Atmos. Chem. Phys. Discuss. https://doi.org/10.5194/acp-2017-231-RC3, 2017 © Author(s) 2017. This work is distributed under the Creative Commons Attribution 3.0 License

## Interactive comment on "Physical and chemical properties of deposited airborne particulates over the Arabian Red Sea coastal plain" by Johann Engelbrecht et al.

#### **Anonymous Referee #1**

Received and published: 19 June 2017

The present manuscript describes and analyzes the measurement of dust deposition at 6 sites at the King Abdullah University of Science and Technology (KAUST) campus along the Red Sea. The description includes local meteorology and instruments used. The analysis includes size distribution, chemical and mineralogical composition of dust. They compare their results with a previous work performed on soil sample of the same area. It is interesting to see their similarity. They also compare with measurements at other locations in the Arabian Peninsula, Middle East and United States.

1. These results could quite useful to better characterize dust in the atmosphere. Unfortunately, their use by the modeling community necessitates assumptions, which have not been discussed. The only thing they provide is a figure showing the number size distribution at one collection site, and they suggest to derive from this figure the mass of particles. This method is inadequate. First, they should provide the values in a Table. Second, this implies assumption concerning shape and density, which varies with soil texture. Third, they should provide variability between sites.

Authors Response: We have made adjustments as suggested below.

2. In addition, some work will be necessary to better structure the text, and to clarify some sentences throughout the manuscript. There are also grammatical errors, and typos to correct. Overall, some efforts have to be done to improve the manuscript and make it more appropriate for publication in Atmospheric Chemistry and Physics, but otherwise it would be a good paper.

Authors' Response: We have made adjustments as suggested below.

3. Detailed comments: Abstract: Page 2, Line 21-22: "These data will also support dust modeling.. mass balance and optical properties". I wish this would be true. But there is no possibility to derive mass balance from one figure of number size distribution. Concerning optical properties, they are strong function of size distribution.

Authors Response: We replaced the single size distribution plot (Fig. 9a) with comparable 12 monthly size distribution plots (Fig. 8a), expressed both as number (Supplement C) and volume percentages (Figure 8a, Appendix A). A table (Table 4) with distribution statistics, and assessments of the < 10µm and

- < 2.5  $\mu m$  mass fractions are given. The table also provides information on particle deposition rates by month.
  - 4. Providing mineralogical data as a function of size will make this paper really useful.

Authors Response: Mineralogical data as measured by XRD per size fraction is not available, only for the total deposition samples (Fig. 7)

5. Introduction: The Introduction should be reworked. Some paragraphs in subsequent sections could be moved in the Introduction to improve the reading of the manuscript. I would suggest the following structure, which hopefully help in my following comments. 1. Introduction 1.1 Importance of dust 1.2 Importance of mineralogy 1.3 Previous work on mineralogy 1.4 Gaps 1.5 How is your work filling the gaps 2. Description of the area 2.1. Meteorology 2.2 Dust sources and deposition.

Authors Response: The chapters 1 and 2 were reworked as suggested to improve the reading of the manuscript

6. Page 3, Line 4. I would rather use Schulz et al. (2012) instead of Bergametti and Foret (2014). It is a more appropriate citation for uncertainties associated with model dust deposition.

Authors Response: We added Schultz et al (2012) to the Bergametti and Foret (2014) reference

7. Page 3. Line 9: "important dust source regions". You may want to cite the comprehensive work on the subject by Prospero et al. (2002) and Ginoux et al. (2012)

Authors Response: Added references Prospero et al. (2002) and Ginoux et al. (2012)

8. Page 3 Line17-21: Limit the number of citations to key papers. Page 4, Line 19 –Page 5, Line 4: this paragraph does not fit in the flow of thinking. I suggest to move it in the proposed Section 2.1 providing description of the general area.

Authors Response: The references will be sorted under each category but retained as such. Moved to new Section 2.1

9. Page 4, Line 5: You never say why mineralogy is important, although this should be the key motivation of this work. You should develop this into a full paragraph (proposed Section 1.2).

Authors Response: Expanded in new Section 1.2. Mineralogy is important in the dust forming process in soils, and the dust transport mechanism. Optical properties such as refractive indices differ amongst minerals.

10. Page 5, Line 9: "However" remove

Authors Response: "However" removed

11. Page 5, Line 12. Break the sentence after the citations, and replace "varying with" by "Its adverse effects will depend on"

Authors Response: Sentence restructured as suggested

12. Page 5 Objectives: This should be articulate within the Introduction. Start by saying why mineralogy is important, then what has been done, then what is the originality of the work, and then finish by providing a succinct outline of the manuscript.

Authors Response: Objectives included in Introduction

13. Page 5, Line 25: "plain to be an" => "plain is an"

Authors Response: Corrected as suggested

14. Page 5, Line 25: remove "province"

Authors Response: "province" removed

15. Page 5, Line 25-26: sentence unclear, and provide a reference.

Authors Response: Rephrased and references, Prakash et al, Anatolii et al provided

16. Page 5, Line 27: remove "inevitably"

Authors Response: "inevitably" removed

17. Page 6, Line 8-18: you repeat yourself. Restructure as suggested.

Authors Response: Restructured?

18. Page 6: Line 20-25: Move to suggested Section 2.2 where you describe the general area.

Authors Response: Moved to suggested Section 2.2

19. Page 8, Line 15 "soils and dusts" replace (as well as all other occurrences) by "soil and dust".

Authors Response: Replace with "soil and dust" although the plural "soils and dusts" is not incorrect

20. Page 8, Line 16-18. This is an argument showing the importance of mineralogy and should be moved in suggested Section 1.2

Authors Response: Lines 16-18 moved to the new Section 1.2

21. Page 8, Line 19-23: It is unclear what are these 3 methods for. Are they all used for mineralogical analysis? What are the benefits of using 3 methods?

Authors Response: This comment is unclear. The three methods measure different components of the dust, XRF for chemical elements, IC for the ions, and XRD for the mineral phases.

