

Article



Compilation and Evaluation of Ambient Respirable Crystalline Silica Air Quality Data near Sand Quarries and Processing Facilities

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Abstract: Ambient respirable crystalline silica air quality is of concern to many communities near mineral processing facilities and to regulatory agencies serving these communities. Accurate air quality data are needed to compare measured respirable crystalline silica concentrations at the fencelines of mineral processing facilities with the published health effect guideline published by the California Office of Health Hazard Assessment (OEHHA). This article is a compilation and evaluation of air quality studies around a diverse set of nineteen sand producing facilities. The respirable crystalline silica air quality data compiled by Air Control Techniques, P.C. and most of the data compiled by other researchers cited in this article have been measured using EPA Reference Method samplers adjusted for respirable crystalline silica sampling and NIOSH Method 7500 X-ray diffraction analyses. The authors conclude that (1) the ambient concentrations in the diverse set of mineral processing facilities were consistently lower than the 3.0 microgram per cubic meter chronic reference exposure level (REL) adopted by OEHHA, (2) upwind-to-downwind fenceline concentration differences were small, and (3) the fenceline t concentrations were often at background concentrations of ambient respirable crystalline silica.

Keywords: ambient respirable crystalline silica; air quality; health effects; fenceline sampling; sand production; ambient air sampling; mineral industries; community air quality

1. Introduction

There have been significant community concerns expressed regarding respirable crystalline silica particulate matter air quality in the vicinity of sand quarries, sand trans-load operations, sand processing facilities, and other mineral processing facilities. Prior to 2005, there were no ambient air quality standards or guidelines to evaluate possible health effects of ambient respirable crystalline silica. In 2005, the California Office of Environmental Health Hazards Assessment (OEHHA) published a chronic reference exposure limit of 3 micrograms per cubic meter for ambient respirable crystalline silica [1]. OEHHA based this guideline on industrial hygiene health effects studies conducted with PM_4 crystalline silica personal occupational exposure samplers. In both the occupational hygiene studies and the air quality studies addressed in this article, PM_4 is defined as particulate matter having aerodynamic sizes equal to or less than 4 micrometers as measured in accordance with NIOSH Method 0600 or equivalent procedures.

OEHHA set their REL at a very low concentration of 3 micrograms per cubic meter $(\mu g/m^3)$ to protect sensitive individuals subject to exposure to respirable crystalline silica in ambient air. The NIOSH 0600 samplers used to assess exposure to the much higher concentrations in occupational work areas are not capable of accurate measurement at this low REL concentration set for ambient air exposure. The NIOSH 0600 samplers have low sample flow rates and are operated only for 8 to 10 h work shifts. To increase measurement



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). sensitivity to the low OEHHA REL level, Richards and Brozell [2] and other researchers have adapted EPA $PM_{2.5}$ Reference Method samplers meeting the requirements of 40 CFR Part 50, Appendix L requirements, which operate at much higher sample flow rates and operate for 24 h periods. The air flow rates in these modified $PM_{2.5}$ samplers were adjusted to (1) provide a 50% cut size of 4 micrometers and (2) maintain a size-efficiency curve closely matching the NIOSH 0600 personal samplers. To provide consistency with the crystalline silica data used by OEHHA, the filter samples from the flow rate adjusted $PM_{2.5}$ samplers were analyzed using X-ray diffraction in accordance with NIOSH Method 7500. With this combined sampling and filter analytical approach, the lower limit of quantification for crystalline silica was $0.31 \ \mu g/m^3$ —a value well below the OEHHA REL of $3.0 \ \mu g/m^3$.

Richards et al. [3] used this newly developed ambient respirable crystalline silica measurement method in 2006 to conduct short-term studies at two facilities in California. The California Air Resources Board and the South Coast Air Quality Management District (Los Angeles area) used a similar sampling method in several short-term studies [4,5]. All of these short-term California studies indicated very low ambient respirable crystalline silica concentrations.

In 2012, Richards and Brozell [6] initiated three-year sampling programs at three frac sand guarries and one sand processing facility located in Wisconsin. Richards and Brozell [7] also conducted tests at six other frac sand facilities in Wisconsin and Minnesota [8]. All of the average ambient respirable crystalline silica concentrations measured in these studies were below the OEHHA REL. The upwind-to-downwind concentrations measured in these sampling programs indicated very little contribution from the sources monitored. The majority of the 24 h concentration measurements summarized in Wisconsin and Minnesota tests [6,7] were below the limit of quantification of 0.31 μ g/m³ and were close to background concentration levels. The Minnesota Pollution Control Agency (MPCA) [8–11] published short-term ambient respirable crystalline silica monitoring programs at several sand quarries and a trans-load facility. Despite using different samplers, the MPCA data were similar to those of Richards and Brozell [6,7] and the California-based studies [3–5] conducted earlier. Peters et al. [12] published ambient monitoring data at seventeen residences located within 800 m of frac sand quarries in western Wisconsin and found ambient respirable crystalline silica concentrations below the OEHHA REL in samples obtained during 48 h sampling periods.

