



A new sampler for the collection and retrieval of dry dust deposition

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ABSTRACT

Atmospheric dust can influence biogeochemical cycles, accelerate snowmelt, and affect air, water quality, and human health. Yet, the bulk of atmospherically transported material remains poorly quantified in terms of total mass fluxes and composition. This lack of information stems in part from the challenges associated with measuring dust deposition. Here we report on the design and efficacy of a new dry deposition sampler (Dry Deposition Sampling Unit (DSU)) and method that quantifies the gravitational flux of dust particles. The sampler can be used alone or within existing networks such as those employed by the National Atmospheric Deposition Program (NADP). Because the samplers are deployed sterile and the use of water to remove trapped dust is not required, this method allows for the recovery of unaltered dry material suitable for subsequent chemical and microbiological analyses. The samplers were tested in the laboratory and at 15 field sites in the western United States. With respect to material retention, sampler performance far exceeded commonly used methods. Retrieval efficiency was > 97% in all trials and the sampler effectively preserved grain size distributions during wind exposure experiments. Field tests indicated favorable comparisons to dust-on-snow measurement across sites (r^2 0.70, p < 0.05) and within sites to co-located aerosol data (r^2 0.57–0.99, p < 0.05). The inclusion of dust deposition and composition monitoring into existing networks increases spatial and temporal understanding of the atmospheric transport on materials and substantively furthers knowledge of the effects of dust on terrestrial ecosystems and human exposure to dust and associated deleterious compounds.

1. Introduction

Several separate lines of research provide strong evidence that dust transport has increased in recent decades over large areas of the western United States as compared to Holocene averages (Brahney et al., 2013; Clow et al., 2016; Hand et al., 2016; Neff et al., 2008). Because current national atmospheric monitoring networks were specifically designed to sample acid deposition (Lamb and Bowersox, 2000) and particles of sizes that affect visibility (UCDavis, 1999), only wet deposition and particles below 10 μm are routinely monitored. However, the bulk of regional dust mass flux occurs in size fractions greater than 20 μm and up to 250 μm (Lawrence and Neff, 2009) leading to several significant data and knowledge gaps. In particular, the atmospheric pathway represents a poorly constrained flux of material within regional biogeochemical cycles (Brahney et al., 2015). For example, a critical unknown is the bioavailability of various phosphorus- and

nitrogen-containing compounds found in dry deposition. In addition, atmospheric dust acts as a vector for the transport of accumulated deleterious compounds including pathogens, heavy metals, organic contaminants, and other toxins (Boxall et al., 2009). All of these can have significant human and ecosystem health consequences. For example, incidents of Valley Fever, caused by inhaling the soil pathogen, *Coccidioides*, are on the rise in the southwestern United States (CDC, 2013; Chow et al., 2016), as are other common aeroallergens related to increased emissions of soils, wildfire particulates, and plant products (Takaro et al., 2013). Dust transport may also represent an important mechanism for the dispersal of microbes across ecosystems (Dastrup et al., 2018). Taken together, the inclusion of particulate deposition monitoring within a comprehensive network (or alone) can be used to quantify the atmospheric transport of constituents that have significant biogeochemical, ecological, and human health consequences.

Dust can be difficult to measure owing to its episodic and spatially

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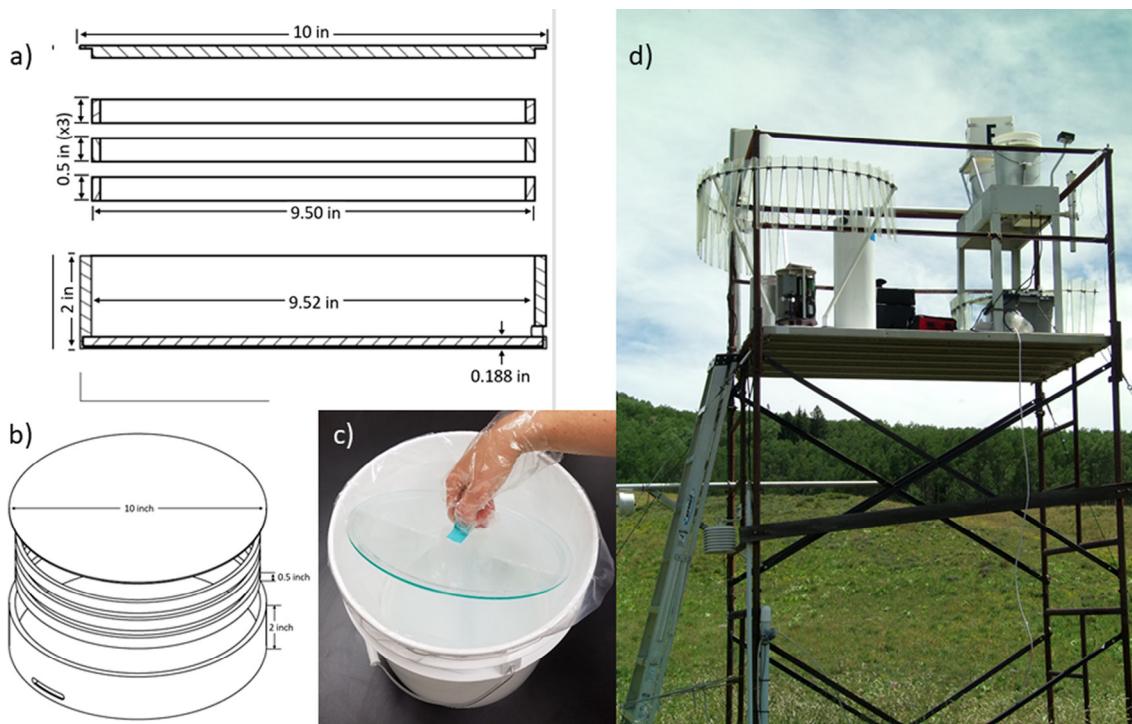


Fig. 1. Design of the Dry Sampling Unit (DSU) manufactured at SensorSpace, Flathead Lake Biological Station, University of Montana. a) and b) the design and dimensions. The hole at the bottom facilitates the removal of dust material. c) The sampler in the NADP bucket and the lid being removed. d) The deployed dry bucket at an NADP station.

heterogeneous characteristics; further, measurement can be influenced by the geometry of the sampler (Goossens, 2010). Current sampling methods for measuring deposition of atmospheric aerosols at a large spatial scale are primarily limited to vacuum filtration systems that only capture particles below 10 μm (PM10) and 2.5 μm (PM2.5). These networks were not designed to capture dust and are not reliable indicators of dust events (Neff et al., 2013; Reynolds et al., 2016). In the United States, monitoring capacity is limited to a small number of total suspended particulate (TSP) samplers within state-operated networks. TSP samplers have similar inadequacies, including (1) they may be inefficient at capturing larger particle class sizes (Neff et al., 2013), which may make up a large fraction of the mass flux (Lawrence and Neff, 2009; Lawrence et al., 2010), (2) they do not capture aerosol deposition in precipitation, which can account for up to 70% of deposition (Lawrence and Neff, 2009), and (3) it is difficult to remove dust samples from TSP filters without altering their chemical compositions.

