1.17	0.69		