To provide a more comprehensive set of air quality data, this paper summarizes ambient respirable crystalline silica concentration measurements over long time periods at numerous additional mineral facilities having diverse process equipment, production capacities, and surrounding terrain features that could potentially affect dispersion of fugitive dust emissions. This much larger and diverse data set created by the addition of recently completed ambient monitoring programs [13] provides an improved basis for evaluating respirable crystalline silica air quality in communities near mineral processing facilities. Acronyms and definitions of terms used are listed at the end of this article.

2. Ambient PM₄ Crystalline Silica Measurement

2.1. Facilities

The facilities sampled had a wide variety of production rates, mineral characteristics, plant process equipment, and surrounding terrain features. A summary of their diverse characteristics is provided in Table 1. Details concerning the characteristics of the facilities, process types, terrain characteristics, sampling location arrangements, and other factors potentially affecting the measured ambient PM_4 respirable crystalline silica are provided in references [3–13].

Location	Туре	Production Rate	Product	Topography and Community Characteristics	Ref.
Wedron, IL	Quarry and Processing	High	Frac Sand	Small hills and a river valley, located close to a small community	[13]
Menomonie, WI	Quarry and Processing	Moderate	Frac Sand	Very small hills, rural area	[13]
Kasota. MN	Quarry and Processing	High	Frac Sand	Flat plain, lightly vegetated, close to town of Kasota, Minnesota	[13]
Sparta, WI	Quarry and Processing	High	Frac Sand	Small hills, lightly vegetated. adjacent to a residential community	[13]
Berkeley Springs, WV	Quarry and Processing	Moderate	Milled Sand	Mountain valley	[13]
Chippewa Falls, WI	Processing	High	Frac Sand	Flat terrain, near residential area	[6]
DS Mine, WI	Quarry	Moderate	Frac Sand	Rolling hills, rural agricultural area	[6]
S&S Mine, WI	Quarry	Moderate	Frac Sand	Rolling hills, rural agricultural area	[6]
DD Mine, WI	Quarry	Moderate	Frac Sand	Rolling hills, rural agricultural area	[6]
Maiden Rock, WI	Quarry and Processing	Large	Frac Sand	Steep river valley	[7]
Cataract Green, WI	Greenfield	None	N/A	Rolling hills, rural agricultural area	[7]
Cataract Green, WI	Quarry and Processing	Moderate	Frac Sand	Rolling hills, rural agricultural area	[14]
Downing, WI	Quarry	Moderate	Frac Sand	Rolling hills, rural agricultural area	[7]
Jordan Sands, MN	Quarry and Processing	Large	Frac Sand	Rolling hills, rural agricultural area	[8]
Winona, MN Stanton, MN	Trans-Load Greenfield	N/A None	Frac Sand N/A	Community Rural	[10] [10]
Shakopee, MN	Quarry and Processing	Moderate	Frac Sand	Rolling hills, rural agricultural area	[9]
Titan, MN	Trans-Load	Moderate	Frac Sand	Rolling hills, rural agricultural area	[10]
Duarte, CA	Quarry and Processing	Large	Frac Sand	Flat terrain at base of mountains	[4,5]
San Diego, CA	Quarry and Processing	Large	Construction Sand	Flat terrain	[3]
Vernalis, CA	Processing	Large	Construction Sand	Flat terrain	[3]

Table 1. Characteristics of the facilities.

Most of the studies had sets of samplers arranged in an upwind-downwind configuration at the fencelines of the facilities. Some of the sampling programs used collocated samplers at the downwind sites to evaluate the precision of the ambient PM_4 crystalline silica 24 h average concentration measurements.

Many of the sampling programs operated samplers on either a once-every-three day or once-every six-day schedule. The sampling programs that included collocated samplers operated those collocated samplers on a once-every twelfth -day schedule. Essentially all the sampling programs operated on the calendar day-specific sampling schedule specified by EPA for each calendar year. Accordingly, the day-by-day respirable crystalline silica concentration variations could be compared with air quality variations measured on the same days by the state agency and EPA PM_{2.5} and PM₁₀ samplers located near the sampling locations.

2.2. Sampling and Analytical Procedures

The characteristics and study periods of the various sampling programs evaluated in this paper are summarized in Table 2.