22. Page 9, Line 1 to 14. I don't see the relevance between your measurement and ambient temperatures. Does it matter? On the other hand, did it rain anytime?

Authors Response: In the light of the current study we find it relevant to provide a brief description of weather conditions at the measurement sites. High ambient temperatures is an indicator of highly turbulent conditions, favorable for dust transport and increased deposition. We do not discuss wet deposition of dust directly, but we note that hazy humid conditions during some mornings probably contribute to the deposition of dust, justifying the discussion of the dewpoint. In 2015, there were only a few light rainfall events at KAUST, and as such not of much importance to our measurements.

23. Page 9, "Gravimetric Analysis": In Figure 5, you did not discuss the peak in dust deposition in August in DT3. This maximum is 3 times higher than the annual mean, and 30% higher than DT4. Why such difference between DT3 and DT4 and all other sites in August? This factor 3 difference will affect your analysis, but first you will have to know its origin. Is it construction?

Authors Response: Added sentence. The higher deposition rate of DT3 for August compared to DT4 is ascribed to the fact that the former is about 100 m closer to construction material handling activities during that month.

24. What is the mineralogy or chemical components of construction dust? Do you see its signature in your data?

Authors Response: We do not have chemical or mineralogical signatures for construction dust. However, it is expected to contain variable amounts of local dirt road dust, sand, and cement products. The composition thereof will vary substantially with each construction activity. The best we can do is to ascribe elevated mass concentration at any one site to the contribution by construction dust (or marine salt in some cases. This explains the anomalously high concentrations at individual sites (Figure 5, August, September, October).

25. Page 10, Line 5-7. Reformulate the sentence. => The dust deposition measured in Kuwait on the other hand, varies substantially between sites due to the contribution from disturbed soils in lowlands during periods of northwesterly Shamal.

Authors Response: Rephrased to read "The dust deposition measured in Kuwait on the other hand, varies substantially between sites due to the contribution from disturbed soils in lowlands during periods of northwesterly *Shamal* winds."

26. Page 10, Line 8-13: Remove, this is repeating what is already in Table 3.

Authors Response: Removed these lines

27. Page 10, AERONET: You should either use the Angstrom exponent to screen out non-dusty days or use SDA coarse mode optical depth.

Authors Response: The KAUST campus is located in the heart of the dust source region. The average optical depth is about 0.4 and there are virtually no non-dusty days. According to CALIPSO, the ratio of "not dust" to "dust" successful retrievals in this region is 2.04% indicating that dust dominates all other types of aerosol (Osipov et al., 2015). Kalenderski and Stenchikov (2016) demonstrated that over the Arabian Peninsula the contribution of non-dust aerosols in the visible optical depth does not exceed 10%. Khan et al. (2015) compared the contributions of fine and coarse modes of aerosols over the similar dust source region in Sahara and showed that the coarse mode dominates the optical depth. Having said this, we consider it unnecessary to further recalculate the AERONET observations to improve the correlation between deposition and AOD because, as we mentioned in the paper, there are other important factors that could affect this relation, which in the context of this paper is treated only qualitatively

28. Page 10, Line 26: "dust particles are predominantly from local sources" but in Abstract you wrote "dust deposits along the Red Sea coast are a mixture of dust emission from local soils, and soils imported from distal sources." This is contradictory.

Authors Response: Replaced "predominantly" with "partly"

29. Page 10 Line 28-30: You should remove and screen AERONET data using low Angstrom values, or use AERONET SDA coarse mode AOD.

Authors Response: See response under comment 27

30. Page 11, Line 10: Merge Figure 6 and 7.

Authors Response: Since the three variables (deposition rate, visibility, AOD) have different units, it will be confusing to the reader have them together on a single plot. The two diagrams (Figures 6, 7) are combined as Figure 6a and 6b.

31. Page 11, Line 13-15: This is irrelevant for this study. Remove.

Authors Response: Sentence and references removed

32. Page 11, Mineral analysis: A point that needs clarification is the units. Are all the % values given by mass or by number? In section 4.6, it is specified by mass. This means that you should be able to provide the mass size distribution!

Authors Response: Correct. The mineral analysis by XRD and chemistry are all mass percentages. To be comparable, the particle size distributions are now presented and discussed as both mass (volume) (Appendix A) and number (Supplement C).

33. Page 12, Line 11: "DT1". Why only one site and not all of them? Why is there no standard deviation in Figure 9a. What is the error associated with these measurements?

Authors Response: See below. Additional SEM analysis were recently performed and are now included (Appendix A, Supplement C, Table 4)

34. Page 12, Line 21-22. "... Figure 9 could be used to distinguish the contribution of PM10 in deposited mass and reconcile models with observations." Are you suggesting that modelers use a ruler to derive approximately some fraction of particle numbers, then assume some density and shape for each sizes? This is an inadequate method. You should provide the values of each dots of Figure 9a in a Table, as well as the errors associated with the measurement, and assumptions on shape and density.

Authors Response: We replaced the single size distribution plot (Fig. 9a) with comparable 12 monthly size distribution plots (Fig. 8a), expressed both as number and volume percentages (Appendix A, Supplement C, Table 4). It was shown that the deposition rates at all the sites are similar, except for the added contributions from local marine and local construction at some sites. A table (Table 4) with distribution statistics, assessments of the < 2.5  $\mu$ m and < 10  $\mu$ m mass fractions are presented. The table also provides information on particle size distribution variability by month.

35. Page 13, Line 3 "soils and dusts" => "soil and dust"

Authors Response: Corrected to read "soil and dust"

36. Page 14, Line 13: "This paper has as its goal the provision" Needs to be reformulated

Authors Response: Rephrased to read "This paper provides new mineralogical, physical and chemical results on deposition samples collected at the KAUST campus during 2015".

37. Page 14, Line 16: "meant to be used for validating dust mass balance." No. The method suggested in Section 4.5 is inadequate.

Authors Response: Deleted "and is meant to be used for validating dust mass balance in the meteorological models with the dust component".