These vacuum filtration systems, deemed 'active' samplers, measure atmospheric particle concentrations rather than deposition. These data can be used to model deposition but rely on high-quality local climate data and particle settling rates, which are not well constrained for larger particle size classes (Wesely and Hicks, 2000). By contrast, three common 'passive' sampling techniques have been used to monitor vertical deposition, or the deposition that results from the gravitational flux of material. These methods include the glass marble 'cake-pan' samplers employed by the U. S. Geological Survey (USGS; Reheis and Kihl, 1995), dust-on-snow collection (Clow et al., 2016; Painter et al., 2007), and the Hubbard-Brook bulk deposition samplers (Eaton et al., 1980). All of these techniques have advantages and disadvantages that can be weighed against the nature of monitoring objectives.

Glass marbles in open vessels have long been used to collect both horizontal (dust emissions from a landscape) and gravitational transport of dust as marbles serve to trap dust and prevent finer particles from being excavated from the sampler (Reheis, 2006). These samplers are typically mounted at least 2 m above the ground to avoid contamination from saltating particles (Reheis and Kihl, 1995). Samples

are nonetheless easily contaminated by vegetation, insects, and bird feces; the latter is typically ameliorated through the use of bird deterrents (Bao and Reheis, 2003) and the former by manually removing the contaminants. Nonetheless, the introduction of contaminants into the sampler introduces uncertainty as vegetation and insects may break apart during manual removal or leach into the collected wet deposition collected with the sampler. The marble sampler has been further criticized for inaccurately measuring horizontal transport because retrieval efficiency is influenced by sampler geometry (Goossens, 2010; Goossens and Offer, 1994; Sow et al., 2006). However, gravitational deposition is less likely affected by sampler shape. Other important challenges with marble samplers include: maintenance of clean and sterile sampling surface, management of cumbersome marbles, inefficient and highly variable retrieval efficiency of material from marbles (Goss et al., 2013), and the required use of water to retrieve dust from samplers. The latter will directly influence the measurement of dust composition through the alteration of mineralogy and as easily leached and exchangeable compounds may be lost rapidly if the water used to recover the sample is not retained (Gao et al., 2017).

Methods used to collect dust-on-snow have some advantages over marble collectors in that they minimize the impact of local soil contamination but have increased contamination potential from large debris produced from above ground vegetation. Other uncertainties include the unequal loading of snow on the landscape and post snowfall wind redistribution. The combination of these effects can lead to a high spatial heterogeneity and challenges in determining a mean areal deposition rate to a catchment. Further, if melt has occurred, then soluble species may be lost (Clow et al., 2016; Lawrence et al., 2010). Importantly, this method cannot capture summer and fall events, which can be significant and even dominate in some regions (Neff et al., 2013; Dahms and Rawlins, 1996; Vicars and Sickman, 2011).

A third commonly used sampler is the bulk deposition sampler that captures both wet and dry deposition (Eaton et al., 1980). Deposition can be separated post collection into wet and dry fractions, albeit with some loss of the dry fraction to dissolution. Disadvantages are similar to

the marble collectors in that bird feces and local vegetation can contaminate the sample unless mitigated by bird rings and filters (Bao and Reheis, 2003; Brahney et al., 2014), and dry deposition that is exposed to water leads to leaching of constituents prior to chemical analyses.

To address these various deficiencies, we developed a modification that can be used alone or with the Aerochem Metrics Model 31 (ACM) wet/dry precipitation collector used by the National Atmospheric Deposition Program (NADP) (Rothert et al., 1997). ACMs have a wet and dry 3.5-gallon bucket. The dry bucket is open to gravitational deposition during dry periods. When precipitation occurs, the sensor is triggered and moves the collection lid over the dry side bucket, leaving the wet side to collect rain. Dry deposition in the dry bucket alone would be subject to wind remobilization of material. The modification utilizes Dry Deposition Sampling Units (DSUs; Fig. 1), which are designed to capture gravitational deposition and fit neatly inside the dry bucket. The DSU improves upon current methods by allowing for retrieval of dust samples year-round, and by preventing contamination through the use of multiple layers of screening. These layers also act as wind baffles to limit dust excavation from the sampler. Further, the DSUs are easy to deploy and retrieve, and are ultraviolet (UV) sterilizable or autoclavable. The capacity to retrieve dust that is uncompromised by water or solvents is critical for determining the chemical composition of dusts and in particular, the bioavailability of limiting nutrients, which is a key motivation for this study.

The objective of this study was to test sampler efficacy in both laboratory and field conditions. Specifically, we tested the DSU's capacity to 1) minimize collector obstruction, (i.e., does the sampler influence the deposition rate by retaining dust on the sampler screens?), and 2) retain dust in the collection plate during heavy winds. In addition, we wanted to compare our measured deposition rates to other aerosol and dust data streams. Specifically, we compared our data to co-located aerosol measurements from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network as well as the deposition rates determined from dust-on-snow measurements.