Figure	Type and Number of PM Samplers	Sampling Frequency	Sampling Period (Month/Year)	Sampler Operator	Ref.
Wedron	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 3	2/15 to 3/16	Contractor	[13]
Menomonie	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 6	7/14 to 11/15	Employee	[13]
Kasota	1 PM ₄ CS Partisol 2000 <i>i,</i> Locations 1, 2, and 3	1 day in 6	3/14 to 4/19	Employees	[13]
Sparta	1 PM ₄ CS, Partisol 2000 <i>i</i> downwind	1 day in 6	9/12 to 2/20	Employees	[13]
Berkeley Springs	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM4 CS, Partisol 2000 <i>i</i> upwind	1 day in 6	7/12 to 7/13	Employee	[13]
Chippewa Falls	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 3	8/12 to 12/14	Contractor	[6]
DS Mine	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 3	8/12 to 12/14	Contractor	[6]
S&S Mine	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 3	8/12 to 12/14	Contractor	[6]
DD Mine	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 3	11/12 to 12/14	Contractor	[6]
Maiden Rock	1 PM ₄ CS, Partisol 2000 <i>i</i> , Loc. 1 1 PM ₄ CS, Partisol 2000 <i>i</i> , Loc. 2 1 PM ₄ CS Partisol 2000 <i>i</i> , Loc. 3	1 day in 3	3/13 to 3/14	Contractor, resident	[7]
Cataract Green	None	N/A	9/12 to 12/13	Contractor	[7]
Cataract Green	1 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 6	12/13 to	Contractor	[7]
Downing	2 PM ₄ CS, Partisol 2000 <i>i</i> downwind 1 PM ₄ CS, Partisol 2000 <i>i</i> upwind	1 day in 6	8/12 to 9/13	Contractor	[7]
Jordan Sands	1 PM ₄ CS, Partisol 2000 <i>i,</i> downwind 1 PM ₄ CS Partisol 2000 <i>i</i> , upwind	1 day in 6	9/14 to 12/16	Employee contractor	[8]
Winona	1 PQ200 CS downwind	1 day in 6	2/14 to 12/14	MPCA	[9]
Stanton	None	1 day in 6	1/14 to 12/14	MPCA	[9]
Shakopee	Unknown	1 day in 12	8/12-12/13	Employee	[11]
Titan	Unknown	1 day in 6	9/13 to 9/15	Employee	[12]
Duarte	1 PM ₄ CS, Unknown downwind	1 day in 6	5/6 to 9/6	SCAQMD	[4,5]
San Diego	$2 PM_4 CS$, Partisol 2000 <i>i</i>	N/A	9/6	ACTPC	[3]
Vernalis	$2 \text{ PM}_4 \text{ CS}$, Partisol $2000i$	N/A	9/6	ACTPC	[3]

Table 2. Sampling program characteristics.

Most of the sampling programs measured PM_4 respirable crystalline silica data using Partisol 2000*i* samplers meeting the performance requirements of 40 CFR Part 50, Appendix L and adjusted to provide a cut size of 4 micrometers rather than 2.5 micrometers (All particulate matter size data are expressed as aerodynamic diameters). These PM_4 sampling procedures were developed by Richards and Brozell in 2005 [2] in response to the publication of the OEHHA REL [1]. The South Coast Air Quality Management District developed and used a similar approach [4,5]. The crystalline silica samples were analyzed using NIOSH reference method 7500 [14].

The quality assurance procedures used for sampling PM₄ respirable crystalline silica were based on EPA specified quality assurance procedures for PM_{2.5} sampling [15]. These quality assurance procedures included routine sampler audits, independent audits of the samplers, blank filter analyses, collocated sampler-primary sampler precision analyses, and detailed laboratory procedures. The Wisconsin Department of Natural Resources (WDNR) and the Minnesota Pollution Control Agency (MPCA) reviewed the sampling protocols for several of the studies and audited the samplers in several of the studies. EPA reviewed the sampling procedures and data for a sampling program at a facility in Illinois.

2.3. Crystalline Silica Characteristics

Quartz, cristobalite, and tridymite forms of crystalline silica were included in the scope of the sampling programs. Of these three forms—quartz is by far the most common. Quartz is the second most common mineral in the Earth's crust and is present in most rocks and soils in most geographical locations.

All three forms of crystalline silica are especially hard and resist size reduction down to particles with an aerodynamic particle size of 4 micrometers. Ambient air concentrations are low due to the low formation rate of particles that can be entrained in the wind. Accordingly, the sampling and analytical procedures used in these air quality sampling studies had to have the capabilities of measuring low ambient mass concentrations and accurately quantifying small amounts on the sampled filters.

