Settling Flux of PAHs on the Ground Surface

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Abstract

Three methods (bucket, downward and upward surface plate, and water vessel) for measuring dry deposition were used at the three different heights (0.5 m, 1.5 m and 12.5 m) to estimate the real settling fluxes of mass and Polycyclic Aromatic Hydrocarbons (PAHs) in TERC (Tsukuba), Japan. Ambient air samples were also collected there using a high-volume sampler. Particle size distributions and composition of PAHs were measured by the low pressure cascade impactor (LP-20) at NIES which is located about 5 km south of TERC. Settling velocities were calculated as the settling fluxes divided by corresponding average ambient air concentration. The results showed that small particles dominate atmospheric dry deposition of PAHs and that bucket deposition fluxes and net fluxes decrease with height due to re-suspension and/or vertical distribution of suspended particles etc. The results also indicated that downward fluxes overestimated and net fluxes underestimated the real fluxes for both mass and PAHs. To estimate the real fluxes, we can assume that net flux is the first approximation.

Keywords: Polycyclic aromatic hydrocarbons; Atmospheric deposition; Net flux; Settling velocity; Size distribution

Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) are organic compounds, containing more than one benzene ring. Sixteen PAH congeners have been classified as priority pollutants by Environmental Protection Agency (EPA), USA due to their carcinogenic property. Interest in atmospheric deposition has increased over the past decade because deposition is an important pathway for the transfer of pollutants from the atmosphere to natural surfaces. Dasch (1985) found that bucket collected more dry deposited material than Teflon, foil, or coated foil surfaces. Previous attempts to quantify PAHs dry depositions have found that there is still no generally acceptable technology for sampling and analyzing dry deposition flux. Therefore, till now, there has been insufficient data to reliably estimate and understand PAH movement in the environment. Recently, a greased, smooth surface was successfully used to measure particulate fluxes of organic and inorganic air pollutants (Yi *et al*., 1997; Tasdemir, 1997; Franz *et al*., 1998; Cakan,

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1999; Shahin *et al*., 1999; Shahin *et al*., 2000; Yi *et al*., 2001; Vadar *et al*., 2002). Since this surrogate surface does not significantly disturb airflow, it gives estimates of the lower limits of dry deposition compared with rougher natural surfaces.

Various surrogate surfaces have been used in the past with the deposition flux shown to be dependent on the shape and properties of the surface (Davidson *et al*., 1985; Vandenberg and knoerr, 1985). Surrogate surfaces are often used because they are relatively inexpensive and do not possess many of the problems associated with the biological surfaces. However, no surface has been established as a standard. The present collection surface was patterned after those used in wind tunnel studies (McCready, 1986) and was designed to provide minimum air flow disruption and thus provide an estimation of the lower limit for dry deposition flux (Davidson *et al*., 1985; Vandenberg and knoerr, 1985).

A comparison of various dry deposition surfaces has shown that the deposition flux is a function of the collector surface geometry and that a smooth surface provides the lowest deposition flux. The flux plate was designed so that it could simultaneously collect particles on the upward and downward surface plate. This provides information on both particle deposition and suspension. Particles collected on the deposition upward and downward surface plates are used to calculate net flux (downward flux - upward flux).

The objectives of this study were:

1. To compare different settling methods to estimate mass and PAHs fluxes at the three different heights.
2. To estimate real fluxes of mass and PAHs on the ground surface.

Experimental section

2.1. Deposition plate

The dry deposition flux was directly measured by using smooth surface plastic plates (0.45 m $\times$ 0.3 m $\times$ 5 mm) of which both surfaces were covered with aluminum sheets (0.3 m $\times$ 0.2 m) coated with approximately 15 mg silicon grease. The plates were fixed to the horizontal bars at the three different heights. We call the flux measured by the upper sheet the downward flux and that measured by the lower one the upward flux. This hydrophobic grease has a high molecular weight and low vapor pressure and is therefore suitable to measure PAH flux and it is suitable for measurement of particulate matter and PAHs. Before sample collection, the grease was sprayed onto the aluminum sheets. Sheets were then put in an oven at 50 for 90 minutes to remove the volatile substances. The sheets were weighed before and after sampling to determine total mass of the particles collected. The greased sheets were then extracted and analyzed for PAHs. Analysis of unexposed aluminum greased sheets (blank) showed that their PAH mass was below the detection limits of this study.