Benefits of widespread monitoring of dust deposition include an increased capacity to model fluxes of material, microbes, and nutrients from atmospheric deposition, and to predict ecological response to elevated dust deposition. The inclusion of atmospheric fluxes of particles greater than 10 μm , specifically of particulate nutrients, has been critically absent from both atmospheric and watershed-scale biogeochemical models primarily due to a lack of real data to parameterize these models (Arnold, et al., 2012; Wiedinmyer et al., 2017)

2. Methods

2.1. Sampler design

The DSUs were designed to capture gravitational deposition and maximize the recovery of particulates while maintaining a sterile sampling environment. The prototype samplers were constructed of glass, acrylic, nylon, and polycarbonate. A DSU consists of a glass baseplate with a high-temperature acrylic rim to collect and retrieve fallen dust. Above the glass sample plate is a series of stackable nylon mesh screens with acrylic support rings that are $\frac{1}{2}$ -inch tall. The nylon screens were pre-heated prior to assembly to avoid shrinkage anticipated from possible autoclaving. The bottom screen pore size is 250 μm , followed by one 500 μm screen, and the uppermost screen is 1000 μm . A lower limit of 250 μm was selected to capture regional to far-travelled dust particles (Lawrence and Neff, 2009). An acrylic lid is used to limit contamination during transportation to and from the field. The entire unit is 10 inches in diameter, and it can be used alone or can fit well inside the dry side bucket of the ACM collector. The screens serve multiple purposes. First, they act to prevent contamination of dust samples by large insect and vegetation fragments, which collect on the upper 1000 μm and 500 μm screens. Second, the two lower screens sort dust size fractions as they are deposited. Collectively, the screen stack is

designed to act as a wind baffle to prevent dust scouring out of the sampler (Fig. 1).

The production design, as opposed to the prototype design described above, is made of fewer materials. Constructed entirely of nylon with a glass baseplate, the production units minimize the use of any glues by using an acoustic weld to permanently fix the nylon screens to each ring layer. This glue-free approach is ideal as adhesives tend to break down after repeated UV sterilization and autoclaving, which may introduce particulates that detrimentally influence quantification. Both designs are fully autoclavable or UV sterilizable so that the sampler can be deployed sterile.

2.2. Laboratory trials on sample retrieval efficiency

To determine sampler efficacy, we performed a series of laboratory trials that include both retrieval efficiency and wind-exposure experiments. The capacity to retrieve deposited material was initially tested in the laboratory using only the glass plate and also using the glass base plate plus full screen stack. Three different mass ranges of local soil sieved to size ranges $< 250 \mu\text{m}$ were sprinkled over the sampler in triplicate to determine if retrieval efficiency would vary by deposition rate, these were 10–15, 50–60, and 100–120 mg (n = 9). Masses were chosen to reflect the range of monthly deposition based on previous studies that retrieved dusts from NADP dry buckets (Brahney, 2012). Dusts were recovered from the base plate and sampler using an ethanol-sterilized ceramic razor blade and synthetic paint brush. Samples were recovered into vacuum-ionized polycarbonate containers and weighed on a Metler-Toledo 105DUExcellence high linearity scale after passing through a Haug ring deionizer. We conducted similar addition and retrieval efficiency experiments with marbles in round plastic containers. Containers were filled to the halfway mark with marbles and dusts were sprinkled into the sampler for the same three size classes as for the DSU and added in triplicate (n = 9). To remove added dusts, we added 250 mL of deionized (DI) water three times to the sampler, swirled to disperse the marbles, and filtered through a pre-weighed 0.45- μm polyethersulfone (PES) filter. Recovered DSU samples were analyzed for grain size pre- and post-addition using a Malvern Mastersizer 3000 laser wet-dispersion particle size analyzer to determine whether inefficiencies in grain size recovery exist. We could not conduct grain size analyses on marble-recovered material as they were impacted on a filter.

2.3. Wind excavation experiments

To establish the effectiveness of the DSU in preventing wind excavation loss, we conducted a series of wind exposure experiments. Dust samples of known mass and pre-determined particle-size distribution were added to a bucket with a DSU sampler and a control consisting of a bucket with just a base plate. The DSU and control buckets were exposed to 30-mile-per-hour winds for 120 s in triplicate. Recovered samples were analyzed for mass and grain size as described above.

2.4. Field trials on sample recovery

To establish efficacy in the field, a pilot study at 15 NADP sites was conducted continually between October 2017 and August 2019 (Fig. 2; Table 1). Prior to deployment, samplers were cleaned using a combination of ethanol and vacuuming and then UV sterilized for 15 min. Sample buckets were lined with a custom clean-room-grade, low-density polyethylene bag, and a vacuum was applied to a hole in the exterior of the bucket to mold the bag to the bucket interior. The DSU was then placed in the bucket using gloves and a clean handling technique developed by the authors. The DSUs were secured in the bag-lined bucket with a polyethylene lid (Fig. 1). The assembled DSUs were transported to each station and deployed by trained site operators. At