The PM₄ particulate matter samples were collected on PVC filters rather than the Telfon®filters used for PM_{2.5} and PM₁₀ sampling. These filter samples were analyzed at qualified laboratories using NIOSH Method 7500 X-ray diffraction. This is the method most often used for industrial hygiene PM₄ crystalline silica sampling. The limit of quantification using the sample flowrate modified Partisol 2000*i* samplers with NIOSH Method 7500 was 0.31 micrograms per cubic meter of crystalline silica. All three common forms of crystalline silica were detectable using this sampling and analytical approach.

3. Ambient Air Concentrations Data, Recently Tested Facilities

The PM₄ respirable crystalline silica data compiled in the various sampling studies described above are summarized in Table 3.

Facility	Sampling Location	Sampling Period (Month/Year)	Number of 24 h Samples	Avg. with ND = LOQ/ $\sqrt{2} \mu g/m^3$	Max., μg/m ³	UCL95%, Average with ND = $LOQ/\sqrt{2}$
	470th St.	3/14 to 4/16	135	0.46	4.89	0.54
77 .	480th St.	4/16 to 4/19	182	0.47	11.58	0.58
Kasota	Town	3/14 to 4/19	307	0.33	2.24	0.35
	Prairie	3/14-4/19	297	0.40	5.05	0.45
Menomonie	North, downwind	7/14 to 7/15	62	0.28	0.81	0.31
	South, upwind	7/14 to 7/15	60	0.24	0.50	0.31
Wedron	North, downwind	2/15 to 3/15	130	1.56	10.1	1.85
	South, upwind	2/15 to 3/15	127	0.25	0.69	0.27
Sparta	One, downwind	1/15 to 2/20	344	0.22	1.81	0.32
Berkeley Springs	One, upwind	7/12 to 7/13	61	0.38	1.91	0.40
	Two, downwind	7/12 to 7/13	60	1.73	5.80	2.05
Cataract Green	One, upwind	12/13 to 10/15	102	0.23	0.75	0.24
(background site)	Two, downwind	12/13 to 10/15	108	0.23	0.56	0.24
Chippewa Falls	North, downwind	8/12 to 12/13	155	0.33 ¹	1.44	0.36
	Southwest, upwind	8/12 to 12/13	153	0.26 ¹	1.44	0.27
	North, downwind	1/14 to 12/14	118	0.31 ¹	1.13	0.34
	Southwest, upwind	1/14 to 12/14	116	0.22 ¹	0.44	0.23

Table 3. Ambient PM₄ respirable crystalline silica concentration data.

Facility	Sampling Location	Sampling Period (Month/Year)	Number of 24 h Samples	Avg. with ND = LOQ/ $\sqrt{2} \mu g/m^3$	Max., µg/m ³	UCL95%, Average with ND = $LOQ/\sqrt{2}$
	Upwind	8/12 to 12/13	151	0.24^{1}	0.63	0.24
DOM	Downwind	8/12 to 12/13	150	0.26 1	1.10	0.27
D5 Mille	Upwind	1/14 to 12/14	121	0.24e ¹	0.88	0.24
	Downwind	1/14 to 12/14	121	0.23^{1}	1.38	0.23
	Upwind	8/12 to 12/13	149	0.30 1	2.13	0.33
S&S Mine	Downwind	8/12 to 12/13	149	0.24^{1}	0.60	0.25
Jas Mille	Upwind	1/14 to 12/14	118	0.27^{1}	0.88	0.29
	Downwind	1/14 to 12/14	117	0.24^{1}	0.75	0.26
	Upwind	11/12 to12/13	139	0.25 1	1.31	0.27
DD Mina	Downwind	11/12 to 12/13	136	0.25 ¹	0.69	0.26
DD Wille	Upwind	1/14 to 12/14	118	$0.22^{\ 1}$	0.50	0.23
	Downwind	1/14 to 12/14	117	0.23 ¹	0.56	0.23
	Town	3/13 to3/14	124	0.25	0.7	0.27
Maiden Rock	Southwest	3/13 to 3/14	125	0.55	2.2	0.61
	Northeast	3/13 to 3/14	124	0.28	2.4	0.34
Cataract Green	Background	9/12 to 12/13	60	0.26	0.81	0.27
Downing -	Southwest	8/12 to 9/13	62	0.29	1.3	0.33
Dowining	Southeast	8/12 to 9/13	63	0.27	0.88	0.30
Jordan Cando	Upwind	8/14 to 8/17	141	0.20 6	1.0	0.25
Jordan Sands	Downwind	8/14 to 8/17	165	0.245 6	0.90	0.30
Winona	Single ^{2,3}	2/4 to 12/14	48	0.23 ⁷	0.40	0.24
Stanton	Single ^{2,3}	2/14 to 12/14	55	0.27 7	0.80	0.30
Titan	Single ^{3,4}	9/13 to 9/15	81	1.28^{4}	6.0	ND ⁸
Shakopee	Single ^{2,3}	8/2 to 12/13	44	0.75 ⁵	1.80	ND ⁸
Duarte	Single	5/6 to 9/7	19	0.60	1.10	0.63
Vernalis —	Upwind	9/6	3	1.10	1.30	N/A
	Downwind	9/6	3	0.77	1.10	N/A
San Diego	Upwind	9/6	3	2.0	2.80	N/A
San Diego —	Downwind	9/6	3	0.57	0.90	N/A
Total Number of 24 h Samples		5226				

Table 3. Cont.