38. Page 15, Line 3-4 contradicts Line 5-6.

Authors Response: Deleted the sentence in line 3 and 4 reading "It is therefore not feasible to explicitly relate the deposition samples to the coastal soils from chemical and mineralogical results on their own."

39. Page 15, Line 12: you may want to add "construction dust".

Authors Response: Added", local construction dust.."

40. Page 15, Line 13: "To better represent." In what sense? By models? This may be a good place to add that the "inclusion of particle size into mineralogical and chemical analysis will provide more effectively data for the modeling community."

Authors Response: Replaced "represent" with "To better model the dust being deposited ......". Added as suggested "Also, inclusion of particle size with mineralogical and chemical measurements will provide more effective data for the modeling community."

41. Page 25, Line 4: "Locality" => Position

Authors Response: Rephrased as suggested

42. Page 25, Line 5: "campus. . .Sea" => on the Arabian Peninsula (red dot)

Authors Response: Rephrased as suggested

43. Page 27: Provide a Figure caption rather than an analysis of the Figure.

Authors Response: Unsure what is meant with analysis of the Figure 3.

44. Page 28: add the color of each lines in the Figure caption in parenthesis.

Authors Response: Parenthesis added in Figure 4 caption

45. Page 30 & 31: Merge the 2 Figures.

Authors Response: Since the three variables (deposition rate, visibility, AOD) have different units, it will be confusing to the reader have them together on a single plot. The two diagrams (Figures 6,7) are retained as such, but combined as one figure (Fig. 6a,b).

46. Page 32: Is there a possibility to split between fine, coarse and super-coarse modes?

Authors Response: The 0.2 -2.5  $\mu$ m, 0.2-10  $\mu$ m and >10 $\mu$ m mass percentages were assessed from the mass percentage/particle size distribution plots, shown in Table 4.

#### 47. Page 33, Figure 9a: Error bars.

Authors Response: Figure 9a is replaced by an averaged plot for the 12-month sampling period, including the uncertainty bars (Fig 8a).

#### References:

Ginoux, P., Prospero, J.M., Gill, T.E., Hsu, N.C. and Zhao, M., 2012. Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products. Reviews of Geophysics, 50(3).

Prospero, J.M., Ginoux, P., Torres, O., Nicholson, S.E. and Gill, T.E., 2002. Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product. Reviews of geophysics, 40(1).

Schulz, M., Prospero, J.M., Baker, A.R., Dentener, F., Ickes, L., Liss, P.S., Mahowald, N.M., Nickovic, S., García-Pando, C.P., Rodríguez, S. and Sarin, M., 2012. Atmospheric transport and deposition of mineral dust to the ocean: implications for research needs. Environmental science & technology, 46(19), pp.10390-10404.

Kalenderski, S., and Stenchikov, G.: High-resolution regional modeling of summertime transport and impact of African dust over the Red Sea and Arabian Peninsula, Journal of Geophysical Research: Atmospheres, 121, 6435–6458, doi:10.1002/2015JD024480, 2016.

Khan, B., Stenchikov, G., Weinzierl, B., Kalenderski, S., and Osipov, S.: Dust plume formation in the free troposphere and aerosol size distribution during the Saharan Mineral Dust Experiment in North Africa, Tellus B: Chemical and Physical Meteorology, 67, 1, 27170, doi: 10.3402/tellusb.v67.27170, 2015.

Osipov, S., Stenchikov, G., Brindley, H., and Banks, J.: Diurnal cycle of the dust instantaneous direct radiative forcing over the Arabian Peninsula, Atmospheric Chemistry and Physics, 15, 9537–9553, doi:10.5194/acp-15-9537-2015, 2015.

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# Interactive comment on "Physical and chemical properties of deposited airborne particulates over the Arabian Red Sea coastal plain" by Johann Engelbrecht et al.

#### **Anonymous Referee #2**

The manuscript presents information on monthly resolved dust deposition rates as well as the mineralogical, chemical, and elemental composition of the deposited dust. The data are based on monthly accumulated samples over 13 months from six sites on the campus of King Abdullah University of Science and Technology (KAUST), located on the Saudi Arabian coastal plain near the Red Sea. These are new data from an understudied region, and the provided information is very valuable for other researchers. It will help with a better assessment of the effects of dust from this region on the environment and human health as well as for the evaluation and constraining of dust simulated with models. The manuscript is clearly written and well structured. It should be published after taking into consideration following few minor points.

1. Page 4, lines 19-25: Information should be provided where the climatological data were sourced.

Authors Response: Data sources added.

With the exception of the area around Jazan in the south, which is impacted by the Indian Ocean monsoon, the Red Sea coastal region has a desert climate characterized by extreme heat. Temperatures measured at the KAUST campus reach 43° C during the summer days, with a drop in night-time temperatures on average of more than 10° C. Also, although the extreme temperatures here are moderated by the proximity of the Red Sea, summer humidity is often 85% or higher during periods of the northwesterly *Shamal* winds. Rainfall diminishes from an annual average of 133 mm at Jazan in the south to 56 mm at Jeddah, and 30 mm at Tabuk in the north <a href="http://worldweather.wmo.int/en/city.html?cityId=699">http://worldweather.wmo.int/en/city.html?cityId=699</a>.

2. Page 5, line 7: Add Scanza et al. (2015) as reference.

Authors Response: Reference was added.

**3.** Page 8, lines 13-24: The authors should mention a possible bias in the results from applying the X-ray diffraction (XRD) technique. XRD is most effectively detecting crystalline material. This could lead to an overestimation of the abundance of those dust mineral types that tend to have a regular crystal structure, like tectosilicates, relative to other minerals such as phyllosilicates whose mass can have a significant and varying amorphous fraction (*Formenti et al.*, 2008; *Kandler et al.*, 2009).

Authors Response: Added text following line 24.

