

CHAPTER 1: IMPROVE NETWORK – PURPOSE, DESIGN, AND HISTORY

INTRODUCTION: REPORT OBJECTIVES

This report is the fourth in a series of periodic reports that describe the data collected by the Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring network. The objectives of this report were to

1. describe the spatial and seasonal variation of aerosol species contributing to visibility impairment from January 2000 through December 2004 for the combined data set from the IMPROVE network and the Environmental Protection Agency's (EPA) Speciation Trends Network (STN);
2. provide a first estimate of the apportionment of visibility impairment to these chemical species;
3. document long-term trends (7–16 years) of various aerosol species and visibility;
4. review a number of special studies that were designed to examine the robustness of algorithms used to make extinction estimates from aerosol mass concentrations;
5. and evaluate and qualify certain uncertainties in the IMPROVE measurements and examine the intercomparability of the data from IMPROVE and the STN.

1.1 OBJECTIVES OF VISIBILITY MONITORING UNDER THE IMPROVE PROGRAM

The Regional Haze Rule [U.S. EPA, 1999] requires monitoring representative of each of the 156 visibility-protected federal Class I areas (CIAs), as shown in Figure 1.1. The monitoring is conducted in order to track progress toward the goal of returning visibility in our national parks and wilderness areas (CIAs) to natural visibility conditions. Required monitoring under the Regional Haze Rule began in 2000. The deciview index, calculated from speciated ambient particle concentrations, was selected to track haze levels. This entails sampling and analysis of the major aerosol components using methods patterned after those utilized since 1987 by the IMPROVE network [Joseph et al., 1987; Sisler, 1996] and consistent with the aerosol monitoring portion of the 1999 Visibility Monitoring Guidance document issued by the EPA [U.S. EPA, 1999].

The IMPROVE program is a cooperative measurement effort designed to

1. establish current visibility and aerosol conditions in mandatory CIAs;
2. identify chemical species and emission sources responsible for existing man-made visibility impairment;
3. document long-term trends for assessing progress towards the national visibility goal;
4. and, with the enactment of the Regional Haze Rule, provide regional haze monitoring representing all visibility-protected federal CIAs where practical.

The program is managed by the IMPROVE steering committee that consists of representatives from the U.S. EPA; the four federal land managers (FLMs)—the National Park Service, U.S. Forest Service, Fish and Wildlife Service, and Bureau of Land Management; the National Oceanic and Atmospheric Administration; four organizations representing state air quality organizations—the State and Territorial Air Pollution Program Administrators/Association of Local Air Pollution Control Officials (STAPA/ALAPCO), Western Regional Air Partnership (WRAP), Northeast States for Coordinated Air Use Management (NESCAUM), and Mid-Atlantic Regional Air Management Association (MARAMA); and an associate member, the State of Arizona Department of Environmental Quality.

1.2 OVERVIEW OF THE IMPROVE MONITORING NETWORK

1.2.1 Current and Historical Sampler Siting

The IMPROVE network initially consisted of 30 monitoring sites in CIAs, 20 of which began operation in 1987, with the others starting in the early 1990s (Table 1.1). An additional approximately 40 sites, most in remote areas, that used the same instrumentation and monitoring and analysis protocols (called IMPROVE protocol sites) began operation prior to 2000 and were operated individually by federal or state organizations. Adjustments to the number of monitoring sites in the network or the suite of measurements collected at an individual site have happened on several occasions, due in some cases to scientific considerations and in others to resource and funding limitations. Many of the sites also included optical monitoring with a nephelometer, a transmissometer, and/or color photography to document scenic appearance. The optical monitoring sites are detailed below in section 1.2.3.

Beginning in 1998, the EPA began providing supplemental support to IMPROVE to expand the network in order to provide the representative speciated particle monitoring required under the Regional Haze Rule for each of the 156 mandatory federal CIAs (Figure 1.1, Table 1.2) where it is practical to do so. The expansion was not as straightforward as installing a new monitoring site within the boundaries of each of the 156 CIAs that did not already have an IMPROVE site. For one thing, many CIAs are designated national wilderness areas, for which the Wilderness Act restricts the siting of man-made items, including environmental monitoring equipment [The Wilderness Act, 1964]. Additionally, even for CIAs where monitoring is allowed (e.g., national parks), practical requirements such as power, security, and access occasionally make it difficult to find a suitable monitoring site within the CIA boundary.

Since regional haze impacts are by definition those that are distributed over a broad geographic region, a representative monitoring site does not necessarily need to be located in the CIA being represented. The practical significance of this concept is that it is possible for a site to 1) be located outside of the CIA boundaries and 2) represent more than one CIA when they are located within the same regional haze region. A clustering process, designed to limit the number of sites necessary for tracking progress under the Regional Haze Rule, identified 110 CIA clusters that require monitoring [Malm et al., 2000]. Locations for the necessary monitoring sites were chosen through a selection process detailed in Malm et al. [2000] that included reviewing the locations of existing IMPROVE sites, horizontal distance from the CIA, site elevation, and local pollution sources. The selection process was completed by the end of 1999 and installations began shortly after. At the time of this report, the network has been expanded to 167 sites, including a representative site for each of these 110 clusters and additionally to fill in the spatial gaps where CIAs are sparse or absent. These monitoring sites provide data that aid in understanding spatial patterns and are often installed to assist the sponsoring agency, such as a state, tribe, or the EPA, in meeting planning or quality assurance responsibilities.

Table 1.1. Discontinued and current IMPROVE particulate monitoring sites. The site groupings are displayed in Figure 1.2.

IMPROVE Site Group	Site Name	Site Code	State	Latitude	Longitude	Elevation (m)	Dates of Operation
Alaska	Ambler	AMBL1	AK	67.099	-157.863	78	07/2004-08/2005
	Denali NP	DENA1	AK	63.723	-148.968	658	03/1988-present
	Petersburg	PETE1	AK	56.611	-132.812	0	07/2004-present
	Simeonof	SIME1	AK	55.325	-160.506	57	09/2001-present
	Trapper Creek	TRCR1	AK	62.315	-150.316	155	09/2001-present
	Tuxedni	TUXE1	AK	59.992	-152.666	15	12/2001-present
Appalachia	Arendtsville	AREN1	PA	39.923	-77.308	267	04/2001-present
	Cohutta	COHU1	GA	34.785	-84.626	735	05/2000-present
	Dolly Sods WA	DOSO1	WV	39.105	-79.426	1182	09/1991-present
	Frostburg	FRRE1	MD	39.706	-79.012	767	04/2004-present
	Great Smoky Mountains NP	GRSM1	TN	35.633	-83.942	811	03/1988-present
	James River Face Wilderness	JARI1	VA	37.627	-79.513	290	06/2000-present
	Jefferson NF	JEFF1	VA	37.617	-79.483	219	09/1994-05/2000
	Linville Gorge	LIGO1	NC	35.972	-81.933	969	03/2000-present
	Shenandoah NP	SHEN1	VA	38.523	-78.435	1079	03/1988-present
	Shining Rock WA	SHRO1	NC	35.394	-82.774	1617	07/1994-present
	Sipsy Wilderness	SIPS1	AL	34.343	-87.339	286	03/1992-present
Boundary Waters	Boundary Waters Canoe Area	BOWA1	MN	47.947	-91.496	527	08/1991-present
	Isle Royale NP	ISLE1	MI	47.46	-88.149	182	11/1999-present
	Isle Royale NP	ISRO1	MI	47.917	-89.15	213	06/1988-07/1991
	Seney	SENE1	MI	46.289	-85.95	215	11/1999-present
	Voyageurs NP #1	VOYA1	MN	48.413	-92.83	426	03/1988-09/1996
	Voyageurs NP #2	VOYA2	MN	48.413	-92.829	429	11/1999-present
California Coast	Pinnacles NM	PINN1	CA	36.483	-121.157	302	03/1988-present
	Point Reyes National Seashore	PORE1	CA	38.122	-122.909	97	03/1988-present
	San Rafael	RAFA1	CA	34.734	-120.007	957	02/2000-present
Central Great Plains	Blue Mounds	BLMO1	MN	43.716	-96.191	473	07/2002-present
	Bondville	BOND1	IL	40.052	-88.373	263	03/2001-present
	Cedar Bluff	CEBL1	KS	38.77	-99.763	666	06/2002-present
	Crescent Lake	CRES1	NE	41.763	-102.434	1207	07/2002-present
	El Dorado Springs	ELDO1	MO	37.701	-94.035	298	06/2002-present
	Great River Bluffs	GRR11	MN	43.937	-91.405	370	07/2002-present
	Lake Sugema	LASU1	IA	40.688	-91.988	210	06/2002-11/2004
	Lake Sugema	LASU2	IA	40.693	-92.006	229	12/2004-present