¹ Values below the LOQ in the studies conducted at these plants were calculated as the LOQ/2 rather than the LOQ/ $\sqrt{2}$ in the other studies included in Table 3. The data shown in Table 3 have been adjusted to values calculated as LOQ/ $\sqrt{2}$. ² All data for these sources have been estimated from bar charts published on the MPCA website; ³ There were long interruptions in this sampling program; ⁴ The LOQ for the Titan data have been assumed to be 1.3 µg/m³; ⁵ The LOQ for the Shakopee data have been assumed to be 1.0 µg/m³; ⁶ The LOQ for the Jordan Sands is 0.31 µg/m³; ⁷ The LOQ for the Winona and Stanton data have been assumed to be 0.31 µg/m³; ⁸ UCL95% confidence values of the arithmetic mean were not calculated due to difficulty in interpreting available data.

The ambient PM4 respirable crystalline silica datasets had numerous values below the 0.31 ug/m³ limit of quantification of the sampling/analytical method in most of the sampling programs summarized in Table 3. The averages have been tabulated using nondetect values expressed as the limit of quantification (LOQ) divided by the square root of 2. The latter approach is based on the method recommended by Hornung [16]. The use of the LOQ/ $\sqrt{2}$ to express the non-detect values is more reasonable than assigning zero values to the non-detects considering that crystalline silica is a ubiquitous material in most rocks and soils in most locations. Some very small amount of crystalline silica is almost certainly present in most ambient air samples. However, with this procedure for expressing non-detect values, the minimum respirable crystalline silica concentration that can be reported is $0.22 \ \mu g/m^3$.

The Titan [10] and Shakopee Sands [11] sampling programs conducted in Minnesota had high quantification limits of 1.0 to $1.3 \ \mu g/m^3$. These LOQ values are three to four times higher than the LOQ values in most of the sampling programs such as [3–7,14]. Expressing the numerous non-detect values in these datasets as the LOQ/ $\sqrt{2}$ increased the UCL95% average values in these sampling programs.

4. Discussion

All of the facilities had UCL95% arithmetic average concentrations ranging from 0.22 to 1.73 μ g/m³ (ND = LOQ $\sqrt{2}$) regardless of facility's production rates, topography, and/or climate conditions. The highest PM4 respirable crystalline silica levels were found at Wedron and Berkeley Springs, two of the largest facilities addressed in this article. However, facility size may not have been the dominant factor influencing the observed concentrations. The Wedron downwind sampling location was especially close to the plant processing equipment and plant buildings. Accordingly, the Wedron data may not be representative of fenceline concentrations in most mineral processing facilities. The Berkeley Springs plant produced a finely milled silica product using a large number of grinding circuits in series. The product mass median size distribution was very small. Furthermore, the Berkeley Springs processing plant and quarry are located in a narrow mountain valley, and the only location with available electrical power to operate the samplers was near the processing area and far from the downwind fencelines. Even considering these atypical conditions favoring higher reported ambient concentrations, these two facilities had average concentrations well below the OEHHA REL of 3.0 μ g/m³.

Conversely, the very low ambient respirable crystalline silica concentrations observed at the Cataract Green facility may not be representative of most quarries or processing plants. This plant is located in a rural area without nearby agricultural operations. It was initially chosen as a background, greenfield site to provide information concerning regional background levels. The measured concentrations did not significantly increase after a moderately sized quarry was installed.

The upwind–downwind concentration differences averaged over the study periods were small except at the Wedron and Berkeley Springs facilities. The upwind–downwind differences are summarized in Table 4.

Facility	Upwind PM ₄ Crystalline Silica Concentration, μg/m ³	Downwind PM ₄ Crystalline Silica Concentration, µg/m ³	Difference, µg/m ³
Menomonie	0.31	0.31	0.00
Wedron	0.27	1.85	1.58
Berkeley Springs	0.40	2.05	1.65
Cataract Green	0.24	0.30	0.06
Chippewa Falls 2012–2013	0.26	0.33	0.07
Chippewa Falls 2014	0.22	0.31	0.09
DS mine 2012–2013	0.24	0.26	0.02
DS Mine 2012–2013	0.24	0.23	-0.01
S&S mine 2012–2013	0.30	0.24	-0.06
S&S mine 2012–2013	0.27	0.24	-0.03
DD Mine-2012-2013	0.25	0.25	0.00
DD Mine 2014	0.22	0.23	0.01
Downing	0.30	0.33	0.03
Jordan Sands	0.25	0.30	0.05

Table 4. Upwind–downwind concentration differences.