A likely bias in the results from applying the X-ray diffraction (XRD) technique together with the RIR method is widely recognized, and therefore our methodology is considered to be semi-quantitative at best. Chung (1974) recognized that if the RIRs of all the crystalline phases in a mineral mixture are known, the sum of all the fractions should add to 100%. However, XRD is effective at measuring crystalline phases such as quartz, calcite, and feldspars, and less so for partly crystalline and amorphous phases including some layered silicates such as clays as well as many hydrous minerals. This could lead to an overestimation of the abundance of the crystalline mineral types in the dust, compared to partly crystalline and amorphous phases (*Formenti et al.*, 2008; *Kandler et al.*, 2009). Other discrepancies could occur from preferred orientation of layered silicates in the sample mounts, and the dust samples were loaded into side-mount holders to minimize this effect.

**4. Page 8, line 25**: "Northwesterly Shamal winds prevailed during all twelve months of 2015 (Fig. 3)"

What about November? It looks like from Figure 3, as an exception, that northeasterly winds were more frequent in that month, although they didn't quite reach the maximum strength of the northwesterly winds.

Authors' Response: Sentence added.

Although the northeasterly winds were more frequent in November, they did not reach the strength of the northwesterlies.

#### References

Formenti, P., J. L. Rajot, K. Desboeufs, S. Caquineau, S. Chevaillier, S. Nava, A. Gaudichet, E. Journet, S. Triquet, S. Alfaro, M. Chiari, J. Haywood, H. Coe, and E. Highwood (2008), Regional variability of the composition of mineral dust from western Africa: Results from the AMMA SOP0/DABEX and DODO field campaigns, *J. Geophys. Res.*, 113, D00C13, doi:10.1029/2008JD009903.

Kandler, K., L. Schütz, C. Deutscher, M. Ebert, H. Hofmann, S. Jäackel, R. Jaenicke, P. Knippertz, K. Lieke, A. Massling, A. Petzold, A. Schladitz, B. Weinzierl, A. Wiedensohler, S. Zorn, and S. Weinbruch (2009), Size distribution, mass concentration, chemical and mineralogical composition and derived optical parameters of the boundary layer aerosol at Tinfou, Morocco, during SAMUM 2006, *Tellus B*, 61 (1), 32{50, doi:10.1111/j.1600-0889.2008.00385.x.

Scanza, R. A., N. Mahowald, S. Ghan, C. S. Zender, J. F. Kok, X. Liu, Y. Zhang, and S. Albani (2015), Modeling dust as component minerals in the Community Atmosphere Model: development of framework and impact on radiative forcing, *Atmos. Chem. Phys.*, 15, 537-561, doi:10.5194/acp-15-537-2015.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-231-RC2, 2017 © Author(s) 2017. This work is distributed under the Creative Commons Attribution 3.0 License.

### Interactive comment on "Physical and chemical properties of deposited airborne particulates over the Arabian Red Sea coastal plain" by Johann Engelbrecht et al.

#### **Anonymous Referee #3**

Received and published: 15 June 2017

The paper presents and analyzes the mineralogical, physical and chemical composition of dust deposited samples at six sites on the KAUST campus. KAUST is located on the Red sea coastal plain of Saudi Arabia. Monthly samples were collected between December 2014 and December 2015. The monthly deposition trends were compared to visibility and sun photometric measurements and to previous mineralogical analysis of soil samples from nearby dust sources. The paper concludes that dust deposits along the Red Sea coast are a mixture of dust emissions from local soils and soils imported from distal sources.

As the authors mention in the abstract and the paper, the type of information obtained has no precedent in the region and can be useful for modelers and other impact areas. This supports publication.

1. In order to make the data useful to others, the authors may include, not only the oxide data in the appendix, but also the mineralogical data and size distribution data. I believe this is the main reason for publication of this manuscript.

Authors Response: Particle size distribution plots of 12 deposition samples collected monthly at the KAUST campus throughout the 2015 period are added as Appendix A. The mineralogical data are added as a table in Appendix B. The chemical data tables are renumbered as Appendix C.

2. While the data will be useful, I find the paper itself overly descriptive. The paper presents the data and a preliminary exploratory data analysis from which it is difficult to extract new insights. The comparison with AERONET, visibility and meteorology is rather superficial. A good example of my argument is Figure 4, where the authors present humidity and temperature data but it is not very clear why they do so. The comparison with AERONET is performed with total AOD. It would have been more appropriate to filter the data by low Angstrom Exponent or use coarse mode AOD.

Authors Response: : See Referee #1, authors response under paragraph 27

3. One of the main conclusions is that dust in the Red Sea is a mixture of dust from local soils and dust imported from distal sources, which is something that is already well-known; the paper makes no attempt to quantify the respective contributions. An additional concern that I have is the selection of the sites to measure dust deposition. It is not very clear what is the impact of local construction activities. The authors should make a clear statement in that respect.

Authors Response: Source apportionment is considered to be a further step in our research, to be documented in a following paper. As an approximation the sampler with the lowest deposition rate can be considered to have negligible or the least amount of local dust and sea salt. In the months of December 2014, January, April, March, June, July, and December 2015, the deposition rates at the four sites were similar, and considered to have no or negligible amounts of dust from local construction, campus roads, marine salt, or other particulates.

Other than that, I think that the paper is well written and well structured. I did not find minor errors or typos.

## First marked up revision

# Physical and chemical properties of deposited airborne particulates over the Arabian Red Sea coastal plain

Johann P. Engelbrecht<sup>1,2</sup>, Georgiy Stenchikov<sup>1</sup>, P. Jish Prakash<sup>1</sup>, Traci Lersch<sup>3</sup>, Anatolii Anisimov<sup>1</sup> and Illia 5 Shevchenko<sup>1</sup>

<sup>1</sup>King Abdullah University of Science and Technology (KAUST), Physical Science and Engineering Division (PSE), Thuwal, 23955-6900, Saudi Arabia.

<sup>2</sup>Desert Research Institute (DRI), Reno, Nevada 89512-1095, U.S.A.

<sup>3</sup> RJ Lee Group Inc., Monroeville, Pennsylvania 15146, U.S.A.