IMPROVE Site Group	Site Name	Site Code	State	Latitude	Longitude	Elevation (m)	Dates of Operation
	Nebraska NF	NEBR1	NE	41.889	-100.339	883	07/2002-present
	Omaha	OMAH1	NE	42.149	-96.432	430	08/2003-present
	Sac and Fox	SAFO1	KS	39.979	-95.568	293	06/2002-present
	Tallgrass	TALL1	KS	38.434	-96.56	390	09/2002-present
	Viking Lake	VILA1	IA	40.969	-95.045	371	06/2002-present
Central Rockies	Brooklyn Lake	BRLA1	WY	41.366	-106.242	3196	09/1993-12/2003
	Great Sand Dunes NM	GRSA1	CO	37.725	-105.519	2498	05/1988-present
	Mount Zirkel WA	MOZI1	CO	40.538	-106.677	3243	07/1994-present
	Rocky Mountain NP HQ	RMHQ1	CO	40.362	-105.564	2408	03/1988-02/1991
	Rocky Mountain NP	ROMO1	CO	40.278	-105.546	2760	09/1990-present
	Storm Peak	STPE1	CO	40.445	-106.74	3220	12/1993-07/1994
	Wheeler Peak	WHPE1	NM	36.585	-105.452	3366	08/2000-present
	White River NF	WHRI1	CO	39.154	-106.821	3414	07/1993-present
Colorado Plateau	Arches NP	ARCH1	UT	38.783	-109.583	1722	03/1988-05/1992
	Bandelier NM	BAND1	NM	35.78	-106.266	1988	03/1988-present
	Bryce Canyon NP	BRCA1	UT	37.618	-112.174	2481	03/1988-present
	Canyonlands NP	CANY1	UT	38.459	-109.821	1798	03/1988-present
	Capitol Reef NP	CAPI1	UT	38.302	-111.293	1897	03/2000-present
	Hopi Point #1	GRCA1	AZ	36.066	-112.154	2164	03/1988-08/1998
	Hance Camp at Grand Canyon NP	GRCA2	AZ	35.973	-111.984	2267	09/1997-present
	Indian Gardens	INGA1	AZ	36.078	-112.129	1166	10/1989-present
	Meadview	MEAD1	AZ	36.019	-114.068	902	09/1991-09/1992 02/2003-present
	Mesa Verde NP	MEVE1	CO	37.198	-108.491	2172	03/1988-present
	San Pedro Parks	SAPE1	NM	36.014	-106.845	2935	08/2000-present
	Weminuche WA	WEMI1	CO	37.659	-107.8	2750	03/1988-present
	Zion Canyon	ZICA1	UT	37.198	-113.151	1215	12/2002-present
	Zion	ZION1	UT	37.459	-113.224	1545	03/2000-08/2004
Columbia River Gorge	Columbia Gorge #1	COGO1	WA	45.569	-122.21	230	09/1996-present
	Columbia River Gorge	CORI1	WA	45.664	-121.001	179	06/1993-present
Death Valley	Death Valley NP	DEVA1	CA	36.509	-116.848	130	10/1993-present
East Coast	Brigantine NWR	BRIG1	NJ	39.465	-74.449	5	09/1991-present
	Swanquarter	SWAN1	NC	35.451	-76.207	-4	06/2000-present

IMPROVE Site Group	Site Name	Site Code	State	Latitude	Longitude	Elevation (m)	Dates of Operation
Great Basin	Great Basin NP	GRBA1	NV	39.005	-114.216	2066	05/1992-present
	Jarbridge WA	JARB1	NV	41.893	-115.426	1869	03/1988-present
Hawaii	Haleakala NP	HALE1	HI	20.809	-156.282	1153	02/1991-present
	Hawaii Volcanoes NP	HAVO1	HI	19.431	-155.258	1259	03/1988-present
	Mauna Loa Observatory #1	MALO1	HI	19.536	-155.577	3439	03/1995-present
	Mauna Loa Observatory #2	MALO2	HI	19.536	-155.577	3439	03/1995-present
	Mauna Loa Observatory #3	MALO3	HI	19.539	-155.578	3400	04/1996-05/1996
	Mauna Loa Observatory #4	MALO4	HI	19.539	-155.578	3400	04/1996-05/1996
Hells Canyon	Craters of the Moon NM	CRMO1	ID	43.461	-113.555	1818	05/1992-present
	Hells Canyon	HECA1	OR	44.97	-116.844	655	08/2000-present
	Sawtooth NF	SAWT1	ID	44.17	-114.927	1990	01/1994-present
	Scoville	SCOV1	ID	43.65	-113.033	1500	05/1992-05/1997
	Starkey	STAR1	OR	45.225	-118.513	1259	03/2000-present
Lone Peak	Lone Peak WA	LOPE1	UT	40.445	-111.708	1768	12/1993-08/2001
Mid South	Caney Creek	CACR1	AR	34.454	-94.143	683	06/2000-present
	Cherokee Nation	CHER1	OK	36.956	-97.031	342	09/2002-present
	Ellis	ELLI1	OK	36.085	-99.935	697	06/2002-present
	Hercules-Glades	HEGL1	MO	36.614	-92.922	404	03/2001-present
	Sikes	SIKE1	LA	32.057	-92.435	45	03/2001-present
	Upper Buffalo WA	UPBU1	AR	35.826	-93.203	723	12/1991-present
	Wichita Mountains	WIMO1	OK	34.732	-98.713	509	03/2001-present
Mogollon Plateau	Mount Baldy	BALD1	AZ	34.058	-109.441	2509	02/2000-present
	Bosque del Apache	BOAP1	NM	33.87	-106.852	1390	04/2000-present
	Gila WA	GICL1	NM	33.22	-108.235	1776	04/1994-present
	Hillside	HILL1	AZ	34.429	-112.963	1511	04/2001-06/2005
	Ike's Backbone	IKBA1	AZ	34.34	-111.683	1298	04/2000-present
	Petrified Forest NP	PEFO1	AZ	35.078	-109.769	1766	03/1988-present
	San Andres	SAAN1	NM	32.687	-106.484	1326	10/1997-08/2000
	Sierra Ancha	SIAN1	AZ	34.091	-110.942	1600	02/2000-present
	Sycamore Canyon	SYCA1	AZ	35.141	-111.969	2046	09/1991-present
	Tonto NM	TONT1	AZ	33.655	-111.107	775	04/1988-present
	White Mountain	WHIT1	NM	33.469	-105.535	2064	01/2002-present
Northeast	Acadia NP	ACAD1	ME	44.377	-68.261	157	03/1988-present
	Addison Pinnacle	ADPI1	NY	42.091	-77.21	512	04/2001-present
	Bridgton	BRMA1	ME	44.107	-70.729	234	03/2001-present
	Casco Bay	CABA1	ME	43.833	-70.064	27	03/2001-present

IMPROVE Site Group	Site Name	Site Code	State	Latitude	Longitude	Elevation (m)	Dates of Operation
	Cape Cod	CACO1	MA	41.976	-70.024	49	04/2001-present
	Connecticut Hill	COHI1	NY	42.401	-76.653	519	04/2001-07/2006
	Great Gulf WA	GRGU1	NH	44.308	-71.218	454	06/1995-present
	Lye Brook WA	LYBR1	VT	43.148	-73.127	1015	09/1991-present
	Martha's Vineyard	MAVI1	MA	41.331	-70.785	3	01/2003-present
	Mohawk Mt.	MOMO1	CT	41.821	-73.297	522	09/2001-present
	Moosehorn NWR	MOOS1	ME	45.126	-67.266	78	12/1994-present
	Old Town	OLTO1	ME	44.933	-68.646	51	07/2001-present
	Proctor Maple Research Facility	PMRF1	VT	44.528	-72.869	401	12/1993-present
	Presque Isle	PRIS1	ME	46.696	-68.033	166	03/2001-present
	Quabbin Summit	QURE1	MA	42.298	-72.335	318	03/2001-present
Northern Great Plains	Badlands NP	BADL1	SD	43.743	-101.941	736	03/1988-present
	Cloud Peak	CLPE1	WY	44.334	-106.957	2471	06/2002-present
	Fort Peck	FOPE1	MT	48.308	-105.102	638	06/2002-present
	Lostwood	LOST1	ND	48.642	-102.402	696	12/1999-present
	Medicine Lake	MELA1	MT	48.487	-104.476	606	12/1999-present
	Northern Cheyenne	NOCH1	MT	45.65	-106.557	1283	06/2002-present
	Thunder Basin	THBA1	WY	44.663	-105.287	1195	06/2002-present
	Theodore Roosevelt	THRO1	ND	46.895	-103.378	853	12/1999-present
	UL Bend	ULBE1	MT	47.582	-108.72	891	01/2000-present
	Wind Cave	WICA1	SD	43.558	-103.484	1296	12/1999-present
Northern Rockies	Bridger WA	BRID1	WY	42.975	-109.758	2627	03/1988-present
	Cabinet Mountains	CABI1	MT	47.955	-115.671	1441	07/2000-present
	Flathead	FLAT1	MT	47.773	-114.269	1580	06/2002-present
	Gates of the Mountains	GAMO1	MT	46.826	-111.711	2387	07/2000-present
	Glacier NP	GLAC1	MT	48.511	-113.997	975	03/1988-present
	Monture	MONT1	MT	47.122	-113.154	1282	03/2000-present
	North Absaroka	NOAB1	WY	44.745	-109.382	2483	01/2000-present
	Salmon NF	SALM1	ID	45.159	-114.026	2788	12/1993-08/2000
	Sula Peak	SULA1	MT	45.86	-114	1896	08/1994-present
	Yellowstone NP 1	YELL1	WY	44.565	-110.4	2442	03/1988-07/1996
	Yellowstone NP 2	YELL2	WY	44.565	-110.4	2425	07/1996-present
Northwest	Lynden	LYND1	WA	48.953	-122.559	28	10/1996-08/1997
	Mount Rainier NP	MORA1	WA	46.758	-122.124	439	03/1988-present
	North Cascades	NOCA1	WA	48.732	-121.065	569	03/2000-present
	Olympic	OLYM1	WA	48.007	-122.973	600	07/2001-present