The following features were considered to be applicable to the smooth surface plate for measuring the deposition fluxes under study:

a. A smooth surface produces minimum deposition velocities for surrogate collectors under atmospheric conditions (Davidson *et al*. 1985).

b. An aerodynamic boundary layer that developed over a smooth, flat, acrylic plate with a sharp leading edge in wind tunnel studies (McCready, 1986) is present.

c. Particles are deposited on the surrogate surface plate due to interactions between the particles and the turbulent motion of the atmosphere if the particles are in the inertial deposition range.

d. The greased surfaces prevent particle bounce (Holsen *et al*. 1991).

2.2. Sampling program

Experimental site is located at Terrestrial Environment Research Centre, University of Tsukuba, Tsukuba, Japan (36° 06'35" N, 140° 6'00" E and an altitude of 27 m).

Experiments were conducted at three different heights *i.e.* 0.5 m, 1.5 m and 12.5 m, using three different settling methods: (1) downward and upward surface plates covered by aluminum sheet (exposed surface area 0.06 m^2) coated with silicon grease, (2) bucket (exposed area 0.046 m^2) and (3) water vessel (exposed area 0.039 m^2).

Deposition fluxes by water vessel method were measured only at 1.5 m height. Simultaneously, air samples were also collected at TERC building roof (about 7 m height) using a high volume sampler. For particle size distribution, air samples were collected using a low pressure cascade impactor (LP-20) on the building roof (about 3 m height) in NIES campus which is located about 5 km south of TERC. The sampling apparatus of dry deposition are shown in Fig. 1.

Simultaneous dry deposition and ambient air samples were collected between July and December, 2002 at TERC and particle size distribution air samples were collected at NIES during November, 2002. During periods of no rain, the dry deposition plates allow adequate material to be deposited for the PAH analysis. Each set of dry deposition samples was typically exposedfor 2-5 for days; each high-volume sample was collected for 24 hrs. Air samples for measuring particle size distribution were collected for 3 days. Wind speed (ms^{-1}) was measured at 1.5 m height during the study period.
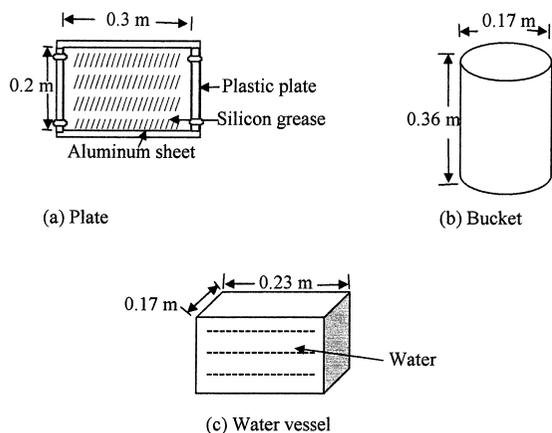

Fig. 1 Different sampling apparatus of atmospheric deposition

2.3. PAHs extraction and analysis

After collecting deposited samples, the silicon greased aluminum sheets and glass fiber filter papers were dried for two days at room temperature and subsequently put into a 50 ml glass bottle and extracted with dichloromethane in an ultrasonic water bath for analysis of particulate PAHs. Extracts were concentrated with N$_2$ before GC/MS analysis. For analysis of solvent PAHs, the eluent was filtered through SepPak tC 18 and PAHs were trapped in it. Entrapped PAHs were extracted with dichloromethane, and the elution was also concentrated with N$_2$. PAHs were analyzed with a GC/MS (Shimadzu). In this study, 16 PAH congeners were analyzed and their abbreviations, molecular weight (MW), chemical formula and structure are shown in Table 1.

Results

3.1. Settling fluxes of mass and PAHs

Mass fluxes decrease with height. At 1.5 m, settling fluxes of bucket, downward surface plate, water vessel, and upward surface plate decrease in this order: bucket > downward plate > water vessel > upward plate (Fig. 2). Net fluxes (downward minus upward) of mass which were nearly the same magnitude as upward fluxes were very low. Similarly, Fig. 3 shows that PAH fluxes decrease with height except the flux of the bucket at 0.5 m and at 1.5 m, and that settling fluxes of water vessel, bucket, downward surface plate, and upward surface plate decrease in this orde (Fig. 3). PAH fluxes of bucket were about four times higher than net flux. These net fluxes were very small because PAH fluxes difference between downward and upward surface plate were very low.