10 Correspondence to: Johann P. Engelbrecht (johann@dri.edu)

#### Abstract

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Mineral dust is the most abundant aerosol, having a profound impact on the Global energy budget. This research continues our previous studies performed on surface soils in the Arabian Peninsula and aims at analyzing mineralogical, physical and chemical composition of dust deposits from the atmosphere at the Arabian Red Sea coast. For this purpose, aerosols deposited from the atmosphere are collected during 2015 at six sites on the campus of the King Abdullah University of Science and Technology (KAUST) situated on the Red Sea coastal plain of Saudi Arabia and subjected to the same chemical and mineralogical analysis we conducted on soil samples. Frisbee deposition samplers with foam inserts were used to collect dust and other deposits, for the period December 2014 to December 2015. The average deposition rate measured at KAUST for this period was 14 g m<sup>-2</sup> per month, with lowest values in winter and increased deposition rates in August to October. The particle size distributions provide assessments of < 10  $\mu$ m and < 2.5  $\mu$ m dust deposition rates, and it is suggested that these furthermore represent proxies for PM<sub>10</sub> (coarse) and PM<sub>2.5</sub> (fine) particle size fractions, respectively.

XRD analysis of a subset of samples confirms variable amounts of quartz, feldspars, micas, and halite, with lesser amounts of gypsum, calcite, dolomite, hematite, and amphibole. Freeze-dried samples were re-suspended onto the Teflon\* filters for elemental analysis by XRF, while splits from each sample were analyzed for water soluble cations and anions by Ion Chromatography. The dust deposits along the Red Sea coast are most probably a mixture of dust emissions from local soils, and soils imported from distal dust sources. It is not possible to identify the exact origin of deposition samples from the mineralogical and chemical results alone. These aerosol data are the first of their kind from the Red Sea region. They will help assess their potential nutrient input into the Red Sea, as well the impact on human health, industry, and solar panel efficiency. These data will also support dust modeling in this important dust belt source area, by better quantifying dust mass balance and optical properties of airborne dust particles.

#### 25 Keywords

Dust mineralogy, chemistry, particle size distribution Frisbee deposition samplers Freeze-dry dust retrieval Dust deposition rates, AOD and visibility Proxies for PM<sub>10</sub> and PM<sub>2.5</sub> deposition rates

#### 1. Introduction

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Dust emission and deposition modeling and measurements are required for the assessment of the dust mass budget. Both emission and deposition are under constrained in atmospheric dust models, leading to large uncertainties (Bergametti and Forêt, 2014; Schulz et al., 2012). To improve simulations, the above authors and others suggested the establishment of dust deposition networks in the vicinity of and away from dust source regions, operating throughout the year. In this paper we are presenting results from a network of dust deposition samplers located on the campus of the King Abdullah University of Science and Technology (KAUST) along the Red Sea coast of Saudi Arabia. This is an important dust source region (Ginoux et al., 2012; Prospero et al., 2002), the effect of which extends thousands of kilometers downwind. To better characterize optical, microphysical, and health effects of dust aerosols we conducted detailed chemical, mineralogical and particle size analysis of deposition samples collected from the air.

#### 1.1 Importance of mineral dust

Mineral dust is the most abundant atmospheric aerosol, primarily from suspended soils in arid and semiarid regions on Earth (Buseck et al., 2000; Washington and Todd, 2005; Goudie, 2006; Muhs et al., 2014), including deserts of the Arabian Peninsula (Edgell, 2006). Dust aerosols profoundly affect climate (Haywood and Boucher, 2000; Hsu et al., 2004; Kumar et al., 2014), cloud properties (Twomey et al., 1984; Wang et al., 2010; Huang et al., 2006), visibility (Kavouras et al., 2009; Moosmüller et al., 2005), air quality (Hagen and Woodruff, 1973), atmospheric chemistry and mineralogy (Sokolik and Toon, 1999; Kandler et al., 2007), biogeochemical cycles in the ocean and over land (Jickells et al., 2005; Mahowald, 2009), human health (Bennett et al., 2006; Bennion et al., 2007; De Longueville et al., 2010; Menéndez et al., 2017), and agriculture (Fryrear, 1981; Nihlen and Lund, 1995).

A further important implication of dust emission/deposition processes is associated with the harvesting of the solar renewable energy in the desert areas. Dust deposits on solar panels are known to have a severe detrimental effect on the efficiency of photovoltaic systems (Goossens and Van Kerschaever, 1999; Hamou et al., 2014; Mejia et al., 2014; Rao et al., 2014; Sulaiman et al., 2014; Ilse et al., 2016), with its adverse effects depending on mineral composition and atmospheric conditions.

#### 1.2 Importance of dust mineralogy

The importance of dust mineralogy was long been recognized (Engelbrecht et al., 2016), but only recently the explicit transport of different mineralogical species is implemented in climate models (Perlwitz et al., 2015a, b; Scanza et al., 2015)

The mineralogy and chemical composition of dust generated from the Red Sea coastal region remains uncertain. The Red Sea coastal plain is a narrow highly heterogeneous piedmont area, and existing soil databases do not have the spatial resolution to represent it adequately (Nickovic et al., 2012).

The specific objective of the present study is to examine mineralogical, chemical and morphological information of deposition samples collected on the KAUST campus. This will help to better quantify the ecological impacts, health effects, damage to property, and optical effects of dust blown across this area (Engelbrecht et al., 2009a, b; Weese and Abraham, 2009). Knowledge of the mineralogy of the dust deposits will provide information on refractive indices, which can be used to calculate dust optical

properties, providing input into radiative transfer models, and to assess the impact of dust events on the Red Sea and adjacent coastal plain.

#### 1.3 Previous dust studies in the region

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This research complements our dust studies performed in the Arabian Peninsula (Engelbrecht et al., 2009a; Kalenderski et al., 2013; Prakash et al., 2015; Prakash et al., 2016) and globally (Engelbrecht et al., 2016).