IMPROVE Site Group	Site Name	Site Code	State	Latitude	Longitude	Elevation (m)	Dates of Operation
	Pasayten	PASA1	WA	48.388	-119.927	1627	11/2000-present
	Snoqualmie Pass	SNPA1	WA	47.422	-121.426	1049	07/1993-present
	Spokane Res.	SPOK1	WA	47.904	-117.861	552	07/2001-06/2005
	White Pass	WHPA1	WA	46.624	-121.388	1827	02/2000-present
Not Assigned	Walker River Paiute Tribe	WARI1	NV	38.952	-118.815	1250	06/2003-11/2005
Ohio River Valley	Cadiz	CADI1	KY	36.784	-87.85	192	03/2001-present
	Livonia	LIVO1	IN	38.535	-86.26	282	03/2001-present
	Mammoth Cave NP	MACA1	KY	37.132	-86.148	235	09/1991-present
	Mingo	MING1	MO	36.972	-90.143	111	05/2000-present
	M.K. Goddard	MKGO1	PA	41.427	-80.145	380	04/2001-present
	Quaker City	QUCI1	OH	39.943	-81.338	366	05/2001-present
Oregon and Northern California	Bliss SP (TRPA)	BLIS1	CA	38.976	-120.103	2131	11/1990-present
	Crater Lake NP	CRLA1	OR	42.896	-122.136	1996	03/1988-present
	Kalmiopsis	KALM1	OR	42.552	-124.059	80	03/2000-present
	Lava Beds NM	LABE1	CA	41.712	-121.507	1460	03/2000-present
	Lassen Volcanic NP	LAVO1	CA	40.54	-121.577	1733	03/1988-present
	Mount Hood	MOHO1	OR	45.289	-121.784	1531	03/2000-present
	Redwood NP	REDW1	CA	41.561	-124.084	244	03/1988-present
	Three Sisters WA	THSI1	OR	44.291	-122.043	885	07/1993-present
	Trinity	TRIN1	CA	40.786	-122.805	1014	07/2000-present
Phoenix	Phoenix	PHOE1	AZ	33.504	-112.096	342	04/2001-present
Puget Sound	Puget Sound	PUSO1	WA	47.57	-122.312	98	03/1996-present
Sierra Nevada	Dome Lands WA	DOLA1	CA	35.699	-118.202	914	08/1994-10/1998
	Dome Lands WA	DOMI1	CA	35.728	-118.138	927	02/2000-present
	Hoover	HOOV1	CA	38.088	-119.177	2561	07/2001-present
	Kaiser	KAIS1	CA	37.221	-119.155	2598	01/2000-present
	Sequoia NP	SEQU1	CA	36.489	-118.829	519	03/1992-present
	South Lake Tahoe	SOLA1	CA	38.933	-119.967	1900	03/1989-06/1997
	Yosemite NP	YOSE1	CA	37.713	-119.706	1603	03/1988-present
Southeast	Breton	BRET1	LA	29.119	-89.207	11	06/2000-present
	Chassahowitzka NWR	CHAS1	FL	28.748	-82.555	4	04/1993-present
	Everglades NP	EVER1	FL	25.391	-80.681	1	09/1988-present
	Okefenokee NWR	OKEF1	GA	30.741	-82.128	48	09/1991-present
	Cape Romain NWR	ROMA1	SC	32.941	-79.657	5	09/1994-present
	St. Marks	SAMA1	FL	30.093	-84.161	8	06/2000-present
Southern Arizona	Chiricahua NM	CHIR1	AZ	32.009	-109.389	1555	03/1988-present

IMPROVE Site Group	Site Name	Site Code	State	Latitude	Longitude	Elevation (m)	Dates of Operation
	Douglas	DOUG1	AZ	31.349	-109.54	1230	06/2004-present
	Organ Pipe	ORPI1	AZ	31.951	-112.802	504	01/2003-present
	Queen Valley	QUVA1	AZ	33.294	-111.286	661	04/2001-present
	Saguaro NM	SAGU1	AZ	32.175	-110.737	941	06/1988-present
	Saguaro West	SAWE1	AZ	32.249	-111.218	714	04/2001-present
Southern California	Agua Tibia	AGTI1	CA	33.464	-116.971	508	11/2000-present
	Joshua Tree NP	JOSH1	CA	34.069	-116.389	1235	02/2000-present
	Joshua Tree NP	JOTR1	CA	34.069	-116.389	1228	09/1991-07/1992
	San Gabriel	SAGA1	CA	34.297	-118.028	1791	12/2000-present
	San Geronio WA	SAGO1	CA	34.194	-116.913	1726	03/1988-present
Urban QA Sites	Atlanta	ATLA1	GA	33.688	-84.29	243	04/2004-present
	Baltimore	BALT1	MD	39.255	-76.709	78	06/2004-present
	Birmingham	BIRM1	AL	33.553	-86.815	176	04/2004-present
	Chicago	CHIC1	IL	41.751	-87.713	195	11/2003-09/2005
	Detroit	DETR1	MI	42.229	-83.209	180	11/2003-present
	Fresno	FRES1	CA	36.782	-119.773	100	09/2004-present
	Houston	HOUS1	TX	29.67	-95.129	7	05/2004-09/2005
	New York City	NEYO1	NY	40.816	-73.902	45	08/2004-present
	Pittsburgh	PITT1	PA	40.465	-79.961	268	04/2004-present
	Rubidoux	RUBI1	CA	34	-117.416	248	09/2004-09/2005
Virgin Islands	Virgin Islands NP	VIIS1	VI	18.336	-64.796	51	10/1990-present
Washington D.C.	Washington D.C.	WASH1	DC	38.876	-77.034	15	03/1988-present
West Texas	Big Bend NP	BIBE1	TX	29.303	-103.178	1067	03/1988-present
	Guadalupe Mountains NP	GUMO1	TX	31.833	-104.809	1672	03/1988-present
	Salt Creek	SACR1	NM	33.46	-104.404	1072	04/2000-present

NF = National Forest

NM = National Monument

NP = National Park

NWR = National Wildlife Refuge

WA = Wilderness Area

Table 1.2. Class I areas and the representative monitoring site.

Class I Area Name	Site Name	Site Code
Acadia	Acadia NP	ACAD1
Agua Tibia	Agua Tibia	AGTI1
Alpine Lakes	Snoqualmie Pass	SNPA1
Anaconda-Pintler	Sula Peak	SULA1
Ansel Adams	Kaiser	KAIS1
Arches	Canyonlands NP	CANY1
Badlands	Badlands NP	BADL1
Bandelier	Bandelier NM	BAND1
Big Bend	Big Bend NP	BIBE1
Black Canyon of the Gunnison	Weminuche WA	WEMI1
Bob Marshall	Monture	MONT1
Bosque del Apache	Bosque del Apache	BOAP1

Class I Area Name	Site Name	Site Code
Boundary Waters Canoe Area	Boundary Waters Canoe Area	BOWA1
Breton	Breton	BRET1
Bridger	Bridger WA	BRID1
Brigantine	Brigantine NWR	BRIG1
Bryce Canyon	Bryce Canyon NP	BRCA1
Cabinet Mountains	Cabinet Mountains	CABI1
Caney Creek	Caney Creek	CACR1
Canyonlands	Canyonlands NP	CANY1
Cape Romain	Cape Romain NWR	ROMA1
Capitol Reef	Capitol Reef NP	CAPI1
Caribou	Lassen Volcanic NP	LAVO1
Carlsbad Caverns	Guadalupe Mountains NP	GUMO1
Chassahowitzka	Chassahowitzka NWR	CHAS1
Chiricahua NM	Chiricahua NM	CHIR1
Chiricahua W	Chiricahua NM	CHIR1
Cohutta	Cohutta	COHU1
Crater Lake	Crater Lake NP	CRLA1
Craters of the Moon	Craters of the Moon NM	CRMO1
Cucamonga	San Gabriel	SAGA1
Denali	Denali NP	DENA1
Desolation	Bliss SP (TRPA)	BLIS1
Diamond Peak	Crater Lake NP	CRLA1
Dolly Sods	Dolly Sods WA	DOSO1
Dome Land	Dome Lands WA	DOME1
Eagle Cap	Starkey	STAR1
Eagles Nest	White River NF	WHRI1
Emigrant	Yosemite NP	YOSE1
Everglades	Everglades NP	EVER1
Fitzpatrick	Bridger WA	BRID1
Flat Tops	White River NF	WHRI1
Galiuro	Chiricahua NM	CHIR1
Gates of the Mountains	Gates of the Mountains	GAMO1
Gearhart Mountain	Crater Lake NP	CRLA1
Gila	Gila WA	GICL1
Glacier	Glacier NP	GLAC1
Glacier Peak	North Cascades	NOCA1
Goat Rocks	White Pass	WHPA1
Grand Canyon	Hance Camp at Grand Canyon NP	GRCA2
Grand Teton	Yellowstone NP 2	YELL2
Great Gulf	Great Gulf WA	GRGU1
Great Sand Dunes	Great Sand Dunes NM	GRSA1
Great Smoky Mountains	Great Smoky Mountains NP	GRSM1
Guadalupe Mountains	Guadalupe Mountains NP	GUMO1
Haleakala	Haleakala NP	HALE1
Hawaii Volcanoes	Hawaii Volcanoes NP	HAVO1
Hells Canyon	Hells Canyon	HECA1
Hercules-Glade	Hercules-Glades	HEGL1
Hoover	Hoover	HOOV1
Isle Royale	Isle Royale NP	ISLE1
James River Face	James River Face WA	JARI1
Jarbidge	Jarbidge WA	JARB1
John Muir	Kaiser	KAIS1

Class I Area Name	Site Name	Site Code
Joshua Tree	Joshua Tree NP	JOSH1
Joyce Kilmer-Slickrock	Great Smoky Mountains NP	GRSM1
Kaiser	Kaiser	KAIS1
Kalmiopsis	Kalmiopsis	KALM1
Kings Canyon	Sequoia NP	SEQU1
La Garita	Weminuche WA	WEMI1
Lassen Volcanic	Lassen Volcanic NP	LAVO1
Lava Beds	Lava Beds NM	LABE1
Linville Gorge	Linville Gorge	LIGO1
Lostwood	Lostwood	LOST1
Lye Brook	Lye Brook WA	LYBR1
Mammoth Cave	Mammoth Cave NP	MACA1
Marble Mountain	Trinity	TRIN1
Maroon Bells-Snowmass	White River NF	WHRI1
Mazatzal	Ike's Backbone	IKBA1
Medicine Lake	Medicine Lake	MELA1
Mesa Verde	Mesa Verde NP	MEVE1
Mingo	Mingo	MING1
Mission Mountains	Monture	MONT1
Mokelumne	Bliss SP (TRPA)	BLIS1
Moosehorn	Moosehorn NWR	MOOS1
Mount Adams	White Pass	WHPA1
Mount Baldy	Mount Baldy	BALD1
Mount Hood	Mount Hood	MOHO1
Mount Jefferson	Three Sisters WA	THSI1
Mount Rainier	Mount Rainier NP	MORA1
Mount Washington	Three Sisters WA	THSI1
Mount Zirkel	Mount Zirkel WA	MOZI1
Mountain Lakes	Crater Lake NP	CRLA1
North Absaroka	North Absaroka	NOAB1
North Cascades	North Cascades	NOCA1
Okefenokee	Okefenokee NWR	OKEF1
Olympic	Olympic	OLYM1
Otter Creek	Dolly Sods WA	DOSO1
Pasayten	Pasayten	PASA1
Pecos	Wheeler Peak	WHPE1
Petrified Forest	Petrified Forest NP	PEFO1
Pine Mountain	Ike's Backbone	IKBA1
Pinnacles	Pinnacles NM	PINN1
Point Reyes	Point Reyes National Seashore	PORE1
Presidential Range-Dry River	Great Gulf WA	GRGU1
Rawah	Mount Zirkel WA	MOZI1
Red Rock Lakes	Yellowstone NP 2	YELL2
Redwood	Redwood NP	REDW1
Rocky Mountain	Rocky Mountain NP	ROMO1
Roosevelt Campobello	Moosehorn NWR	MOOS1
Saguaro	Saguaro NM	SAGU1
Saint Marks	St. Marks	SAMA1
Salt Creek	Salt Creek	SACR1
San Gabriel	San Gabriel	SAGA1
San Geronio	San Geronio WA	SAGO1
San Jacinto	San Geronio WA	SAGO1