Settling fluxes of mass measured by the previous studies were reported as 175 mgm^{-2}d^{-1} by Noll *et al*. (1989), 193 mgm^{-2}d^{-1} by Holsen and Noll (1992), 163 mgm^{-2}d^{-1} by Lin *et al*. (1993) and 134 mgm^{-2}d^{-1} by Sheu *et al*. (1996). The settling fluxes of mass measured in this study were about one seventh to one fourth of the

Table 1 Measured PAHs and their abbreviations, molecular weight (MW), chemical formula and structure

| PAH	Abbreviation	MW	Formula	Structure
Acenaphthylene	ACY	152.2	$C_{12}H_8$	
Acenaphthene	ACE	154.2	$C_{12}H_{10}$	
Fluorene	FLU	166.2	$C_{13}H_{10}$	
Phenanthrene	PHEN	178.2	$C_{14}H_{10}$	
Anthracene	ANTH	178.2	$C_{16}H_{10}$	
Fluoranthene	FLUR	202.3	$C_{16}H_{10}$	
Pyrene	PYR	202.3	$C_{16}H_{10}$	
Benz[a]anthracene	B(a)A	228.3	$C_{18}H_{12}$	
Chrysene	CHR	228.3	$C_{18}H_{12}$	
Benzo[b]fluoranthene	B(b)F	252.3	$C_{20}H_{12}$	
Benzo[k]fluoranthene	B(k)F	252.3	$C_{20}H_{12}$	
Benzo[e]pyrene	B(e)P	252.3	$C_{20}H_{12}$	
Benzo[a]pyrene	B(a)P	252.3	$C_{20}H_{12}$	
Benzo[g,h,i]perylene	B(g,h,i)P	276.3	$C_{22}H_{12}$	
Indeno[1,2,3-cd]pyrene	I(1,2,3-cd)P	276.3	$C_{22}H_{12}$	
Dibenz[a,h]anthracene	DB(a,h)A	278.4	$C_{22}H_{14}$	
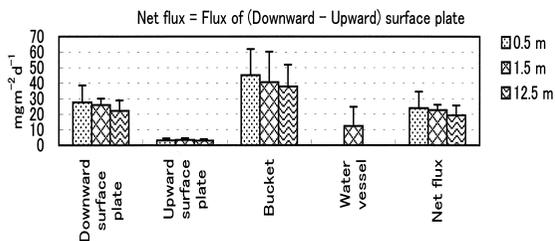
Fig. 2 Averaged mass flux ($mgm^{-2}d^{-1}$) at TERC, Tsukuba
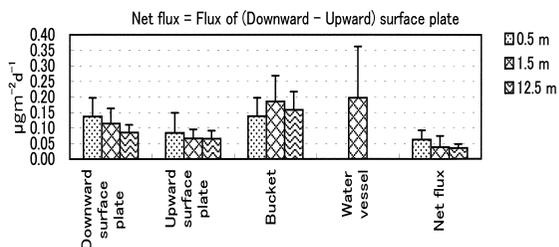
Fig. 3 Averaged 16 PAH congeners ($\mu gm^{-2}d^{-1}$) at TERC, Tsukuba
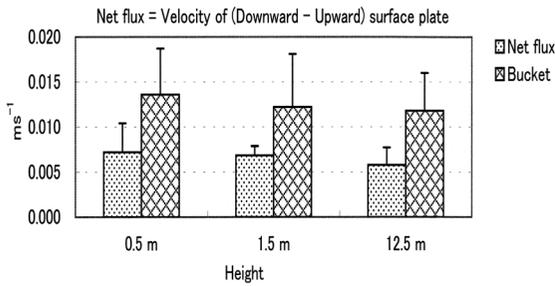
Fig. 4 Average settling velocity (ms^{-1}) of particulate matter at TERC, Tsukuba
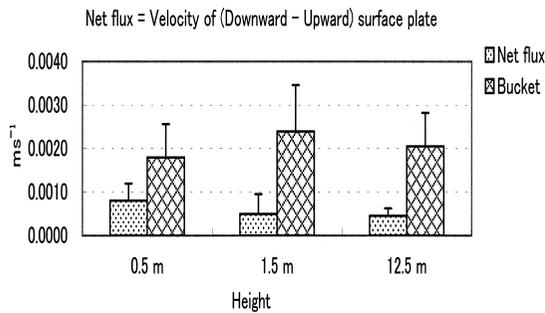

Fig. 5 Average settling velocity (ms^{-1}) of PAHs (16 PAH congeners) at TERC, Tsukuba

value measured by the previous studies because these studies involved urban areas.