The highest maximum single day 24 h average concentration of 11.58 μ g/m³ was observed at Wedron. Maximum concentrations at the upwind sampling location in some of

the studies were greater than 2 μ g/m³—probably due to the fugitive dust emissions from nearby agricultural operations.

The ambient PM_4 respirable crystalline silica data measured in all the sampling programs were quite similar. All of the UCL95% confidence levels for the arithmetic means were well below the healthbased OEHHA chronic exposure REL.

Climate conditions do not appear to a major factor in the differences in the UCL95% levels measured. For example, the PM₄ respirable crystalline silica UCL95% levels measured in essentially all the sampling programs in the midwestern U.S. with moderate rainfall levels were similar to the 0.4 to $1.1 \,\mu\text{g/m}^3$ concentrations measured over a summertime four-month period by the South Coast Air Management District in the semi-arid Duarte, California [4,5] area.

The Duarte study conducted by the South Coast Air Quality Management District (Los Angeles area) is also of interest because the sampling location was on the grounds of a school surrounded on three sides by nearby large quarries and processing plants. The sampling location was also close to two major interstate highways with near-constant heavy traffic. A usually dry creek bed was on one side of the school. The daily off-shore and down-valley winds created by the nearby Pacific Ocean and the San Gabriel mountains usually generated air flow passing over at least one of the mineral industry sites, the interstate highways, and/or the dry creek bed toward the school sampling location. Despite these conditions, the ambient respirable crystalline silica conditions were only slightly higher than those measured in most of the midwestern U.S. sampling sites and were well below the California OEHHA chronic exposure REL.

The probable influence of agricultural operations on ambient concentrations is suggested by the differences in the Winona study [9] results and those from Stanton [10]. The Winona study consisted of a single PQA-200 instrument located on the roof of a YMCA building near a trans loading operation. During the 10-month study, two of the 48 samples obtained at Winona had greater than the detectable concentration limit of $0.31 \,\mu\text{g/m}^3$. The concentrations at Winona were similar to other facilities discussed earlier in this article. The PM₄ respirable crystalline silica concentrations at Stanton [9] were slightly higher than at Winona despite the lack of a nearby mineral processing facility. The slightly elevated concentrations observed in this 10-month study at Stanton were probably due to fugitive dust emissions from agricultural operations. Similar agricultural operation impacts were observed at the DS mine study in Wisconsin.

The influence of the crystalline silica content of the material being handled does not appear to be large in this dataset. The crystalline silica levels of the construction sand handled in the San Diego and Vernalis [3] facilities sampled in California were lower than the >95% levels of crystalline silica in the frac sand-oriented sampling programs in the numerous studies in the upper midwestern U.S. However, the ambient PM₄ respirable crystalline silica levels were higher than in the frac sand related studies. The probable impact of nearby unpaved roads and agricultural operations in the California studies appears to overcome any differences due to mineral material crystalline silica content.

The sampling programs at the Titan trans-load facility [10] and at Shakopee Sands [11] used a sampler different from the Partisol 2000*i* samplers used in most of the studies discussed earlier. The estimated limit of quantification values ranged from $1.0 \,\mu\text{g/m}^3$ at Shakopee to $1.3 \,\mu\text{g/m}^3$ at Titan. These LOQ values are three to four times higher than those used in the studies conducted by Richards and Brozell [3,6,7,13] and by Jordan Sands [8]. Due to the high LOQ values at Titan and Shakopee, the average values and the UCL95% values are artificially inflated. Furthermore, the MPCA report concerning Titan indicated frequent quality assurance issues with the sampler flow rates.

Shiraki and Holmen [17] measured PM_{10} crystalline silica at a sand and gravel facility located near the Tracy, California airport. The authors did not detect respirable crystalline silica in the $PM_{2.5}$ particulate matter fractions—a conclusion that suggests low concentrations in the PM_4 respirable fraction. The PM_{10} crystalline silica data reported in this study cannot be accurately equated to the PM_4 respirable size fraction. Furthermore, the sand and gravel facility sampled was almost completely surrounded by other sand and gravel producing facilities, the immediately adjacent Tracy airport, and nearby active agricultural operations. These adjacent sources may have influenced the measured PM_{10} crystalline silica levels.