Class I Area Name	Site Name	Site Code
San Pedro Parks	San Pedro Parks	SAPE1
San Rafael	San Rafael	RAFA1
Sawtooth	Sawtooth NF	SAWT1
Scapegoat	Monture	MONT1
Selway-Bitterroot	Sula Peak	SULA1
Seney	Seney	SENE1
Sequoia	Sequoia NP	SEQU1
Shenandoah	Shenandoah NP	SHEN1
Shining Rock	Shining Rock WA	SHRO1
Sierra Ancha	Sierra Ancha	SIAN1
Simeonof	Simeonof	SIME1
Sipsy	Sipsy WA	SIPS1
South Warner	Lava Beds NM	LABE1
Strawberry Mountain	Starkey	STAR1
Superstition	Tonto NM	TONT1
Swanquarter	Swanquarter	SWAN1
Sycamore Canyon	Sycamore Canyon	SYCA1
Teton	Yellowstone NP 2	YELL2
Theodore Roosevelt	Theodore Roosevelt	THRO1
Thousand Lakes	Lassen Volcanic NP	LAVO1
Three Sisters	Three Sisters WA	THSI1
Tuxedni	Tuxedni	TUXE1
UL Bend	UL Bend	ULBE1
Upper Buffalo	Upper Buffalo WA	UPBU1
Ventana	Pinnacles NM	PINN1
Virgin Islands	Virgin Islands NP	VIIS1
Voyageurs	Voyageurs NP #2	VOYA2
Washakie	North Absaroka	NOAB1
Weminuche	Weminuche WA	WEMI1
West Elk	White River NF	WHRI1
Wheeler Peak	Wheeler Peak	WHPE1
White Mountain	White Mountain	WHIT1
Wichita Mountains	Wichita Mountains	WIMO1
Wind Cave	Wind Cave	WICA1
Wolf Island	Okefenokee NWR	OKEF1
Yellowstone	Yellowstone NP 2	YELL2
Yolla Bolly-Middle Eel	Trinity	TRIN1
Yosemite	Yosemite NP	YOSE1
Zion	Zion	ZION1

NF = National Forest

NM = National Monument

NP = National Park

NWR = National Wildlife Refuge

WA = Wilderness Area

1.2.2 Aerosol Sampling and Analysis

The current IMPROVE protocol for particle sampling requires that the sampler operate at ambient temperatures. To accomplish this, samplers are generally housed in a ventilated shelter that provides shielding from direct sunlight. Shelter design across the network is varied to meet differing practical and aesthetic concerns for specific sites.

A few protocol changes with respect to the particle monitoring in the network were implemented as part of the expansion to make the IMPROVE network more consistent with the EPA's fine mass and fine speciation particulate monitoring networks and to add additional quality control measures. The primary changes included changing the twice-weekly, 24-hour duration sampling schedule to an every-third-day schedule that corresponds to the schedule of the national particulate networks operated by state and local governments and the addition of replicate sampling and analysis for PM_{2.5} mass and composition to evaluate measurement uncertainty. A new version of the IMPROVE particle sampler was designed and produced at the University of California, Davis, to allow for these protocol changes. The version I sampler is described in previous IMPROVE-related publications [Malm et al., 1989; Malm et al., 1994; Malm et al., 2000], and the version II IMPROVE sampler is described below. Installation of the version II samplers at all 110 IMPROVE sites, new and existing, began in November 1999 and continued through the spring of 2000. All sites installed since 2000 have the version II sampler.

The IMPROVE samplers (versions I and II) consist of four independent modules (see Figure 1.3). Each module incorporates a separate inlet, filter pack, and pump assembly. It is convenient to consider a particular module, its associated filter, and the parameters measured from the filter as a channel of measurement (e.g., module A). Modules A, B, and C are equipped with a 2.5 µm cyclone, while module D is fitted with a PM₁₀ inlet. For module B, the sampled air is drawn through a carbonate denuder tube in the inlet to remove gaseous nitric acid.

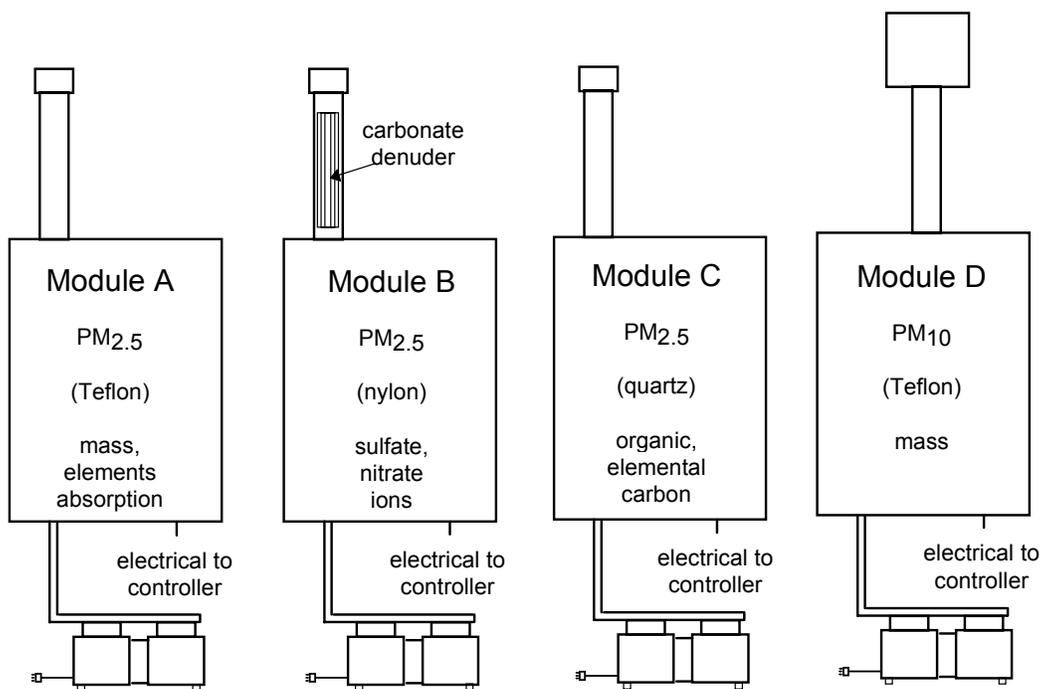


Figure 1.3. Schematic view of the IMPROVE sampler showing the four modules with separate inlets and pumps. The substrates with analyses performed for each module are also shown.

For the version II sampler, the objective was to build a sampler that would be comparable to the version I sampler from a sample collection perspective but use then state-of-the-art microprocessor technology to increase the control and provide feedback on operating status. The version II sampler was designed to be identical to the version I sampler in the design of the four

sampling modules (shown in Figure 1.3), including using the same sample collection substrates (filter materials) and the same materials and dimensions for each module, from the sample inlet to the face of the filter, and with the same flow rates. Preliminary tests of the samplers confirmed the qualitative comparability of the aerosol samples collected via the version I and II samplers [Malm et al., 2000; Eldred et al., 2001].

One of the improvements in the version II sampler is a microprocessor-based controller that can be programmed to sample any period of time on any schedule, which replaced the 7-day timer/controller. The microprocessor includes a memory card reader/writer that is used to record flow rate, sample temperature, and other performance-related information reported every 15 minutes throughout the sample period. For the original version I sampler, the flows were manually checked at the beginning and end of each sample period, and the seasonal mean site temperature and pressure were used for flow calculations. Beyond the improved tracking and calculation of flow and sample air volume, the microprocessor also permits programming changes to be distributed to the controller on chips that are installed during annual maintenance visits. This allows for programming changes to be implemented consistently and without requiring programming in the field.

To accommodate the new sampling schedule, the version II sampler has a four-filter manifold for each module, in place of the version I sampler two-filter manifolds. The manifold with the solenoids sits directly above the filter cassettes and is raised or lowered as a unit to unload and load the filters. The four filter cassettes are held in a cartridge (shown in Figure 1.4) that is designed to only allow one orientation in the sampler. Fully prepared date- and site-labeled filter cartridges, along with memory cards, are sent from the analysis laboratory to the field and are returned in special mailing containers to prevent confusion concerning the order of sampling among the filters. If filter change service is performed on a sample day, the operator moves the cassette containing that day's filter to the open position in the newly loaded cartridge. The few minutes that it takes to perform this sample change is recorded by the microprocessor on the memory card so that the correct air volume is used to calculate concentrations.