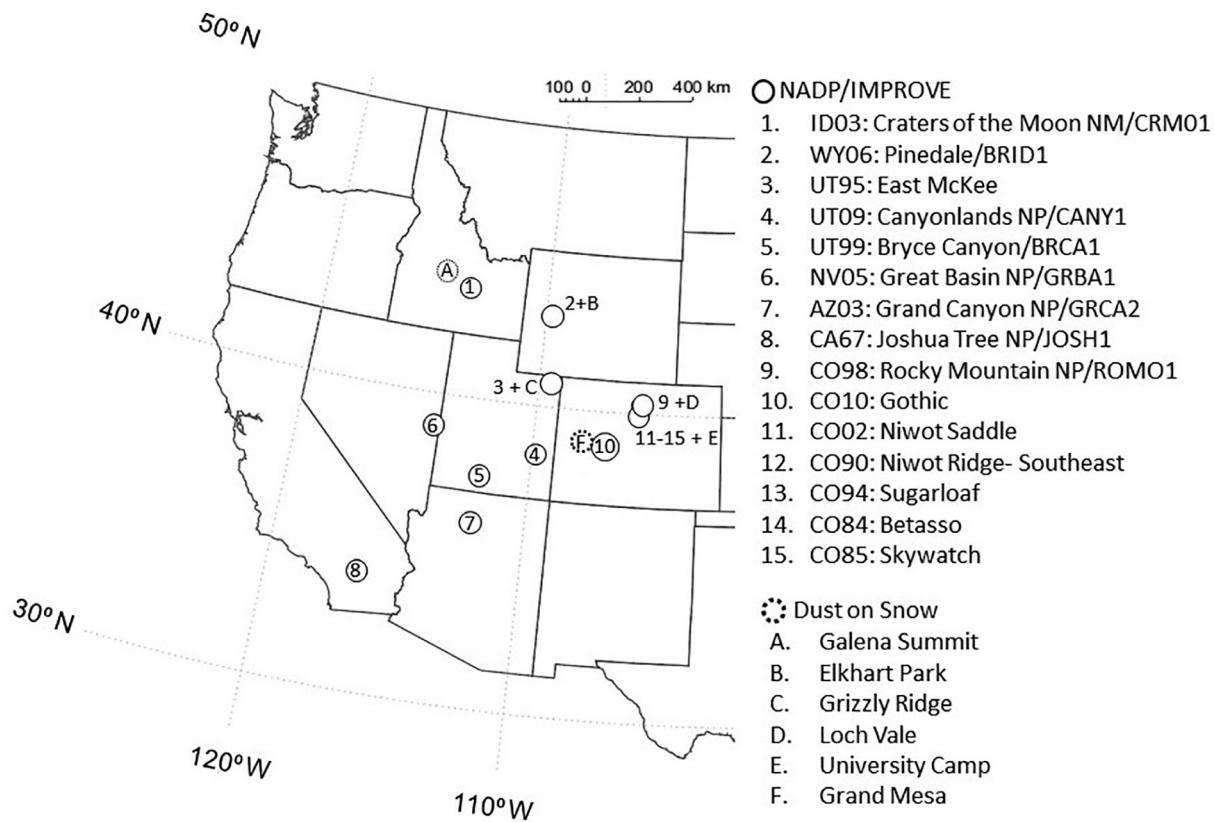


Fig. 2. Locations of NADP field sites and co-located USGS dust-on-snow (DOS) sites (Ingersoll, et al., 2002).

each site, the operators removed the bucket lids with sterile gloves and placed it in a clean Whirl-Pak™ bag for storage. On the first Tuesday of every month, the site operators resealed the DSUs with the stored lids and shipped them to Utah State University (USU). Samples were collected monthly when possible. In the laboratory, dust was retrieved and weighed as above, and containerized samples were catalogued and stored. To account for the total mass deposited, residual dust particles adsorbed to the polyethylene bags were removed by a triple DI wash

that was then filtered first through a 250 μm screen to remove large particles followed by a pre-weighed 0.45 μm PES filter. Note as discussed above, this fraction would be less suitable for chemical analyses. Filters were first desiccated for 24 h before re-weighting and storage. The PES-filtered mass was added to the dry particulate mass for calculation of the monthly mass of dry-deposited dust at each site.

All methods of measuring gravitational deposition have merits and limitations (see introduction). Therefore, it is not possible to compare

Table 1

Study site location information. The DSU were placed at the 13 listed NADP sites and the 2 Boulder Creek Critical Zone Observatory (BC CZO) sites. Six of the IMPROVE sites were co-located with the NADP sites, while two were a few miles distant (BRID1, ROMO1).

Site Name	Site Code	Agency	Latitude	Longitude	Elevation (masl)	Setting
Crater of the Mood NP, ID	ID03/CRM01	NADP/IMPROVE	43.4605	-113.5551	1807	Plains
Wind River Range, WY	WY06	NADP	42.9290	-109.7875	2388	Foothills
Unita NF, UT	UT95	NADP	40.7543	-109.4671	2522	Montane
Canyonlands NP, UT	UT09/CANY1	NADP/IMPROVE	38.4584	-109.8210	1797	Desert
Bryce Canyon NP, UT	UT99/BRCA1	NADP/IMPROVE	37.6186	-112.1728	2477	Plateau
Great Basin NP, NV	NV05/GRBA1	NADP/IMPROVE	39.0054	-114.2170	2066	Foothills
Grand Canyon NP, AZ	AZ03/GRCA2	NADP/IMPROVE	36.0586	-112.1840	2071	Plateau
Joshua Tree NP, CA	CA67/JOSH1	NADP/IMPROVE	34.0695	-116.3889	1239	Desert
Rocky Mountain NP, CO	CO98	NADP	40.2878	-105.6628	3159	Subalpine
Gothic, CO	CO10	NADP	38.9561	-106.9860	2915	Foothills
Niwot Saddle, CO	CO02	NADP	40.0547	-105.5891	3520	Alpine
Niwot Ridge- Southeast, CO	CO90	NADP	40.0360	-105.5441	3030	Subalpine
Sugarloaf, CO	CO94	NADP	39.9939	-105.4800	2524	Montane
Betasso, CO	CO84	BC CZO	40.0139	-105.3463	1975	Foothills
Skywatch, CO	CO85	BC CZO	40.0099	-105.2422	1600	Plains
Bridger, WY	BRID1	IMPROVE	42.9749	-109.7579	2616	Foothills
Rocky Mountain NP, CO	ROMO1	IMPROVE	40.2783	-105.5457	2752	Montane
Galena Summit, ID	GS	USGS DOS	43.8744	-114.7144	2683	Subalpine
Elkhart Park, WY	EP	USGS DOS	43.0027	-109.7570	2877	Foothills
Grizz Ridge, UT	GR	USGS DOS	40.7489	-109.5051	2914	Montane
Loch Vale, CO	LV	USGS DOS	40.2903	-105.6667	3215	Subalpine
University Camp, CO	UC	USGS DOS	40.0328	-105.5760	3159	Montane
Grand Mesa, CO	GM	USGS DOS	39.0328	-107.9775	3133	Valley

the DSU catch efficiency directly to other methods. However, there is utility in comparing our dust deposition rates to other data streams. Specifically, we compared our monthly dust recovery (mg m^{-2}) to aerosol concentration measurements from IMPROVE ($\mu\text{g m}^{-3}$). In addition, mean winter deposition measurements from the field trials were compared to dust-on-snow samples that capture both wet and dry particulate deposition. Depth-integrated snow samples were collected at 6 locations across the study area in late March or early April (Fig. 2). At each location, a snowpit was dug to the ground surface, snowpack physical properties (e.g., snow depth, snow water equivalent, and snow temperature) were measured, and a single depth-integrated snow sample was collected and composited into a clean Teflon bag using sterile techniques (Ingersoll, et al., 2002). All snow samples were preserved and shipped frozen to USU. Snow samples were allowed to melt in the Teflon bags at room temperature prior to filtration through pre-weighed $0.45\text{ }\mu\text{m}$ PES filters. Each bag was rinsed three times with DI water and filtered to maximize recovery of materials from within the Teflon bag. Six of the 15 NADP sites had nearby dust-on-snow samples. Mean daily deposition rates for dust-on-snow were determined based on the total mass recovered and the period of snow cover; the start date was determined based on recorded snow accumulation measurements at nearby snow telemetry (SNOWTELE) (USDA, 2019) stations and the sampling date marked the end of the accumulation period. SNOWTELE sites were less than 10 km from most DSUs and ~ 25 km from the ID03 site. Wet particulate deposition rates were determined for the NADP sites from the $0.45\text{-}\mu\text{m}$ filters used in preparation of wet deposition analyses from the NADP Central Analytical Laboratory (CAL) (SOP# PR-1055) at the Wisconsin State Laboratory of Hygiene. Daily mean wet and dry deposition rates ($\text{mg m}^{-2} \text{ day}^{-1}$) were compared between the paired NADP and USGS snow sampling locations for the 2017/2018 snow year. No samples were collected in the 2018/2019 snow year.