Settling velocities for mass and PAHs fluxes were calculated by dividing the deposition flux by the ambient particle concentration. Settling velocity of mass and PAHs fluxes were nearly equal at different heights (Fig. 4 and Fig. 5) because no significant difference was observed for the settling velocity at the different heights. Fig. 4 shows that settling velocity of mass measured by bucket was about two times higher than that of net flux. Fig. 5 shows that settling velocity of PAHs measured by bucket was about 2 ~ 5 times higher than that of net flux. The calculated settling velocities for PAHs flux in this study were one-tenth to one-fourth of the value measured by the previous studies reported as 0.0067 - 0.018 ms^{-1} by Sheu *et*

al. (1996), 0.065 ms^{-1} by Tasdemir (1997), 0.004 - 0.037 ms^{-1} by Franz *et al*. (1998), 0.067 ms^{-1} by Odabasi *et al*. (1999), and 0.045 ms^{-1} by Vadar *et al*. (2002).

An average overall particle phase dry deposition velocities of the PAHs with molecular weights between 154 and 202 was 0.14 cms^{-1}, and that of the PAHs with molecular weights between 228 and 276 was 0.04 cms^{-1}. This decrease in deposition velocity with increasing molecular weight is supported by other experimental studies which have shown that a greater fraction of the higher molecular weight PAHs are associated with smaller particles in comparison with the lower molecular weight compounds (Winberry *et al*., 1988; Pistikopoulos *et al*., 1990; Lewis *et al*., 1991; Aceves and Grimalt, 1993; Poster *et al*.,1995; Allen *et al*., 1996; Kiss *et al*., 1998; Kaupp and McLahlan, 1999; Odabasi *et al*., 1999; Kaupp and McLahlan, 2000; Vadar *et al*., 2002).

Fluxes of mass and PAHs measured by downward surface plate and bucket method were nearly proportional to wind speed (Fig. 6 and Fig. 7), but fluxes of mass measured by upward plate did not change with wind speed.

The correlation coefficients between wind speed and deposition fluxes of mass and PAHs in bucket methods were 0.91 and 0.89, while the correlation coefficients between wind speed and the net fluxes of mass and PAHs were only 0.24 and 0.18, respectively. Deposition fluxes of mass and PAHs on the downward surface plates weakly correlate with wind speed (r = 0.48 and 0.52). The deposition fluxes of PAHs on the downward surface plate correlated well with the flux to the upward surface plate (r = 0.75), while deposition fluxes of mass on the downward surface plate insignificantly correlated with the flux to the upward surface plate (r = 0.04).

The data in this study showed that deposition flux and velocity increase with wind speed. The previous finding by Noll *et al*. (1988) found that the mass fluxes ratio of upward surface plates to downward surface plates was 0.33, while it was 0.13 in this study. The fluxes ratio of
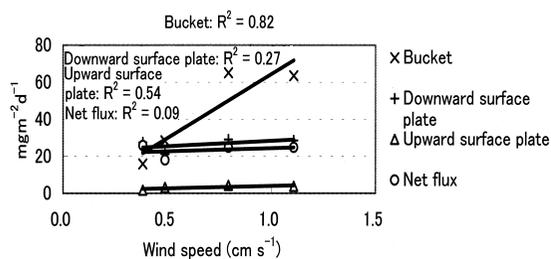
Fig. 6 Correlation analysis on deposition fluxes of mass (mgm^{-2}d^{-1}) with wind speed (cms^{-1}) at 1.5 m height
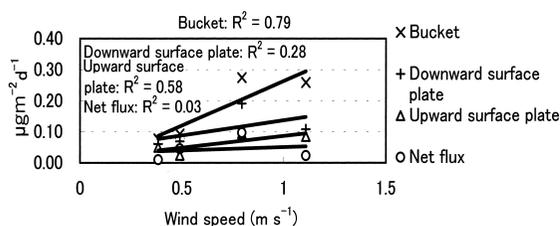
Fig. 7 Correlation analysis on PAHs deposition fluxes (ΣPAH congeners; μgm^{-2}d^{-1}) with wind speed (ms^{-1}) at 1.5 m height

the downward surface plate and upward surface plate provides information that can determine if particles remain suspended in the atmosphere.