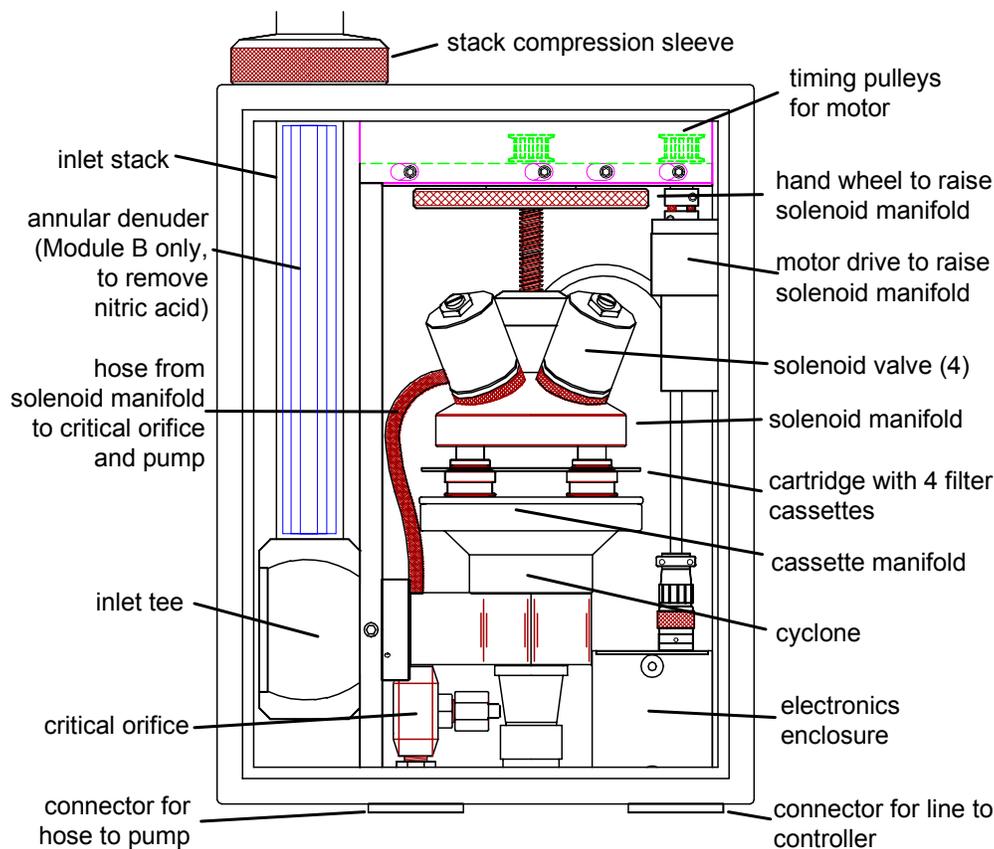


Figure 1.4. Schematic of a new version of the IMPROVE sampler PM_{2.5} module.

Additionally, the version II IMPROVE sampler makes it simple to add a fifth module at the monitoring sites to accommodate replicate sampling and analysis for mass and composition. This quality assurance module will be operated for each sample period and will collect a replicate sample for one of the four modules (A, B, C, or D) so that, over time, relative precision information can be developed for each parameter. Starting in 2003, collocated modules were installed at 24 sites across the network, providing ~4% replication for each of the four modules (Table 1.3).

Table 1.3. Sites with a fifth collocated module.

Site Name	Site	A	B	C	D	Start Date	End Date
Mesa Verde NP	MEVE1	X				8/13/2003	
Olympic NP	OLYM1	X				11/8/2003	
Proctor Maple Research Facility	PMRF1	X				9/3/2003	
Sac and Fox	SAFO1	X				11/20/2003	
St. Marks	SAMA1	X				11/18/2004	
Trapper Creek	TRCR1	X				6/22/2004	
Big Bend NP	BIBE1		X			8/30/2003	
Blue Mounds	BLMO1		X			9/16/2004	
Frostburg	FRRE1		X			4/15/2004	
Gates of the Mountains	GAMO1		X			9/23/2003	
Lassen Volcanic NP	LAVO1		X			4/18/2003	
Mammoth Cave NP	MACA1		X			5/12/2003	

Site Name	Site	A	B	C	D	Start Date	End Date
Everglades NP	EVER1			X		7/11/2003	
Hercules-Glades	HEGL1			X		8/24/2004	
Hoover	HOOV1			X		8/13/2003	
Medicine Lake	MELA1			X		9/25/2003	
Saguaro West	SAWE1			X		3/25/2004	
Seney	SENE1			X		8/10/2003	
Houston	HOUS1				X	4/30/2004	9/1/2005
Jarbidge WA	JARB1				X	6/30/2004	
Joshua Tree NP	JOSH1				X	8/7/2003	
Quabbin Summit	QURE1				X	9/4/2003	
Swanquarter	SWAN1				X	11/9/2004	
Wind Cave	WICA1				X	9/17/2004	

NP = National Park

WA = Wilderness Area

In both the version I and II samplers, the four modules independently collect aerosol samples onto a variety of filter types (Figure 1.3). The D module collects PM₁₀ aerosol on Teflon filters. The A, B, and C modules collect PM_{2.5} aerosol on Teflon, nylon, and quartz fiber filters, respectively. The different filter media facilitate the collection of particular aerosol species or a specific form of chemical analysis. Gravimetric analysis is routinely performed on the A and D module filters. Elemental analysis and aerosol absorption measurements are routinely performed on the A module filter. Ion analysis is routinely performed on the B module filter, and carbon analysis is routinely performed on the quartz fiber filter.

The gravimetric analysis performed on both the PM_{2.5} A and PM₁₀ D module Teflon filters allows for the estimation of the coarse aerosol fraction through differencing. Teflon filters are prone to losses of semivolatile NH₄NO₃ and thus provide only a lower estimate of the actual ambient aerosol concentrations [Hand and Malm, 2006]. Unfortunately, nylon filters that effectively capture the NH₄NO₃ are not ideal for gravimetric analysis because they are heavier than Teflon filters, and thus the calculation of aerosol mass through pre- and post-weighing is more error prone since aerosol mass represents a smaller fraction of the total mass. The quartz filters are not suitable for gravimetric analysis because they are more friable and subject to the same NH₄NO₃ losses as Teflon. A further complication on the interpretation of the gravimetric measurements is the variable size cut on the PM_{2.5} modules. The size cut of the cyclone used to collect and separate PM_{2.5} aerosols is controlled by the flow rate of the sampler. Variations in the flow rate can have impacts on the size range of aerosols being collected and termed “fine”. This issue can be important when interpreting fine mass versus coarse mass, as well as affect the chemical composition of the fractions since different aerosol types tend to be dominant in each size fraction.

The forms of elemental analysis conducted past and present in the IMPROVE network are proton elastic scattering analysis (PESA), proton induced X-ray emission (PIXE), and X-ray fluorescence (XRF). Since the network’s inception, PESA has been and continues to be used for quantifying elemental hydrogen. PIXE has been used for quantifying nearly all elements with atomic weights ≥11 (Na) and ≤82 (Pb). Beginning in 1992, analysis of the heavier elements, those with atomic weights from 26 (Fe) to 82 (Pb), was changed from PIXE to XRF with a Mo anode source. PIXE was discontinued in late 2001 and analysis of the lighter elements with atomic weights from 11 (Na) to 25 (Mn) was changed from PIXE to XRF using a Cu anode

source. Also, in late 2001, the analysis of Fe was changed from Mo anode XRF to Cu anode XRF. In both cases the change from PIXE to XRF provided lower minimum detection limits (mdl) for most elements of interest, as well as better sample preservation for reanalysis. The exceptions were Na, Mg, Al, and to a lesser extent Si, where the change to Cu XRF resulted in significantly increased mdl and uncertainty. The details on the transitions from PIXE to XRF are provided in section 1.3 below.

The material collected from the B module nylon filters is extracted ultrasonically in an aqueous solution that is subsequently analyzed by ion chromatography for the anions sulfate, nitrate, nitrite, and chloride. Nylon filters have been shown to be more effective at capturing and retaining NO₃ from semivolatile NH₄NO₃ than Teflon filters [Yu et al., 2005]. Field blanks for the B module are collected to correct for positive artifacts of all the reported anions. A field blank nylon filter is placed in an unused port in the filter cassette, so it is exposed to all aspects of the filter handling process, except it does not have sample air drawn through it [McDade et al., 2004]. Approximately 70 field blanks are collected each month, constituting around 4% of the total filters collected [McDade et al., 2004]. Each site receives a nylon filter field blank every two to three months, on average [McDade et al., 2004]. A single artifact correction is applied for each species for every site in the network for the time period being processed. Currently, the data are processed in monthly batches; prior to June 2002, seasonal quarters were used. The artifact correction is calculated as the median of the filter blank values and is subtracted from all reported concentrations. Analysis of artifacts on the nylon filter suggests that sulfate ion artifacts are typically less than 10% of the ambient concentration, and nitrate artifacts range between 10% and 20% for the filters used prior to 2004 [McDade et al., 2004]. The filters introduced in 2004 were significantly cleaner, with typical median blank values of 0.00 (below the mdl) for sulfate and nitrate and 0.01 µg/m³ for chloride, which is approximately 100 times smaller than the chloride blank values observed prior to 2004.

Module C utilizes quartz fiber filters that are analyzed by thermal optical reflectance (TOR) for particulate organic and elemental carbon (OC and EC, respectively) [Chow et al., 1993]. Quartz filter field blanks are also collected at approximately the same frequency as for nylon filters, and they are analyzed for the components of OC and EC. These field blanks are examined routinely to identify potential problems, but they are not used for artifact corrections in the IMPROVE database. In the IMPROVE program, secondary filters (after-filters) are used to correct for positive artifacts resulting from the adsorption of organic gases onto the filter. The after-filters are placed directly behind the primary quartz filter and are collected at six sites, Chiricahua, AZ; Grand Canyon, AZ; Yosemite, CA; Okefenokee, GA; Shenandoah, VA; and Mount Rainier, WA, to determine the artifact corrections for OC and EC. The Chiricahua and Okefenokee sites were added in 2001. The number of sites is limited by financial and logistical constraints and was selected to cover a variety of regions and aerosol conditions. The quartz after-filters are collected during every sampling period at the six after-filter sites. Typical artifacts for OC can correspond to half of the reported ambient concentration [McDade et al., 2004]. Negative artifacts due to the volatilization of particulate organics are not accounted for because they are thought to be small [Turpin et al., 2000], although some studies suggest they could be important [Hand and Malm, 2006].