3. Results

3.1. Laboratory trials on sample retrieval efficiency

There was no statistical difference between the retrieval efficiency from the glass base plate alone or with the full DSU screen stack (Table 2), indicating the sampler screens do not trap material $< 250\text{ }\mu\text{m}$. Retrieval efficiency from the DSU was an average of $97.9 +/ - 1.2\%$ for initial masses ranging $10\text{--}15\text{ mg}$, $97.4 +/ - 0.9\%$ for initial masses between 50 and 60 mg , and $97.7 +/ - 0.4\%$ for initial masses ranging from $100\text{--}120\text{ mg}$. The average loss during base plate and DSU recovery was only $1.1 +/ - 1.0$ and $1.4 +/ - 0.3\text{ mg}$ of material (Table 1). Importantly, no statistical differences occurred between the grain size mass distribution of the initial sample and the DSU recovered sample ($p = 0.44$; Fig. 3). Visual inspection indicated that dust material passed easily through the screen stack without measurable electrostatic retention of material on the screens themselves. Data for the marble-based samplers indicated variable mass recoveries ranging from 57 to 83% for the 10 mg trial, 36 to 83% in the 50 mg trial, and 49 to 71% in the 100 mg trial (Table 2).

Table 2

Results of laboratory experiments ($n = 3$ for each size class) indicating similar retrieval efficiencies of material from the glass base plate alone (control) as compared to the DSU sampler. Data also show improved retrieval efficiency for the DSU as compared to marble based samplers. Data in brackets represent the standard error.

Initial weight (mg)	Control % (+/-)	DSU % (+/-)	T-test p-value	Marble sampler % (+/-)
10–15	97.2 (1.4)	97.9 (1.2)	0.54	72.7 (7.8)
50–60	97.6 (1.5)	97.4 (0.9)	0.79	55.2 (14)
100–120	98.6 (1.6)	97.7 (0.4)	0.38	58.9 (6.3)
Average	97.8	97.7		62.3

3.2. Wind excavation experiments

Wind exposure experiments showed the control samples, using a glass base plate only, had a mean recovery of $46.5 +/ - 6.7\%$ and average mass loss of $87 +/ - 47\text{ mg}$ (Table 3). By comparison, sample recovery from the DSUs exceeded 99% with an average mass loss of $1.2 +/ - 0.05\text{ mg}$. Visual inspection confirmed no disturbance of dust material within the glass base plate after wind exposure of the DSUs. Differences or similarities in grain size distribution were determined using ANOVA. There was no change in grain-size distribution of the initial sample and that recovered from the DSU post wind-exposure ($F = 0.59, p = 0.52$), whereas the grain-size distribution in the control experiment indicated a significant loss of fines influencing the mean grain-size distribution (Fig. 3) ($F = 4.51, p < 0.05$).

3.3. Field trials on sample recovery

Throughout the field trials several adjustments were made to the design. These adjustments primarily consisted of adjusting the type of glue because decomposition of the glues holding the screens to the polycarbonate rings was observed. Samples sometimes arrived wet because the precipitation sensor on the Aerochem Metrics collector is not sensitive to light snow fall and occasional malfunction of the collectors can occur. Wet samples were placed in a desiccator until completely dry. Field trials demonstrated that the DSU effectively prevents sample contamination by bird feces and large fragments of vegetation. In 3 of 192 samples, bird feces were noted on the surface screen but did not pass through to screen stacks below. Though rare, large vegetation fragments were effectively retained on the uppermost screen. The lower screen sorted size fractions greater than $> 250\text{ }\mu\text{m}$, the mass of which was determined during the double filtration ($250\text{ }\mu\text{m}$ screen and pre-weighed $0.45\text{ }\mu\text{m}$ PES filter) as described above. Electrostatic retention of dust on the polyethylene bags was observed and this mass is accounted for from pre- and post-weighing of the filters.

There were strong agreements for within-site month to month comparisons of DSU dust deposition rates and IMPROVE aerosol concentrations (Table 4, Fig. 4). Masses recovered from dust-on-snow (wet and dry) and those recovered in the DSU (wet + dry) showed a strong relationship ($r^2 = 0.70, p < 0.05$) though dust recovered from snow was higher (Fig. 5). Dry deposition made up 44 to 93% of the total deposition (average 67%) (Table 4).

4. Discussion

The controlled laboratory experiments, field trials, and wind exposure experiments demonstrate that the DSU offers a considerable improvement over existing dry recovery methods in multiple respects. First, our design allows for the effective capture and retrieval of material across a wide range of dust grain size classes from clays ($< 2\text{ }\mu\text{m}$) to coarse silts and fine sands ($250\text{ }\mu\text{m}$). In comparison, vacuum filtration systems may be inefficient at capturing particle sizes at or near their upper limit of $100\text{ }\mu\text{m}$ (Neff et al., 2013). Secondly, retrieval efficiencies from the DSU ranged from 97 to 99%. Further, our dust retrieval efficiencies were similar over the range of sample masses (10 to 120 mg). The performance of the DSU shows a substantial improvement over the variable retrieval efficiencies reported for marble-based samplers shown here (36–83%) and elsewhere (Goss et al., 2013). Thirdly, the DSU effectively separated contamination including large fragments of vegetation (leaves, needles) and bird feces from the gravitational flux of small particles. Contamination by birds and vegetation is a confounding problem for other gravitational-based systems including marble, bulk, and dust-on-snow methods. Fourthly, the DSU allows for the recovery of dry material without the use of water, leaving the sample intact. Marble and bulk samplers require the use of water, and recovery of dust from filters requires the use of a solvent to remove the filter from the material for subsequent analyses; both of these