The profile of dry deposition flux of individual PAHs were fairly similar regardless of sampling heights and sampling methods (Fig. 8). Phenanthrene, fluoranthene and pyrene were the most abundant compound measured in this study. The profile of dry deposition flux of individual PAHs in this study was nearly the same magnitude as the previous studies (Odabasi *et al*., 1999; Vadar *et al*., 2002).

3.2. Ambient particle phase concentration

The suspended particles were rich in high-molecular PAHs (Fig. 9). The particles captured by upward surface plates were rich in low-molecular PAHs. PAH compositions were fairly similar regardless of sampling

heights. As to suspended particles, the smaller particles were very rich in low and high molecular weight PAHs compared with larger particles (Fig. 10). The small particles captured by low pressure cascade impactors showed higher PAH composition than that of deposited particles in bucket. In addition, a smaller particle has a higher specific area, and a higher attachment rate for organic pollutants. It may therefore contain a greater amount of organic carbon, which allows more PAH adsorption. PAH compositions of deposited matter were fairly similar regardless of sampling heights. The dominances of the contribution of small particles to the total PAHs in this study were also of nearly the same magnitude as the result obtained by Hildemann *et al*. (1991), Venkataraman *et al*. (1994) and Chen *et al*. (1997).
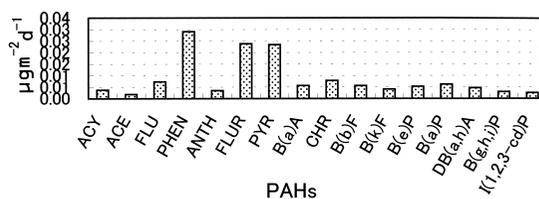
Fig. 8 Averaged individual PAH fluxes (μgm^{-2}d^{-1}) by downward surface plate method at 0.5 m height
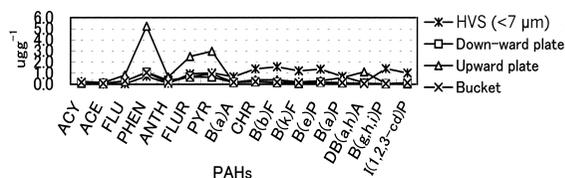
Fig. 9 PAH composition (ugg^{-1}) of the deposition samples at 1.5 m height and the suspended particles captured by high volume sampler (HVS) at TERC, Tsukuba
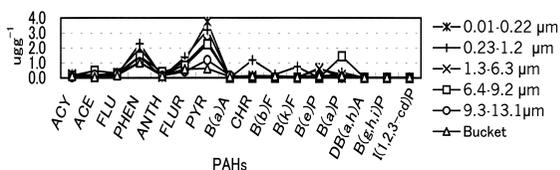
Fig. 10 PAH composition (ugg^{-1}) of the deposition samples measured by bucket and particle size measured by low pressure cascade impactor (LP-20)

Discussion

1. Difference between the methods

The deposition fluxes of mass and PAHs measured by the bucket method are about 1.6 and 1.4 times those by the downward surface plate methods, respectively, and about 1.9 and 3.6 times their net fluxes (downward minus upward), respectively (Fig. 2 and Fig. 3). The higher ratio of bucket flux to net flux for PAHs compared with mass, which resulted from the smaller difference in the fluxes between the downward and upward surface plate methods, indicates the difference in settling behaviors of the particles between with high and poor PAHs contents.

As for the water vessel, we can not explain the small deposited flux of mass in comparison with those by bucket and downward surface plate, suggesting the measurement errors; on the other hand, the coincidence of deposited PAHs fluxes between the bucket and water vessel appears reasonable.