1.2.3 Optical Sampling and Analysis

Optical monitoring is conducted at a subset of the IMPROVE sites (Tables 1.4 and 1.5). Routine optical monitoring includes light extinction as measured by transmissometers and aerosol scattering as measured by nephelometers.

Transmissometers are calibrated to measure the irradiance, at a wavelength of 550 nm, of a light source after the light has traveled over a finite atmospheric path. The transmittance of the path is calculated by dividing the measured irradiance at the end of the path by the calibrated initial intensity of the light source. Bouger's law is applied to calculate the extinction. Because of the relatively clean atmospheres found in the western United States, path lengths of a few kilometers are required to achieve the necessary sensitivity to resolve extinctions near the Rayleigh limit.

The transmissometers used in IMPROVE are the Optec, Inc., LPV-2 instruments, which have been in use since 1986. Their use in remote locations such as national parks is discussed by Molenaar et al. [1989], while their use in urban settings is presented by Dietrich et al. [1989]. Data processing algorithms that incorporate corrections for interferences are thoroughly discussed by Molenaar and Malm [1992].

Molenaar et al. [1989] discuss the inherent uncertainties associated with the measurement. The accuracy of the transmission measurement, as determined by field and laboratory calibrations, is better than 1%. However, the accuracy of the derived extinction is dependent on the accuracy of the transmission measurement in field conditions. The transmission calculation is determined from an absolute (as opposed to relative) measurement of irradiance of a light source of known intensity that is located some known distance from the receiver. The measurement is made through optics that are exposed to the ambient atmosphere but are assumed to be free of dust or other films that tend to build up on the optical surfaces. The uncertainties associated with these parameters contribute to the overall uncertainty of the measurement. For a typical 5-km path length, the estimated uncertainty is about 4 Mm^{-1} .

Table 1.4. Transmissometer receiver and transmitter locations.

Location	Site Name	Receiver Lon (deg)	Lat (deg)	Elevation (m)	Bearing (deg)	Transmitter Lon (deg)	Lat (deg)	Elevation (m)	Mean Elevation	Elevation Angle (deg)	Distance	Start Date	End Date	Sponsor
ACAD1	Acadia NP	-68.26	44.37	134	134	-68.23	44.35	466	300	5	4	9/1/1987	8/31/1993	NPS
BADL1	Badlands NP	-101.9	43.79	806	239	-101.95	43.77	805	805	-0.01	4.151	12/1/1987		NPS
BAND1	Bandelier NM	-106.26	35.78	2011	315	-106.3	35.81	2143	2077	1.65	4.058	9/1/1988		NPS
BRID1	Bridger WA	-109.79	42.93	2390	11	-109.77	42.97	2568	2479	2.01	5.083	9/1/1988		USFS
CANY1	Canyonlands NP	-109.82	38.46	1806	73	-109.75	38.48	1774	1790	-0.29	6.426	12/1/1986		NPS
CHIR2	Chiricahua NM	-109.39	32.01	1567	97	-112.54	32.01	1682	1625	2.07	3.18	12/1/1998		NPS
CHIR1	Chiricahua NM	-109.39	32.01	1567	84	-109.32	32.01	2235	1901	6.26	6.123	12/1/1988	2/28/1999	NPS
GLAC1	Glacier NP	-113.94	48.56	968	232	-113.99	48.53	975	972	0.08	5.276	12/1/1988		NPS
GRBA1	Great Basin NP	-114.21	38.99	2130	315	-114.24	39.02	2365	2248	3.44	3.913	9/1/1992		NPS
GRCA1	Grand Canyon NP	-111.99	36	2256	81	-111.93	36.01	2170	2213	-0.85		12/1/1986		NPS
	Grandview (on the rim)													
GRCW1	Grand Canyon NP	-112.12	36.07	2145	205	-112.09	36.11	755	1450	-15.78	5.11	12/1/1989		NPS
	Yavapai (in canyon)													
GUMO1	Guadalupe Mountains NP	-104.81	31.83	1616	249	-104.86	31.82	1317	1467	-3.53	4.858	12/1/1988		NPS
PEFO1	Petrified Forest NP	-109.77	35.08	1772	173	-109.75	34.94	1690	1731	-0.3	15.44	8/1/1987	8/31/1987	NPS
PEFO2	Petrified Forest NP	-109.8	34.9	1690	48	-109.75	34.95	1700	1695	0.1	5.938	6/1/1987		NPS
PINN1	Pinnacles NM	-121.15	36.47	448	317	-121.18	36.5	428	438	-0.25	4.799	3/1/1988	8/31/1993	NPS
ROMO1	Rocky Mountain NP	-105.58	40.36	2535	305	-105.63	40.39	2932	2734	4.31	5.274	12/1/1987	8/31/1997	NPS
ROMO2	Rocky Mountain NP	-105.58	40.37	2502	302	-105.63	40.39	2932	2717	5.01	4.921	9/1/1998		NPS

Location	Site Name	Receiver Lon (deg)	Lat (deg)	Elevation (m)	Bearing (deg)	Transmitter Lon (deg)	Lat (deg)	Elevation (m)	Mean Elevation	Elevation Angle (deg)	Distance	Start Date	End Date	Sponsor
SAGO1	San Gorgonio WA	-116.91	34.19	1710	211	-116.94	34.16	1731	1721	0.29	4.099	3/1/1988		USFS
SHEN2	Shenandoah NP	-78.43	38.51	1073	310	-78.44	38.52	1061	1717	-0.49	1.412	6/1/1991		NPS
TONT1	Tonto NM	-111.03	33.62	733	115	-111.11	33.65	786	760	0.42	7.203	3/1/1989	8/31/1991	USFS
YELL1	Yellowstone NP	-110.69	44.97	1836	125	-110.65	44.95	1951	1894	1.54	4.285	6/1/1989	8/31/1993	NPS
YOSE2	Yosemite NP	-119.7	37.71	1608	236	-119.75	37.69	1475	1542	-1.71	4.468	12/1/1994		NPS
YOSE1	Yosemite NP	-119.7	37.71	1608	242	-119.73	37.7	1370	1489	-5.04	2.711	9/1/1988	11/30/1994	NPS

NM = National Monument

NP = National Park

WA = Wilderness Area

Integrating nephelometers measure the scattering of light over a defined band of visible wavelengths from an enclosed volume of air. Historically, integrating nephelometers used in most major field studies have underestimated scattering because of

1. modification of the ambient aerosol by heating when a large fraction of the sampled aerosol is hygroscopic;
2. inlet, sampling train, and optical chamber designs that limit the size of particles that make it into the sampling chamber;
3. optical geometry that causes a truncation of the true scattering volume;
4. and electronics that display large nonlinear drifts in zero and span values.

The Optec NGN-2 ambient integrating nephelometer was developed to minimize these limitations. The instrument, which measures light scattering at an effective wavelength of 550 nm, is described in some detail by Molenaar et al. [1989]. It is an “open air” design that has minimal heating characteristics, and because it is open it allows a larger distribution of particle sizes to pass through the instrument. However, the cutpoint of the instrument has not been characterized. It is also designed with solid-state electronics that are very stable over wide temperature and humidity shifts. It still has an inherent limitation of an abbreviated acceptance angle in that it only samples light scattered between 5° and 175°. Calibration of the instrument and data validation and processing algorithms are also discussed in detail in Molenaar and Malm [1992]. Unlike transmissometers, where an uncertainty in transmittance leads to an additive error in extinction, uncertainties in nephelometer calibration lead to a multiplicative error in measured scattering. Typical uncertainties for the Optec instrument are on the order of 5–10% [Molenaar and Malm, 1992].

During high humidity and precipitation events, the nephelometer can report erroneously high scattering values. This is due to water condensing on the walls of the nephelometer and spray from rain drops impacting the screen on the nephelometer inlet. This water collects in the light trap and reflects light directly into the scattered-light detector, causing extremely high readings. In order to minimize this problem, the door of the nephelometer closes during heavy precipitation events, and a wick was added to the light trap to facilitate the removal of any collected water.

Table 1.5. IMPROVE nephelometer network site locations.

Site	Code	State	Latitude	Longitude	Elevation	Dates of Operation
Upper Buffalo WA	UPBU1	AR	35.83	-93.20	722	12/1991-present
Muleshoe Ranch	MUSR1	AZ	32.35	-110.23	1402	07/1997-present
Rucard Canyon	RUCA1	AZ	31.78	-109.30	1637	02/1997-05/2001
Indian Gardens	INGA1	AZ	36.08	-112.13	1166	10/1989-present
Sycamore Canyon	SYCA1	AZ	35.14	-111.97	2046	09/1991-07/1992
Hance Camp at Grand Canyon NP	GRCA2	AZ	35.97	-111.98	2267	09/1997-present
Sierra Ancha	SIANI	AZ	34.09	-110.94	1600	02/2000-present
McFadden Peak	MCFD1	AZ	34.00	-111.00	2175	10/1997-02/2000
Phoenix	PHON1	AZ				12/1996-present
Estrella Mountain Regional Park	ESTR1	AZ	33.39	-112.38	290	
Petrified Forest NP	PEFO3	AZ	34.91	-109.80	1690	