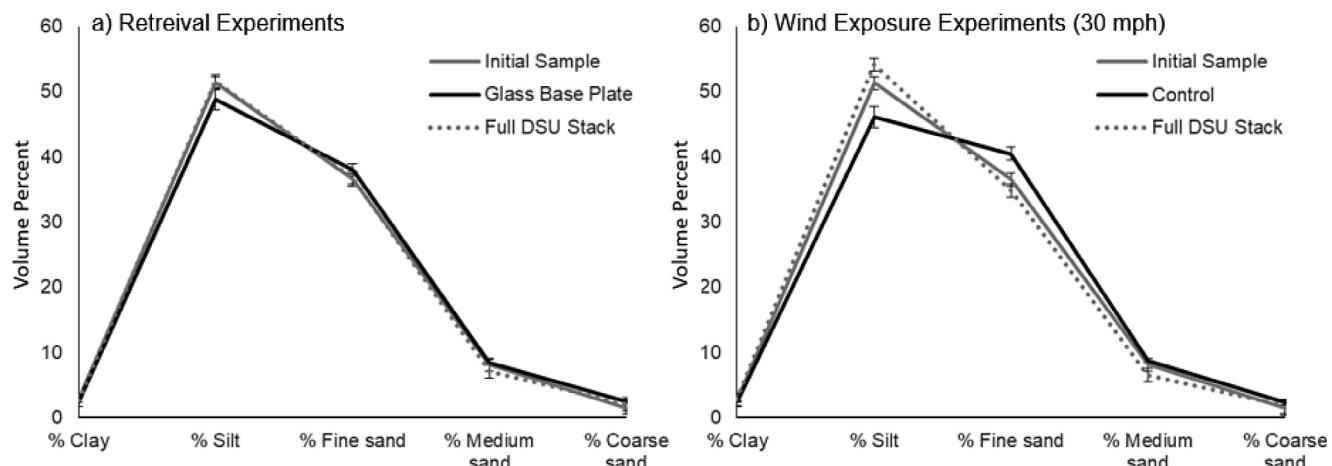


Fig. 3. Results of experimental trials a) grain size distribution of recovered material from the base plate alone and full DSU stack. No statistical significances between sample pairs were found. b) grain-size distribution of recovered material from NADP dry-side bucket using the base plate alone (control) and the full DSU stack. The control experiment showed a significant loss of fines as compared to the initial sample and DSU recovery.

Table 3

Results of wind exposure experiments comparing buckets with only a glass base plate and the Dry Deposition Sampler (DSU) stacked filter samplers, $n = 6$.

Initial mass (mg)	Post-Wind Exposure		
	Mass recovered (mg)	Percent recovered (%)	Mass loss (mg)
Glass base plate			
1	228.88	89.01	38.9%
2	102.72	50.76	49.4%
3	141.95	72.75	51.3%
Average		46.5%	87.01
DDS			
1	189.37	188.12	99.3%
2	139.97	138.82	99.2%
3	186.32	185.1	99.3%
Average		99.3%	1.21

Table 4

Coefficients of determination for the relationship between measured dust deposition and IMPROVE aerosol concentrations, and Dust-on-Snow (DOS). The proportion of particles falling dry is also reported.

Site Name	Site Code	r^2	p	Fraction Dry
Crater of the Mood NP, ID	ID03/CRMO1	0.99	< 0.001	0.75
Wind River Range, WY	WY06/BRID1	0.60	< 0.001	0.76
Canyonlands NP, UT	UT09/CANY1	0.71	< 0.05	0.93
Bryce Canyon NP, UT	UT99/BRCA1	0.96	< 0.001	0.55
Great Basin NP, NV	NV05/GRBA1	0.82	< 0.001	0.66
Grand Canyon NP, AZ	AZ03/GRCA2	0.99	< 0.001	0.81
Joshua Tree NP, CA	CA67/JOSH1	0.83	< 0.001	0.87
Rocky Mountain NP, CO	CO98/ROMO1	0.57	< 0.05	0.44
Unita NF, UT	UT95	N/A	N/A	0.58
East River, CO	CO10	N/A	N/A	0.55
Niwot Saddle, CO	CO02	N/A	N/A	0.53
Niwot Ridge- Southeast, CO	CO90	N/A	N/A	0.44
Sugarloaf, CO	CO94	N/A	N/A	0.64
Betasso, CO	CO84	N/A	N/A	0.65
Skywatch, CO	CO85	N/A	N/A	0.91
DSU vs DOS	All sites	0.70	< 0.05	

procedures fundamentally alter the mineralogy and chemistry of the sample and introduce uncertainty. Finally, the fully autoclavable or UV sterilizable design allows the sampler to be deployed sterile so that fungal and microbial analyses of dust material may be possible. These latter two features are critical for the characterization of dust elemental forms and availability as well as microbiological assessment.

We found low and variable retrieval efficiencies from marble-based samplers similar to other studies (Goss et al., 2013) (Table 2). Low retrieval efficiency likely arises for several reasons. The primary reason appears to be low recovery from the large surface area that marbles provide, a simple rinse and agitate does not appear to be enough to completely bring all particulates into solution. In addition, marbles are heavier than water and impact upon each other, likely trapping some particles. Lastly, some soluble salts, alkaline minerals, and nutrients may be lost to solution (Clow et al., 2016; Chu et al., 2018; Chen and Chen, 2008). The latter can be ameliorated if the sample is kept and evaporated, though notable changes in mineralogy can occur.

We found strong relationships between IMPROVE aerosol atmospheric concentrations and the DSU in co-located sites (r^2 0.71–0.99) (Fig. 4). The weakest relationship was between the IMPROVE and DSU in Rocky Mountain National Park (r^2 0.57), where the sites are approximately 11 km and several mountain peaks apart. Though the month to month variations were generally in agreement, these discrepancies highlight the spatial variability in dust deposition in mountain landscapes. Dust-on-snow deposition rates were higher than those measured from the DSU (Fig. 5). Potential reasons for lower measured dust deposition rates could be related to the inefficiencies associated with horizontal transport and deposition during heavy winds, where the presence of the sampler itself causes perturbations of the air flow that result in an area of low dust deposition (Goossens, 2010). However, since the intention of this sampler is to measure gravitational (vertical) rather than erosional (horizontal) deposition, this may add an advantage in that it may diminish contamination for local erosional sources. Nevertheless, adding wind-baffles around the larger sampling unit could potentially increase particle capture during heavy winds (Reheis and Kuhl, 1995). Though specific studies on gravitational deposition and sampler geometry could not be identified, there is likely airflow disturbance along the rim that alters particle flow (Goossens and Offer, 1994). Potential limitations of the dust-on-snow data include (1) high local variability in dust-on-snow due to redistribution and dust focusing, (2) greater efficiencies in the atmospheric scavenging of particulates from the greater surface area of snow, and/or (3) all dust-on-snow samples contained large fragments of vegetation (> 250 μ m) that could contaminate the measured sample. This fraction was typically > 200% of the weight of the < 250 μ m fraction. Thus, any fragments from these larger fractions that contaminate the < 250 μ m fraction can influence the mass recorded. Despite these uncertainties, dust-on-snow and the DSU showed across-site agreement in relative deposition rates.