Fig. 11 shows the possible errors induced by the three methods for measuring the settling fluxes. The downward and upward surface plates have a possibility of underestimating the actual fluxes because of the release of once-attached particles from the plate due to strong wind and/or gravitational force, reflection of particles due to their inertia, etc. This underestimation leads to the underestimation of the real flux estimated by the net flux.

On the other hand, the bucket and water vessel would overestimate the real flux because they substantially suppress the upward flux. Therefore, it is supposed that the real flux is present between the net flux and the bucket flux (or water vessel flux).

Then, we estimate the real flux. The hypotheses for the estimation are as follows:

1) The reduced rate { α : (actual flux - measured flux) / actual flux} for downward surface plate is identical to that for upward surface plate.

(1) Release from the board due to strong wind
(2) Gravitational force
(3) Reflection

May be underestimation of deposited flux

(1) Reduce upward flux

Overestimation of deposited flux

(1) Diffusion from the air
(2) Neglected up ward flux

Overestimation of deposited flux
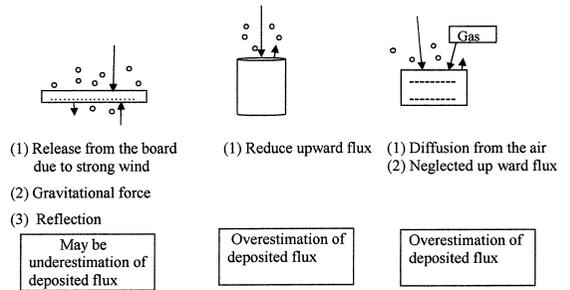
Fig. 11 Measurement errors of deposition flux

2) The reduced rate is identical for mass and PAHs.
3) The actual downward flux is measured by the bucket method.

Then, (1 - α) is estimated to be 0.6 (mass) to 0.7 (PAHs) by the downward surface plate flux divided by the bucket flux. Accordingly, the real flux is calculated to be the net flux divided by (1 - α); the ratios of the real flux to the flux measured by bucket are 1.2 for mass and 2.5 for PAHs. Because the calculated (1 - α) is underestimated due to the hypothesis 3), ratios of the real flux to flux measured by bucket are larger than the values above.

In conclusion, the deposition fluxes of mass are similar between the net (downward minus upward) and bucket methods because the upward flux of large particles representing mass is negligibly small compared with the downward flux even during strong wind period (Fig. 12). There would be insignificant differences in estimating the real mass flux between the net and bucket methods.

Small particles; PAHs rich particles

Gas

Larger particles; PAHs poor particles
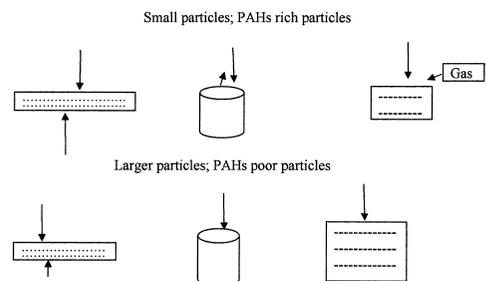
Fig. 12 Larger differences in smaller particles between net flux and bucket deposition flux

On the other hand, the upward flux should not be neglected for small particles e.g. the ones with high PAHs content. In such a case, the net flux is the better estimate for the real flux.

2. Height difference

It can be supposed that the reason why net fluxes decrease with height because real fluxes decrease with height; whereas decrease in the bucket flux with height can be a result of re-suspension from ground. This decrease in real flux with height is probably due to the vertical distributions of suspended particles and/or the vertical distribution of settling velocity. The vertical distributions of suspended particles and settling velocity depend not only on the positions of the sources but also the vertical mixing rate.

Conclusions

This study focused to estimate real fluxes of mass and PAHs on the ground surface using different settling methods at the three different heights. The main conclusions are summarized as follows:

(1) Comparing the different heights, we have noted that bucket deposition fluxes and net fluxes decrease with height due to re-suspension and/or vertical distributions of suspended particles and settling velocity.

(2) Comparing the different settling methods, we have obtained higher settling flux in bucket method compared with net-flux, particularly for PAHs. The bucket method probably over-estimates and net-flux method underestimates the real deposition flux.

(3) To estimate real fluxes on the ground surface, we can use the net flux.

(4) PAHs are rich in small particles.

Acknowledgements

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