Site	Code	State	Latitude	Longitude	Elevation	Dates of Operation
Tucson Central	TUCN1	AZ			762	04/1997-present
Tucson Mountain #1	TUMO1	AZ	32.28	-111.17	754	12/1996-present
Ike's Backbone	IKBA1	AZ	34.34	-111.68	1297	04/2000-present
Humble Mountain	HUMB1	AZ	33.98	-111.78	1586	03/1997-present
Mazatzal	MAZA1	AZ	33.91	-111.43	2164	03/1997-08/2000
Tucson	CRAY1	AZ	32.20	-110.88	1707	
Greer Arizona	GRER1	AZ	34.07	-109.43	2513	
Tucson Mountain #2	TUMO2	AZ				
Lake Tahoe Blvd.	LTBV1	CA	38.95	-118.04	1902	
Bliss SP (Tahoe Regional Planning Agency)	BLIS1	CA	38.98	-120.10	2130	11/1990-present
Mount Zirkel WA	MOZI1	CO	40.54	-106.68	3243	07/1994-present
Okefenokee NWR	OKEF1	GA	30.74	-82.13	48	09/1991-present
Cedar Bluff	CEBL1	KS	38.77	-99.76	665	
Mammoth Cave NP	MACA1	KY	37.13	-86.15	235	09/1991-present
Acadia NP	ACAD1	ME	44.38	-68.26	157	03/1988-present
Seney NWR	SENY1	MI	46.29	-84.05	216	
Boundary Waters Canoe Area	BOWA1	MN	47.95	-91.50	526	08/1991-present
Shining Rock WA	SHRO1	NC	35.39	-82.77	1617	07/1994-present
Great Gulf WA	GRGU1	NH	44.31	-71.22	453	06/1995-present
Brigantine NWR	BRIG1	NJ	39.47	-74.45	5	09/1991-present
Gila WA	GICL1	NM	33.22	-108.24	1775	04/1994-present
Jarbridge WA	JARB1	NV	41.89	-115.43	1869	03/1988-present
Quaker City	QUAK1	OH	39.94	-80.66	372	01/1900-present
Wichita Mountains	WIMO1	OK	34.73	-98.71	509	03/2001-present
Three Sisters WA	THSI1	OR	44.29	-122.04	885	07/1993-present
Cape Romain NWR	ROMA1	SC	32.94	-79.66	4	09/1994-present
Great Smoky Mountains NP	GRSM1	TN	35.63	-83.94	810	03/1988-present
Big Bend NP	BIBE1	TX	29.30	-103.18	1066	03/1988-present
Lone Peak WA	LOPE1	UT	40.44	-111.71	1768	12/1993-present
James River Face WA	JARI1	VA	37.63	-79.51	289	06/2000-present
Shenandoah NP	SHEN1	VA	38.52	-78.43	1079	03/1988-present
Virgin Islands NP	VIIS1	VI	18.34	-64.80	51	10/1990-present
Lye Brook WA	LYBR1	VT	43.15	-73.13	1015	09/1991-present
Snoqualmie Pass	SNPA1	WA	47.42	-121.43	1049	07/1993-present
Columbia River Gorge	CORI1	WA	45.66	-121.00	178	06/1993-present
Mount Rainier NP	MORA1	WA	46.76	-122.12	439	03/1988-present
Columbia River Gorge #2	COGO2	WA	45.57	-122.21	243	
Mayville	MAYV1	WI	43.44	-87.47	306	
Dolly Sods WA	DOSO1	WV	39.11	-79.43	1182	09/1991-present
Green River Visibility Study	GRVS1	WY	41.84	-109.61	1950	06/1996-10/2000

NP = National Park

NWR = National Wildlife Refuge

SP = State Park

WA = Wilderness Area

1.3 PROTOCOL AND EQUIPMENT CHANGES

While consistency through time is critical to a monitoring program interested in trends, changes in protocol are inevitable. Significant changes in sampling, analysis, and data processing have occurred in the history of the IMPROVE network. Most of the changes were

implemented to improve the quality or usefulness of the IMPROVE data set or to increase the overall effectiveness of the network within available resources. Some of the key changes, including the reasoning behind the decision and the ramifications for the IMPROVE data set, are described below and listed in Table 1.6.

1.3.1 Analytical Changes

1.3.1.1. Transition from PIXE to XRF

Elemental analysis was transitioned from proton induced X-ray emission (PIXE) to X-ray fluorescence (XRF) in two stages. The initial transition was elected to lower the mdl of parameters important to aerosol source apportionment. The first transition from PIXE to XRF using a Mo anode occurred in mid 1992 and applied to the analysis of elements with atomic weights from Fe to Pb. The second transition from PIXE to XRF using a Cu anode occurred in late 2001 and applied to the analysis of the lighter elements with atomic weights from Na to Mn. Also, in late 2001, the analysis of Fe was changed from Mo anode XRF to Cu anode XRF. These transitions had both positive and negative impacts on the data quality of the elemental measurements.

One of the positives was the improved detection limits for most elements of interest. Another positive development was the decreased filter degradation with the XRF system as compared to PIXE and PESA. The proton beam used for PIXE and PESA weakens the bonds in the Teflon filters. Over long exposures and high doses, the samples become brittle and will fall apart from small disturbances such as applying vacuum or vibration. Dependent on the filter loadings, a sample can be destroyed with as little as 100 seconds of proton exposure at 50 nano amps, which is a typical exposure condition for rural IMPROVE samples. This filter destruction places a limitation on the PIXE and PESA quality control system because it prevents the repeated reanalysis of the same samples. The option of reanalyzing the same batch of filters numerous times, either as part of a precision study or over time as a check on calibration drift, is a significant addition to the quality control measures of the IMPROVE program.

A negative impact of the move to XRF as compared to PIXE was poorer quantification of the lightest elements Na, Mg, and Al. This is because the number of X-rays detected for Na, Mg, Al, and to a lesser extent Si using the Cu XRF system is much lower than with PIXE. The physical configuration and operating procedures for the XRF systems have continued to evolve to address quality issues as they are identified.

1.3.1.2. Alternate Nylon Filter Extraction Procedure

The filter extraction process for ion analysis was changed from the basic anion eluent to deionized water. This was first done for three sites in 1997 to allow for NH_4^+ analysis (Table 1.6). Starting in 2001, deionized water was used for all sites for the same purpose. The transition dates and the affected sites are detailed in Table 1.6. Recent studies have shown that both extraction solutions are equally effective at extracting particulate nitrate from nylon filters when sonication is used [Yu et al., 2005].

1.3.2 Sampling Equipment Changes

1.3.2.1. Transition from Version I to Version II IMPROVE Sampler

As described in section 1.2, the IMPROVE sampler was modified to accommodate the transition from the twice-weekly, 24-hour duration sampling schedule to an every-third-day schedule, the addition of replicate sampling, and analysis for PM_{2.5} mass and composition to evaluate precision. The new schedule corresponds to the schedule of the EPA's national particulate network operated by state and local governments. The updated sampler is comparable from a sample collection perspective but uses microprocessor technology to increase control and provide feedback on operating status. The changes were implemented into the network through the installation of version II samplers during late 1999 through early 2001.

1.3.2.2. Denuder Coating Modified

The module B denuder coating was altered in 1996 to include glycerin to maintain the efficiency of the denuder for capturing SO₂ and HNO₃ gases for the entire year in which each denuder is deployed. The glycerin was expected to keep the denuder wet and thereby more reactive. Recent studies have shown that qualitatively comparable nitrate concentrations are collected with both the original and current denuder coatings [Ashbaugh et al., 2004; Yu et al., 2005]. The IMPROVE sampler B module inlet/denuder sampling train has been exposed to known concentrations of nitric acid in the laboratory, and nitric acid removal efficiencies have been shown to lie consistently between 98% and 99%.

1.3.2.3. Changes in Nylon Filter Size

Larger nylon filters (47 mm) were initially used to ensure that the pressure drop at the filter, which can impact sampler flow rate and thereby the cutpoint for the sample, was not too high. Improved filter quality allowed the move to smaller 25 mm filters in 1994 that were consistent in size with the other modules and, due to the smaller size, had smaller artifact corrections due to manufacturing contamination, all without negative impacts on pressure drop. In 1996, procuring nylon filters of sufficient quality at the 25 mm size became difficult. With the development of the version II sampler (deployment in 2000–2001), it was decided to increase the nylon filter size to 37 mm so that the sampler would experience less pressure drop. Tests involving collocated samples with 25 and 37 mm diameter filters showed qualitatively comparable nitrate concentrations [McDade et al., 2004].

1.3.2.4. Changes in Nylon Filter Manufacturer

Prior to 1996, IMPROVE purchased nylon filters from Pall/Gelman. However, since Pall/Gelman ceased manufacturing those filters in 1996, IMPROVE transitioned to MSI/Osmonics. Unfortunately, the MSI/Osmonics filters exhibited increased and inconsistent contamination levels of all major ions (especially chloride) as compared to the Pall/Gelman filters. Pall/Gelman resumed manufacturing nylon filters, and after testing at Crocker Nuclear Laboratory (CNL) confirmed that the pressure drops and artifact-corrected ambient concentrations were statistically equivalent to the MSI/Osmonics filters, IMPROVE transitioned back to Pall/Gelman filters in January of 2004. The new Pall/Gelman filters have significantly

lower monthly median artifact values for all major ions as compared to both the MSI/Osmonics filters and the original Pall/Gelman filters.

1.3.3 Data Processing Changes

1.3.3.1. Change in the Reporting of Gravimetric Measurements

Beginning in 2002, it was decided that gravimetric measurements below measurement detection limit, even those less than 0, would be retained in the data set so as not to bias statistical analyses. With the resubmission of data for the period 2000–2004 in October 2005, below mdl gravimetric measurements were added back into the IMPROVE database for that period. This change made the decision-making process for the reporting of the gravimetric data set consistent with the speciation analyses.

1.3.3.2. Change in Batch Size Used in Data Processing Routines at CNL

In 2002 it was decided to change the batch size used in the data processing routines—blank corrections, data validation, and reporting—from seasonal quarters to months. The transition from seasonal quarters to months allowed for the release of a calendar year of data as soon as the December data were fully validated. The downsides of this protocol change include less robust blank correction and uncertainty statistics, noncompatibility with the existing filter storage system, and data management system inefficiencies in the XRF lab.