Peters et al. [12] measured ambient PM_4 respirable crystalline silica at seventeen residences within 800 m of the fencelines of quarries and other mineral processing facilities in Western Wisconsin. The measured PM_4 crystalline silica levels were above the detection limit of 0.4 µg/m³ in seven of the seventeen 48 h average samples. The authors concluded that the measured PM_4 respirable crystalline silica concentrations were below the OEHHA REL of 3.0 µg/m³ adopted in California and Minnesota.

5. Summary

This paper summarizes PM_4 respirable crystalline silica concentrations at a wide variety of mineral producing facilities. The data were compiled during periods of one to three years at facilities of differing production rates, product characteristics, crystalline silica content of the minerals, climate conditions, and terrains. Most of the data were obtained during the 2012 to 2018 period when there was high demand for mineral products. The large majority of the data were obtained in strict accordance with EPA reference method procedures and quality assurance procedures. Accordingly, these studies help to characterize the range of ambient concentrations or PM_4 crystalline silica that exists in a broad sector of mineral industry sources.

The PM₄ respirable crystalline silica concentrations measured at the fifteen mineral producing facilities for which UCL95% values could be calculated had upper mean 95% average values from a low of 0.23 μ g/m³ to a high of 2.05 μ g/m³. None of the UCL95% values of the facility-specific data sets approached or exceeded the OEHHA lifetime REL of 3.0 ug/m³.

Only two of the fifteen facility datasets had UCL95% values above $1.00 \ \mu g/m^3$. Both of these sources were large facilities with downwind sampling locations very close to processing equipment and/or limited dispersion due to unfavorable terrain.

The upwind–downwind concentration differences confirm that the contributions of mineral processing facilities to the ambient air at the downwind fencelines are very small and are often near to the lower limit of quantification.

The similarities of the upwind-downwind concentrations data and the similarities of the maximum observed concentrations indicate that the PM_4 respirable crystalline silica concentrations at the downwind fencelines of mineral industry sources are at or near the background concentrations. Additional studies are needed to more fully characterize background respirable crystalline silica concentrations near mineral facilities, agricultural operations, unpaved roads, construction sites, and arid, unvegetated soil. These future studies may need sampling times higher than 24 h to reduce the limit of quantification below 0.31 μ g/m³.

The extensive PM_4 respirable crystalline silica data consisting of more than 5000 24 h average concentration values at 19 separate facilities compiled in this paper indicate that mineral processing facilities have a minimal effect on downwind ambient concentrations and do not cause exceedances of the OEHHA health-based REL. This is not a surprising conclusion considering that mineral processing facilities do not use process equipment or procedures that are sufficient to break down much of the very hard crystalline silica into the very small PM_4 size range.

The air quality conclusions based on the data evaluated are generally consistent with the conclusions of other researchers—including the Institute for Wisconsin's Health [18] and the Texas Commission on Environmental Quality [19].

6. Recommended Further Study

Additional study is needed to evaluate the seasonal variability of the background levels of PM₄ respirable crystalline silica concentrations in arid and semi-arid areas, near

unpaved roads, in urban areas with active building construction, in areas downwind of controlled burning and wildfires, and in agricultural areas.

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Conflicts of Interest: Air Control Techniques, P.C. is an independent, third-party engineering and testing consulting firm. We have designed and supervised ambient respirable crystalline silica sampling programs for industrial firms needing to respond to community groups and regulatory agencies requesting ambient air quality data. Air Control Technique, P.C. is a member company of the National Stone Sand and Gravel Association (NSSGA), which had no involvement in any of these ambient air sampling programs. No financial support was solicited or provided for the preparation of this article.

Abbreviations

Acronyms and Definitions

ACTPC	Air Control Techniques, P.C.
Crystalline Silica	Quartz, Cristobalite, and Tridymite
EPA	U.S. Environmental Protection Agency
LOQ	Limit of quantification
MPCA	Minnesota Pollution Control Agency
ND	Non-detectable concentration
NAAQS	EPA National Ambient Air Quality Standards
NIOSH	National Institute of Occupational Safety and Health
OEHHA	California Environmental Health Hazard Assessment
DM	Particulate matter equal to and smaller than an aerodynamic diameter
1 1/14	of 4.0 micrometers as measured in accordance with NIOSH Method 0600
PVC	Polyvinyl chloride
Respirable Crystalline	Crystalline silica in the PM, size range
Silica	Crystalline shied in the 1 Wi4 size funge
SCAQMD	South Coast Air Quality Management District
TCEQ	Texas Commission on Environmental Quality
WDNR	Wisconsin Department of Natural Resources
UCL95%	95% Upper confidence limit of the arithmetic mean
XRD	X-ray diffraction
Units of Measure	
$\mu g/m^3$	Micrograms per cubic meter at actual conditions when referring to PM_4 particulate matter.