Inclusion of the DSU into networks such as the NADP will allow for

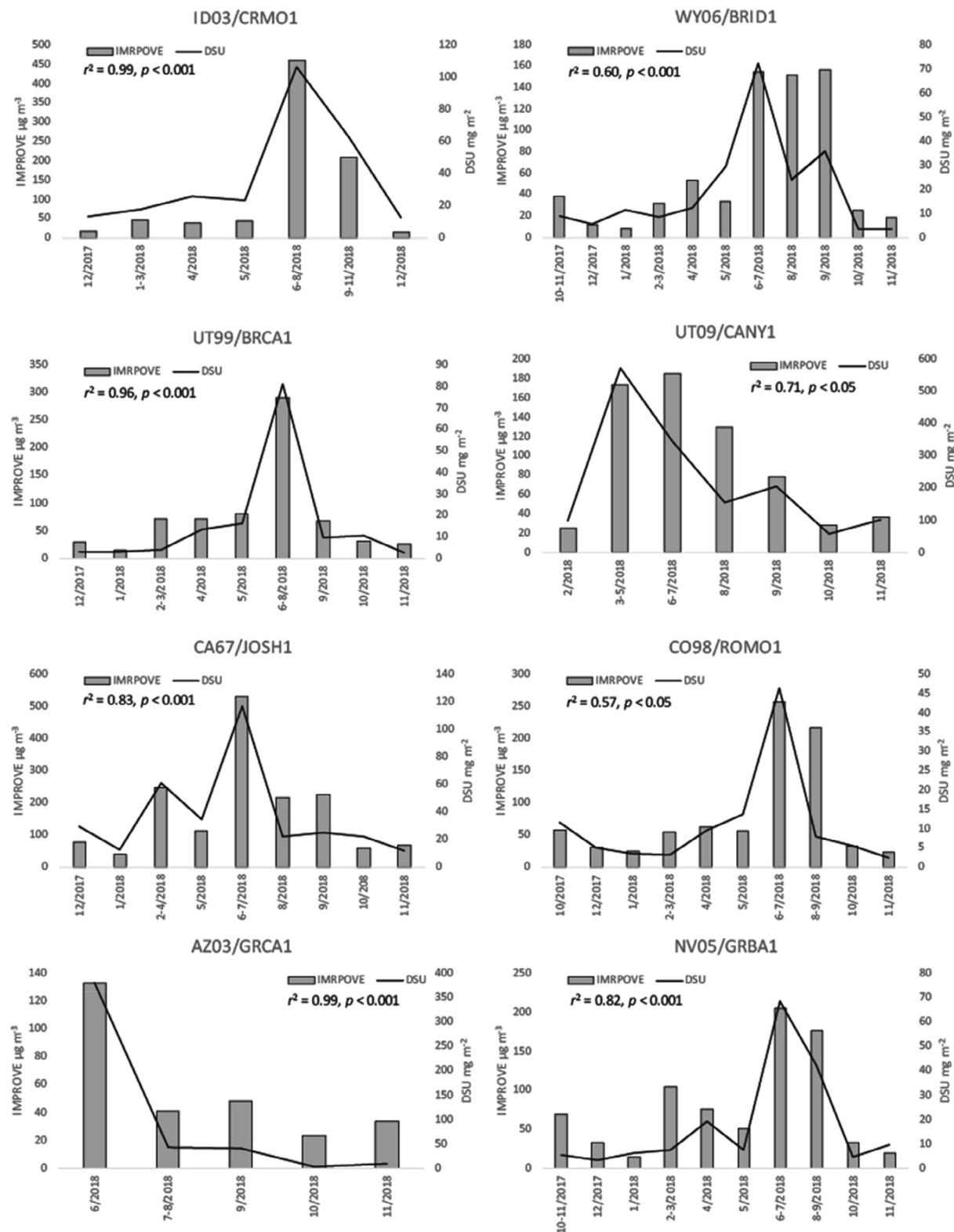


Fig. 4. Comparison between monthly DSU deposition data (mg m^{-2}) and IMPROVE aerosol concentration data ($\mu\text{g m}^{-3}$).

the first time the full quantification of ecologically relevant constituents in both the dry and wet portions of atmospheric deposition sampled independently. Because significant amounts of both nutrients and alkaline minerals can be transported as dust particles (Brahney et al., 2013; Rogora et al., 2004; Tait and Thaler, 2000; Wu et al., 2013), sampling wet deposition alone will not fully capture nutrient or ion loads. Of interest is the atmospheric deposition of the key nutrient

phosphorus (P) because it is often the most limiting nutrient in many terrestrial and freshwater ecosystems (Elser et al., 2007). Dust can contain appreciable amounts of phosphorus (Lawrence et al., 2010; Brahney et al., 2014; Mahowald et al., 2017; Aciego et al., 2017; Zhang et al., 2018), and there is a growing body of evidence that dust-associated phosphorus can produce measurable consequences in lake ecosystems (Brahney et al., 2015; Brahney, 2014; Reche, 2009; Ballantyne,