1.3.3.3. Change in Flow Rate Validation Flag Definitions

In 2005, flow rate validation flags were redefined to be more objective and quantitative in nature and make more complete use of the 15-minute flow rate data [McDade, 2005]. The new flags were adopted for future use and also applied to the 2000–2004 data so that they were determined quantitatively and consistently across the entire Regional Haze Rule 5-year base period. The data prior to 2000 do not use the new flow validation flags. The version I sampler did not collect 15-minute flow rate data, and thus the necessary data are not available to formulate the new flags for data prior to 2000.

1.3.3.4. Change in Flow Rate Calculations

Flow rates were recalculated for the 2000–2004 period to correct an error in the calculation that existed prior to January 2004 [McDade, 2005]. The flow calibration coefficients were incorrectly referenced to the temperature at the time of annual calibration rather than a standard temperature. The range of bias resulting from the calculation error extended from about a 5% high to about a 4% low, with over 80% of the instances falling within a bias of $\pm 2\%$. A single calibration temperature was applied at each site for the entire period between calibrations, typically about a year. Thus the bias did not appear as random fluctuations but rather as offsets in annual blocks of data. The pre-2000 temperature data are not of sufficient quality to warrant applying the small calibration temperature correction.

1.3.3.5. Spectral Corrections to S and Al Data from the XRF Cu Anode System

In both PIXE and XRF analysis, sulfur is subject to a small positive interference from lead, and aluminum is subject to a small positive interference from bromine. The corrections are

$$S \text{ (corrected)} = S - 0.74 * Pb$$

$$Al \text{ (corrected)} = Al - 0.62 * Br$$

These corrections were initially not applied to the XRF data from December 2001 through 2004; the corrections were applied to the 2000–2004 redelivery and will be applied to future XRF data.

1.3.3.6. Change in the Reporting of Organic Pyrolyzed Carbon (OP) Concentrations

In the TOR carbon analysis, the sample is first heated in a non-oxidizing He atmosphere to volatilize the OC. During this phase of the analysis, some of the OC on the filter pyrolyzes to EC in the absence of O₂. The organic pyrolyzed carbon (OP) fraction corrects the OC and EC fractions for this pyrolyzed carbon. However, oxidizing minerals [Fung, 1990; Sciare et al., 2003], catalysts [Lin and Friedlander, 1988a,b] in the sampled aerosol, or oxygen leaks in the TOR analyzer can oxidize [Chow et al., 2005] EC, which can be released during the non-oxidizing phase of the TOR analysis. In these cases, the OP fraction can be negative. Previously, negative OP values were reported as 0. With the resubmission of the carbon analysis data for 2000–2004, the negative OP values were reported unmodified.

Approximately one in ten OP values was affected by this change [Chow et al., 2005]. This change in reporting procedure will also be applied to future data deliveries. While the OP correction could be applied to data prior to 2000, it was decided that this one small correction did not warrant the effort involved in reprocessing 12 years of data.

Table 1.6. Major network-wide changes in sampling, analysis and data reporting.

Change Date	Change Description
9/15/1990	Ion analysis contractor switched from Research Triangle Institute (RTI) to Global Geochemistry Company (GGC). Ion samples extracted using anion eluent.
6/1/1992	Analysis of elements with atomic weights from Fe to Pb was changed from PIXE to XRF by Mo anode, decreasing their minimum detection limits (mdl). The cyclotron time for the PIXE analysis was reduced increasing the mdl for elements below FE.
3/1/1994	Optical absorption measurement changed from Laser Integrating Plate Method (LIPM) to Hybrid Integrating Plate/Sphere Analysis (HIPS).
6/1994	Changed nylon filter size from 47mm diameter to 25mm.
4/1995-present	Began removing Module A filter masks, effectively changing the sample area from 2.2 sq. cm to 3.5 sq. cm. Transition still in progress as of the date of this writing.
5/23/1995	Ion analysis switched to Research Triangle Institute (RTI). Ion samples extracted using anion eluent.
1996	Added glycerin to Module B denuder. The new model denuders were installed during annual maintenance visits.
10/1996	Changed nylon filter manufactures from Gelman to MSI.
6/1/1997	Ion samples extracted using DI water at GRSM1, SHEN1, DOSO1. All other sites extracted with anion eluent.
1/28/1999	Ion samples extracted using DI water at all sites.
12/1999 - 4/2001	Transitioned the new and existing 110 IMPROVE sites to version II IMPROVE samplers.
4/2000 -1/2001	Changed nylon filter size from 25 mm to 37 mm.

Change Date	Change Description
10/11/2000	Ion samples extracted using anion eluent at all sites except GRSM1, SHEN1, and DOSO1 where extraction is with DI water.
4/5/2001	Ion samples extracted using DI water at all sites.
12/1/2001	Analysis of elements with atomic weights from Na to Mn was changed from PIXE to XRF by Cu anode.
2002	Started reporting below-mdl gravimetric measurements.
6/1/2002	Changed from quarterly to monthly medians to estimate artifact corrections from field blanks & secondary filters.
10/1/2002	Standardized XRF run times at 1000 seconds.
3/2003	Installation of collocated extra modules began.
11/2003	Installation of collocated modules with Speciated Trends Network began.
1/2004	Changed module B filter supplier from Osmonics to Pall-Gelman.
9/2004	Changed from monthly to quarterly medians to estimate artifact corrections from field blanks & secondary filters.
10/2005	Redelivery of 2000–2004 data to back-correct data for several data processing changes including new definitions of the flow rate validation flags, a correction to the flow rate calculation, a correction to the XRF results, and a change in the way negative OP fractions were reported.

1.4 THE COMPARISON OF CONCENTRATIONS FROM COLLOCATED IMPROVE AND STN MONITORING SITES

Chapters 2 and 3 examine the annual spatial patterns and the seasonal patterns of the major fine aerosol constituents from 159 IMPROVE sites from 2000 through 2004. The IMPROVE network collects samples in primarily remote rural areas, thus providing little information on the aerosol composition and concentrations in and near population centers. To fill in these gaps, data from the EPA’s Speciated Trend Network (STN) from 84 sites were incorporated into the spatial and seasonal pattern analyses. The STN collects speciated aerosol data similar to the IMPROVE network, but the sites are located primarily in urban/suburban settings. Incorporation of the STN data extends the spatial aerosol patterns from the surrounding remote areas into urban areas, providing insights into the fraction of the particulate matter (PM) that is contributed by regional and local sources.

IMPROVE and the STN both collect 24-hour PM_{2.5} samples on similar filter media on a 1-in-3-day sampling schedule for quantifying PM_{2.5} mass and its chemical constituents. Both networks use similar gravimetric analysis for quantifying PM_{2.5} mass, ion chromatography for NO₃⁻ and SO₄⁼, and XRF for elements including S, Al, Fe, Ca, Si, and Ti. However, IMPROVE uses thermal optical reflectance (TOR) to measure carbon, and the STN uses thermal optical transmittance (TOT). These two techniques are known to produce similar total carbon concentrations but different splits between OC and light-absorbing carbon (LAC) concentrations. The TOR analysis generally has higher LAC concentrations than the TOT technique. IMPROVE and the STN also use different samplers and different standard operating procedures for sample collection and analysis and maintain independent quality assurance programs.

The two networks have collocated IMPROVE and STN samplers in several urban and rural locations. These collocated data were compared to identify potential biases between the annual IMPROVE and STN concentrations that could impact the interpretation of results from the combined data sets. This was done using data from six collocated sites in 2002 and five collocated sites in 2003. For each site and year, annual averages of the major particulate

composite components were calculated, resulting in 11 pairs of annual average values for each particulate component. The calculation of the composite components and their aggregation are described in Chapter 2. A summary of the results for PM_{2.5} and the major particulate composite components are presented in Table 1.7. Appendix E provides a detailed analysis of the differences between a subset of the IMPROVE and STN measured species. Note that the STN does not generally blank correct the OC concentration to account for positive artifacts, but IMPROVE does. The EPA has developed OC adjustments for the STN concentrations to correct for the positive artifacts. The STN OC concentrations used in this analysis were adjusted for the carbon artifact. Appendix E contains a comparison of the unadjusted STN and IMPROVE carbon concentrations.

As shown in Table 1.7, the errors between the annual average values were 16% or smaller for all parameters except fine soil, which was 35%. The bias was 1.3% or smaller for PM_{2.5}, ammonium sulfate, ammonium nitrate, and organics. However, the bias for LAC was -10.4%, indicating the IMPROVE annual average LAC is 10% greater than for the STN. For the fine soil, the IMPROVE annual average concentrations were 30% greater than for the STN.

The spatial and seasonal analyses in Chapters 2 and 3 used 5-year average values. Therefore, the random error between the IMPROVE and STN data will likely be smaller than reported in Table 1.7. This, combined with the fact that PM_{2.5}, ammonium sulfate, ammonium nitrate, and organics have small biases, indicates that the IMPROVE and STN data are sufficiently similar to combine the data. These results indicate that the STN LAC concentrations will be systematically smaller than IMPROVE's by about 10%. This bias needs to be considered when comparing the IMPROVE and STN concentrations. The 30% bias in the fine soil is sufficiently large that the combined fine soil patterns should be treated as suspect.

Table 1.7. Comparison of annual average concentrations between collocated IMPROVE and STN monitoring sites.

	PM _{2.5}	Ammonium Sulfate	Ammonium Nitrate	Organics	Light Absorbing Carbon	Fine Soil
Average IMPROVE (µg/m ³)	9.1	3.3	1.1	4.0	0.6	1.4
Average STN (µg/m ³)	9.2	3.2	1.0	4.1	0.5	0.9
¹ Error (%)	8.3	5.5	13.2	16.0	15.9	35.5
² Bias (%)	1.3	-0.6	-0.5	-0.8	-10.4	-30.8

¹ Error = median $\left(\left| \frac{\bar{X}_i - \bar{Y}_i}{\bar{Y}_i} \right| \right)$ where \bar{X}_i and \bar{Y}_i are the annual average STN and IMPROVE concentrations, respectively.

² Bias = $\frac{1}{N} \sum_i \frac{\bar{X}_i - \bar{Y}_i}{\bar{Y}_i}$ where N is the number of annual average concentrations.

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