The Arabian Peninsula is one of Earth's major sources of atmospheric dust, contributing as much as 11.8% (22–500 Mt  $a^{-1}$ ) of the total (1,877–4000 Mt  $a^{-1}$ ) global dust emissions (Tanaka and Chiba, 2006). The Red Sea, being enveloped by the Arabian and African deserts is strongly impacted by windborne mineral dust. Along with profound influence on the surface energy budget over land and the Red Sea (Kalenderski et al., 2013; Osipov et al., 2015; Brindley et al., 2015), dust is an important source of nutrients, more so for the oligotrophic northern Red Sea waters (Acosta et al., 2013). From preliminary assessments it is estimated that 5 to 6 major dust storms per year impact the Red Sea region, depositing about 6 Mt of mineral dust into the Red Sea (Prakash et al., 2015). Simulations and satellite observations suggest that the coastal dust contribution to the total deposition flux into the Red Sea could be substantial, even during fair weather conditions (Jiang et al., 2009; Anisimov et al., 2017). Therefore, the correct representation of the regional dust balance over the Red Sea coastal plain is especially important. Here we specifically focus on the dust deposition in this area, which helps to constrain the dust mass balance, as well as the dust mineralogy and chemical composition. Dust sources impacting on the Arabian Red Sea coastal region were shown to vary by season, coming from local haboobs and low level jets, delivered from the Tokar delta of Sudan in summer (Kalenderski and Stenchikov, 2016), and transported from the west coast of the Arabian Peninsula (Kalenderski et al., 2013).

Minerals previously identified in continental soils from Middle East dust generating regions include quartz, feldspars, calcite, dolomite, micas, chlorite, kaolinite, illite, smectite, palygorskite, mixed-layer clays, vermiculite, iron oxides, gypsum, hornblende and halite (Engelbrecht et al., 2009b; Engelbrecht et al., 2016; Goudie, 2006; Prakash et al., 2016; Pye, 1987; Scheuvens and Kandler, 2014). It could be expected that similar mineral assemblages would occur in variable proportions in the dust deposition samples collected in the region.

#### 2. Meteorology and climate

With the exception of the area around Jazan in the south, which is impacted by the Indian Ocean monsoon, the Red Sea coastal region has a desert climate characterized by extreme heat. Temperatures measured at the KAUST campus reach 43° C during the summer days, with a drop in night-time temperatures on average of more than 10° C. reaching 43° C during the summer days, with a drop in night-time temperatures on average more than 10° C. Although the extreme temperatures here are moderated by the proximity of the Red Sea, summer humidity is often 85 % or higher during periods of the northwesterly Shamal winds. Rainfall diminishes from an annual average of 133 mm at Jazan in the south to 56 mm at Jeddah, and 24 mm at Tabuk in the north. <a href="http://worldweather.wmo.int/en/city.html?cityId=699">http://worldweather.wmo.int/en/city.html?cityId=699</a>.

Vegetation is sparse, being restricted to semi-desert shrubs, and acacia trees along the ephemeral rivers (wadis), providing forage for small herds of goats, sheep and dromedary camels.

During infrequent but severe rainstorms, run-off from the escarpment along wadis produce flash floods in lowland areas. With such events, fine silt and clay deposits are formed on the coastal plain, which are transformed into dust sources during dry and windy periods of the year. The resultant dust is transported and deposited along the coastal plain itself and adjacent Red Sea, by prevailing northwesterly to southwesterly winds, with moderate breezes (wind speed >5.5 m s<sup>-1</sup>) at the coast (http://www.windfinder.com/weather-maps/report/saudiarabia#6/22.999/34.980).

#### 3. Objectives

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This study is meant to complement the recently published papers by our research group that characterize the effect of dust storms (Prakash et al., 2015; Kalenderski et al., 2013), evaluate radiative effect of dust (Osipov et al., 2015), analyze soils from the Red Sea coastal plain (Prakash et al., 2016) and dust emissions in the same region (Anisimov et al., 2017). It aims to provide mineralogical, physical and chemical compositions of deposition samples collected largely during 2015 at six sites on the campus of KAUST, located approximately 80 km north of Jeddah, along the central part of the Red Sea coastal plain of Saudi Arabia, (Fig. 1).

#### 3.1 Regional dust sources

The coastal plains of the Arabian Peninsula along the Red Sea and Persian Gulf are among the most populated areas in this region, hosting several major industrial and residential centers. Airborne dust profoundly affects human activities, marine and land ecosystems, climate, air-quality, and human health. Satellite observations suggest that the narrow Red Sea coastal plain to be an important dust source-province, augmented by the fine-scale sediment accumulations, scattered vegetation, and variable terrain. Airborne dust inevitably carries the mineralogical and chemical signature of a parent soil (Prakash et al., 2016). The purpose of a previous study on 13 soil samples from the Arabian Red Sea coastal area (Prakash et al., 2016) was to better characterize their mineralogical, chemical and physical properties, which in turn improve assessment of dust being deposited in the Red Sea and on land, affecting environmental systems and urban centers. It was found that the Red Sea coastal soils contain major components of quartz and feldspar, as well as lesser but variable amounts of amphibole, pyroxene, carbonate, clays, and micas, with traces of gypsum, halite, chlorite, epidote and oxides. The mineral assemblages in the soil samples were ascribed to the variety of igneous and metamorphic provenance rocks of the Arabian Shield forming the escarpment to the east of the coastal plain.

#### 4. Sampling and analysis

Anisimov et al. (2017) estimated that the eastern Red Sea coastal plain emits about 5–6 Mt of dust annually. Due to its close proximity, a significant portion of this dust is likely to be deposited into the Red Sea, which could be comparable in amount to the estimated annual deposition rate from remote sources during major dust events (Prakash et al., 2015). Therefore, we expect that the total dust deposition into the Red Sea is of the order of 10 Mt a<sup>-1</sup>, but this figure still needs to be confirmed.

In the past few decades wind tunnel and field tests to compare their efficiencies, had been performed on different designs of deposition samplers and sand traps, including marble dust collectors (MDCO), inverted Frisbees, and glass surfaces (Goossens and Rajot, 2008; Sow et al., 2006; Goossens et al., 2000; Goossens and Offer, 2000). Most of the experiments performed in wind tunnels failed to completely mimic the field conditions, which resulted in an underestimation of the dust deposition, more so for the

<10 µm size fraction (Sow et al., 2006). Based on the field evaluations by Vallack (1995) and suggestions by Vallack and Shillito (1998) the decision was taken to deploy inverted Frisbee samplers with foam inserts.