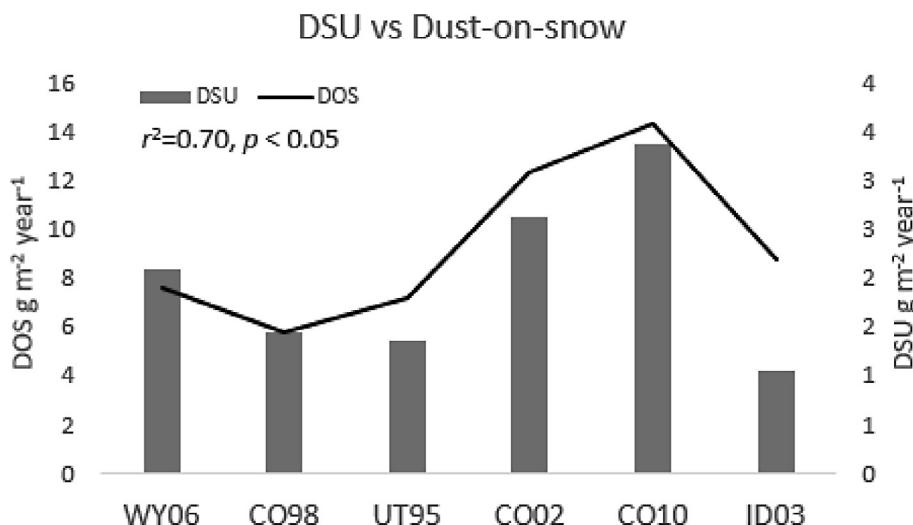


Fig. 5. Comparison between dust deposition rates determined using the DSU and dust-on-snow (DOS) during the winter of 2017/2018. Data were scaled to $\text{g m}^{-2} \text{yr}^{-1}$ to align with common reporting units in the dust literature. Note DOS sites were not co-located with DSU sites. Distances were WY06: 9 km, CO98: 0.5 km, UT95: 3 km, CO02: 3 km, CO10: 90 km, ID03 60 km.

2011; Camarero and Catalán, 2012; Sickman, 2003; Vicars et al., 2010; Brahney, 2015; Stoddard, 2016). In addition, relatively few studies in the continental United States have quantified the form and availability of other nutrients and metals in particulate deposition (C, N, S, Ca, etc.) and those that have done so only collected data over small spatial and temporal scales (Lawrence et al., 2010; Brahney, 2014; Munroe, 2014; Rutter, 2011; Neff et al., 2002; Mladenov et al., 2012). Though aerosol and wet deposition of nitrogen are well monitored and studied, nitrogen contributions from dry dust material are rarely analyzed and a potentially underrepresented component of the atmospheric nitrogen contribution to mountain ecosystems (Neff et al., 2002; Cornell, 2011).

Other constituents relevant to the NADP network include acid anions and base cations. The dissolution of calcium carbonate and other dust minerals either in precipitation or in depositional water bodies can provide readily available Ca^{2+} and acid neutralizing capacity (ANC) to remote mountain lakes and soils (Brahney et al., 2013; Rogora et al., 2004; Wu, 2013; Loyer-Pilot et al., 1986; Hedin and Likens, 1996; Kopáček et al., 2016; Rodhe et al., 2002; Bowman, 2018). For regions with catchments having low natural calcium abundances, this contribution may be a critical source of calcium to aquatic organisms with relatively high calcium requirements, e.g., *Daphnia*. Dust has been shown to support the Ca requirements of *Daphnia* (Jiménez et al., 2018), a keystone genus in aquatic systems (Jeziorski et al., 2008). The DSU offers the capability of obtaining a sample that is uncompromised by leaching and handling allowing for more in-depth studies on dust composition and dust nutrient bioavailability. Specifically, examining collected dusts for the solubility of key nutrients (P, N, C, Ca), their form (adsorbed, complexed, etc.) as well as other elements, e.g., the bioavailability of metal ions. Obvious next steps include testing the efficacy of the sampler in maintaining sample integrity with respect to chemical and microbial composition as compared to other methods.

Beyond the mission of the NADP, the collection, measurement, and archiving of dust materials from regional networks will create new opportunities for scientific investigation. For example, diverse bacterial and fungal communities are associated with airborne dust particles, which can serve as a vehicle of microbial dispersal (Dastrup, 2018; Yamaguchi et al., 2012). With respect to societal concerns, dust deposition data can be used to identify regions where air quality is significantly impacted by dust transport and deposition as well as regions where the transport of toxins may affect large populations. For example, in Utah, nearly 2 million people live directly downwind of eroding lakebeds that may contain cyanotoxins, fungal pathogens, heavy metals, and/or pesticides. The desiccation of terminal lake basins is occurring throughout the Great Basin of the United States (Moore, 2016; Wurtsbaugh, 2017) and elsewhere around the world (Jellison

et al., 2008; Gholampour, 2015; Brahney et al., 2019), generating new sources of atmospheric particulates near large population centers and underscoring the need for improved monitoring of particulate mass and composition transport through the atmosphere.

5. Applications

The results of this proof-of-concept study indicate that the inclusion of DSU samplers into the NADP network would allow for the full quantification of both wet and dry atmospheric deposition of nutrients and major ions across North America. The Aerochem Metrics sensor in the National Trends Network (NTN) used in this study is not as sensitive to activating under conditions of light precipitation (e.g., mist, sleet, snow flurries) as the Modified Aerochem Metrics sensor in the Mercury Deposition Network (MDN). Therefore, the dry side bucket of the Modified Aerochem Metrics mercury collectors would likely provide superior DSU sample collection. In addition, because the MDN collectors use a hydrochloric acid pre-charged sampling container, they would further allow for the quantification of total P deposition via inductively couple plasma mass spectrometry (ICP-MS) analysis of their collected wet deposition. Total wet and dry phosphorus deposition is a critical missing piece of the NADP nutrient deposition monitoring. The DSU samplers can also be used independently of the NTN or MDN networks placed on a platform with an independent precipitation sensor. Note that more than one-third of the NTN and MDN combined sites have N-CON Systems Co., Inc., precipitation collectors that do not have a dry side bucket available for dust sampling.

6. Conclusion

The DSUs offer substantial improvements on the measurement of atmospheric dry deposition as compared to other currently available methods. Specifically, the DSU samplers allow for > 97% recovery of deposited dust across all relevant mass deposition rates whereas existing methods have highly variable and poor recovery rates. Unlike alternative methods, the DSU allows for the collection of unaltered particulate material that can be used for a variety of chemical and microbial analyses. Furthermore, no bias on grain-size distributions of samples was determined. The inclusion of dry deposition data into the NADP network will significantly improve understanding and quantification of the atmospheric transport of key nutrients and the influence of atmospheric transport on recipient ecosystems.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.aeolia.2020.100600>.

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