References

- 1. California Office of Environmental Health Hazards. Crystalline Silica. 2005. Available online: https://oehha.ca.gov/airs/ document/adoption-chronic-reference-exposure-level=silica-crystalline-respirable (accessed on 3 July 2021).
- Richards, J.; Brozell, T. Ambient PM₄ Crystalline Silica Monitoring Method Development. Report to the Coalition for the Responsible Regulation of Naturally Occurring Minerals (Now Part of CalCIMA); California Construction & Industrial Materials Association: Sacramento, CA, USA, 2006.
- 3. Richards, J.; Brozell, T.; Rea, C.; Boraston, G.; Hayden, J. PM₄ crystalline silica emission factors and ambient concentrations at aggregate-producing sources in California. *Air Waste Manag. J.* **2009**, *59*, 1287–1296. [CrossRef] [PubMed]
- 4. South Coast Air Quality Management District. *Sampling and Analysis of Samples Collect in the Cities of Duarte and Azusa, Report MA* 2006-01; South Coast Air Management District: Diamond Bar, CA, USA, 2006.
- South Coast Air Quality Management District. Sampling and Analysis of Samples Collect in the Cities of Duarte and Azusa, Follow-Up #4 Report MA-2008-03. Report MA 2006-01; South Coast Air Management District: Diamond Bar, CA, USA, 2008.
- Richards, J.; Brozell, T. Community exposure to ambient respirable crystalline silica near frac-sand producing facilities. *Atmosphere* 2015, 6, 960–982. [CrossRef]
- Richards, J.; Brozell, T. Fenceline PM₄ crystalline silica concentrations near sand mining and processing facilities in Wisconsin. *Min. Eng.* 2015, 67, 53–59.
- Minnesota Pollution Control Agency. Jordan Sands-Mankato, MN. 2018. Available online: www.pca.state.mn.us/sites/default/ files/g-7-01.pdf (accessed on 3 July 2021).
- 9. Minnesota Pollution Control Agency. Winona Community Ambient Air Monitoring. 2014. Available online: www.pca.state.mn. us/sites/default/files/g-85-03.pdf (accessed on 3 July 2021).
- 10. Minnesota Pollution Control Agency. Titan Lansing Transload Ambient Air Monitoring. 2015. Available online: www.pca.state. mn.us/sites/default/files/9-13-03.pdf (accessed on 3 July 2021).
- 11. Minnesota Pollution Control Agency. Shakopee Sand Ambient Air Monitoring. 2015. Available online: www.pca.state.mn.us/ sites/default/files/g-70-03.pdf (accessed on 3 July 2021).
- Peters, T.M.; O'Shaughnessy, P.T.; Grant, R.; Altmaier, R.; Swanton, E.; Falk, J.; Osterberg, D.; Parker, E.; Wyland, N.G.; Sousan, S.; et al. Community airborne particulate matter from mining for sand used as hydraulic fracturing proppant. *Sci. Total Environ.* 2017, 609, 1475–1482. [CrossRef] [PubMed]
- 13. Brozell, T.; Richards, J. Ambient PM₄ Respirable Crystalline Silica Concentrations at the Fencelines of Six Facilities in Minnesota, Wisconsin, and Illinois; Air Control Techniques, P.C.: Cary, NC, USA, 2021.
- 14. National Institute of Occupational Health and Safety. Method 7500, XRD Analyses of Crystalline Silica. 2003. Available online: https://www.cdc.gov/niosh/docs/2003-154/pdfs/7500.pdf (accessed on 3 July 2021).
- 15. U.S. EPA. *Quality Assurance Guideline Document;* EPA Publication 454/B-13-03; U.S. Environmental Protection Agency: Washington, DC, USA, 2013; Volume II.
- 16. Hornung, R.W.; Reed, L.D. Estimation of average concentration in the presence of nondetectable values. *Appl. Occup. Environ. Hyg.* **1990**, *5*, 46–51. [CrossRef]
- 17. Shiraki, R.; Holmen, B.A. Airborne respirable silica near a sand and gravel facility in central California: XRD and elemental analyses to distinguish source and background quartz. *Environ. Sci. Technol.* **2002**, *36*, 4956–4961. [CrossRef] [PubMed]
- 18. Institute for Wisconsin's Health. *Health Impact Assessment of Industrial Sand Mining in Western Wisconsin*; Institute for Wisconsin's Health Inc.: Madison, WI, USA, 2016.
- 19. Texas Commission of Environmental Quality. Crystalline Silica, Ambient Air Monitoring and Evaluation of Community Health Impacts near Aggregate Production Operations. December 2020. Available online: https://www.tceq.texas.gov/assets/public/ comm_exec/pubs/as/202.pdf (accessed on 3 July 2021).