At each sampling site the particulate deposits were collected into a 227 mm diameter inverted Frisbee 5 dust deposit sampler, each with a polyester foam insert and bird strike preventers (Hall et al., 1993; Vallack and Chadwick, 1992, 1993; Vallack and Shillito, 1998) (Fig. 2). The purpose of the foam insert is to enhance the particulate collection capacity of the dust gauge (Vallack and Shillito, 1998) by better collecting and retaining wet (from fog, dew, rain) and dry, fine and coarse particles being deposited into the inverted Frisbee dish, under stable meteorological conditions, during severe dust events,

10 northwesterly Shamal winds, and by daily coastal winds,

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For the period December 2014 to March 2015, four Frisbee samplers were located at the New Environmental Oasis (NEO) site, about 50 m apart. The gravimetric information from the four samplers were similar, with small variations amongst them ascribed to the impact from local construction activities. Due to the similarity of these gravimetric results, and to obtain a better representation of dust deposition onto the KAUST campus, two of the samplers (DT1 and DT2) were moved in March, the first (DT1) to a residential area and the other (DT2) to the quay adjacent to the Coastal & Marine Resources Core Lab (CMOR) (Table 1). (Site meta-data provided in the Supplement A).

The deposition samples were collected for intervals of a calendar month, starting in December 2014 and ending December 2015. At the end of each month, the samples are retrieved by flushing the dust deposit with distilled water from the foam insert and collection dish into the downpipe and plastic bottle. Both the insoluble particles and dissolved salts in the water suspension are retrieved in the laboratory by a freeze-drying (sublimation) procedure.

A total of 52 deposition samples were collected at the six sampling sites on the KAUST campus (Fig. 1b) over a period of 13 months, largely in 2015. Representative subsets of these samples were selected for X-ray diffraction (XRD), (27 samples) and chemical analysis (29 samples).

Freeze-dried sample splits were re-suspended in the laboratory onto Teflon filters, for elemental analysis by X-ray Fluorescence (XRF) spectrometry by using a miniaturized version of a dust entrainment facility (Engelbrecht et al., 2016) http://www.dri.edu/atmospheric-sciences/atms-laboratories/4185dust-entrainment-and-characterization-facility ). With this modified system the dust sample is drawn into a vertically mounted tubular dilution chamber, and the re-suspended dust collected onto a 47 mm diameter Teflon® filter, for chemical analysis.

The samples re-suspended onto the Teflon® filters were chemically analyzed for elemental content by XRF, including for Si, Ti, Al, Fe, Mn, Ca, K, P, V, Cr, Ni, Cu, Zn, Rb, Sr, Y, Zr, and Pb (US EPA, 1999). Splits of about 2 mg from each freeze-dried sample were analyzed for water soluble cations of sodium (Na<sup>+</sup>), potassium (K<sup>+</sup>), calcium (Ca<sup>2+</sup>) and magnesium (Mg<sup>2+</sup>), and anions of sulfate (SO<sub>4</sub><sup>2-</sup>), chloride (Cl<sup>-</sup>), phosphate (PO<sub>4</sub><sup>3-</sup>) and nitrate (NO<sub>3</sub>-), by Ion Chromatography (IC) (Chow and Watson, 1999).

A subset of 27 samples from the total of 52 samples, representing all months of the year, was selected for X-ray diffraction (XRD) analysis. XRD is a non-destructive technique particularly suited to identifying and characterizing minerals such as quartz, feldspars, calcite, dolomite, clay minerals and iron oxides, in fine soil and dust. Dust reactivity in seawater as well as optical properties depend on its mineralogy, e.g. carbonates and sulfates are generally more soluble in water than silicates such as feldspars, amphiboles, pyroxenes or quartz. A Bruker D8 $^{\circ}$  X-ray powder diffraction system was used to analyze the mineral content of the dust deposition samples. The diffractometer was operated at 40 kV and 40 mA, with Cu K $\alpha$  radiation, scanning over a range of 4-50 $^{\circ}$  2 $\theta$ . The Bruker Topas $^{\circ}$  software and relative intensity ratios (RIRs) were applied for semi-quantitative XRD analyses of the dust deposition samples (Rietveld, 1969; Chung, 1974; Esteve et al., 1997; Caquineau et al., 1997; Sturges et al., 1989).

A likely bias in the results from applying the X-ray diffraction (XRD) technique, together with the RIR method is widely recognized, and therefore our methodology is considered to be semi-quantitative at best. Chung (1974) recognized that if the RIRs of all the crystalline phases in a mineral mixture are known, the sum of all the fractions should add to 100%. However, XRD is effective at measuring crystalline phases such as quartz, calcite, and feldspars, and less so for partly crystalline and amorphous phases, including some layered silicates such as clays as well as many hydrous minerals. This could lead to an overestimation of the abundance of the crystalline mineral species in the dust, compared to partly crystalline and amorphous phases (Formenti et al., 2008; Kandler et al., 2009). Other discrepancies could occur from preferred orientation of layered silicates in the sample mounts. To minimize this effect the dust samples were loaded into side-mount holders.

Electron microscopy provided information on the individual particle size and shape of micron-size particles, important for determining the optical parameters for modeling of dust (Moosmüller et al., 2012). The scanning electron microscope (SEM) based individual particle analysis was performed on a subset of twelve deposition samples collected for each month of 2015. For each sample, the portion of deposition sample was suspended in isopropanol and dispersed by sonication. The suspension was vacuum filtered onto a 0.2 µm pore size polycarbonate substrate. A section of the substrate was mounted onto a metal SEM stub with colloidal graphite adhesive. The sample mounts were sputtercoated with carbon to dissipate the negative charge induced on the sample by the electron beam. The automated analysis was conducted on a Tescan MIRA 3° field emission scanning electron microscope (FE-SEM) by rastering the electron beam over the sample while monitoring the resultant combined backscattered electron (BE) and secondary electron (SE) signals. Based on the grayscale levels, preset threshold values segmented the image into particles of interest and background. The system was configured to automatically measure the size and shape of anywhere from 5,000 to 15,000 particles per sample measuring > 0.2 µm in average diameter. A digital image was acquired of each particle, for measurement, and stored for subsequent review. Size measurements were based on Feret diameters obtained from the projected area of each particle, by tracing their outer edges. This information was used to calculate the shape-dependent particle volumes. The particles were grouped into "bins" by their size. The field emission electron source allows for high magnifications and sharp secondary electron images (SEI), as well as for the detailed study of particle size distributions.

#### 5. Results

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#### 5.1 Meteorology

Northwesterly *Shamal* winds prevailed during all twelve months of 2015 (Fig. 3). Four to five severe dust storms lasting three to five days each, contributed to hot humid conditions during the summer months. Weaker northeasterly winds were experienced in October and November of that year. Although the

northeasterly winds were more frequent in November, they did not reach the maximum strength of the northwesterlies.

The first four months in the second half of 2015 experienced the highest ambient temperatures (Table 2), with an average temperature of 35° C for August followed by 34° C for September, bracketed by 33° C for both July and October. The highest single temperature was 43° C, recorded in October, with the coolest temperature of 17° C in January of that year. The range of temperatures was the greatest through fall, winter and spring, with large diurnal temperature fluctuations during these seasons. The humidity at KAUST is consistently high (Table 2), with averages varying from 57 % for December and 61% for January, to as high as 82 % for August and 80% for September. Dewpoints were calculated for each set of hourly measurements, applying the August-Roche-Magnus approximation (Alduchov and Eskridge, 1996; August, 1828; Magnus, 1844). The highest dewpoint temperatures were calculated in August (31° C) and September (30° C) while the month with the greatest frequency of humidity measurements (96) in excess of 90 % was also recorded in August (Table 2, Fig. 4). The lowest monthly frequency (4) for humidity exceeding 90 % was December. In 2015, there were only a few light rainfall events at KAUST, and as such not of much importance to our measurements.

#### 5.2 Gravimetric analysis

With a few exceptions, the monthly gravimetric measurements from the four samplers (DT1 – DT4) are comparable (Fig. 5), changing similarly by month and season. The deposition rates were at their lowest for December 2014 (avg. 4 g m<sup>-2</sup>), increasing steadily for four months to a peak value for March, 2015 (avg. 20 g m<sup>-2</sup>) before decreasing over the subsequent four months to a low for July (avg. 5 g m<sup>-2</sup>). The deposition rates increased sharply for August (28 g m<sup>-2</sup>), September (23 g m<sup>-2</sup>) and October (28 g m<sup>-2</sup>), before diminishing in November (14 g m<sup>-2</sup>) and December (11 g m<sup>-2</sup>). The NEO terrain is close to several building construction sites, about 400 m to the east and southeast of the installed deposition samplers, which periodically created substantial amounts of local airborne dust. This, together with the windy conditions are held responsible for elevated dust concentrations measured at the two NEO sites (DT3, DT4). The higher deposition rate at DT3 for August, compared to DT4, is ascribed to the fact that the former sampler is about 100m closer to construction material handling activities during that month. Wind-blown sea spray during stormy conditions was responsible for elevated deposition levels of sea salt at the CMOR (DT2) quay-side site, for the months of September and October 2015.

Bearing in mind that the dust deposition samplers, sampling procedures, as well as conditions and sampling periods were different to those of this study, some comparisons to similar studies in desert regions are listed in Table 3. The deposition rates from this study, both on average (14 g m<sup>-2</sup> month<sup>-1</sup>) and in range (4-28 g m<sup>-2</sup> month<sup>-1</sup>), were found to be similar to those previously recorded by Offer and Goossens (2001) in the Negev Desert, Israel (average 17 g m<sup>-2</sup> month<sup>-1</sup>, range 10-25 g m<sup>-2</sup> month<sup>-1</sup>), and West Niger (Goossens and Rajot, 2008) (average 13 g m<sup>-2</sup> month<sup>-1</sup>, range 6-21 g m<sup>-2</sup> month<sup>-1</sup>). A campaign in the Saudi Arabian capital of Riyadh (Modaihsh, 1997; Modaihsha and Mahjoub, 2013) during the dusty months of January to March showed average monthly deposition rates of 42 g m<sup>-2</sup>, and a range of 20-140 g m<sup>-2</sup>. The dust deposition measured in Kuwait on the other hand, varies substantially between sites due to the contribution from disturbed soils in lowlands during periods of north-westerly *Shamal* winds. For these reasons the dust loadings varied during the campaigns in the N-E Bay area of Kuwait during the 2002/3 period (Al-Awadhi, 2005) (average 28 g m<sup>-2</sup>-month<sup>-1</sup>, range 3-58 g m<sup>-2</sup> month<sup>-1</sup>) to samples collected in Kuwait City during the 2011/12 period (Al-Awadhi and AlShuaibi, 2013) (average

53 g m<sup>-2</sup> month<sup>-1</sup>, range 2-320 g m<sup>-2</sup> month<sup>-1</sup>) and samples collected during the 1979/80 campaign in the N-W Gulf area of Kuwait (Khalaf and Al-Hashash, 1983) (average 191 g m<sup>-2</sup> month<sup>-1</sup>, range 10-1003 g m<sup>-2</sup> month<sup>-1</sup>).

#### **5.3 AERONET** and visibility measurements

5 The Aerosol Optical Depth (AOD) is one of the best observed aerosol characteristics. It defines the aerosol radiaitive effect and reflects the abundance of aerosols in the atmosphere. A CIMEL Robotic Sun Photometer is installed on the rooftop of the CMOR building on the campus of the KAUST and operated by our group since 2012, as a part of the NASA- AERONET, providing aerosol optical depth (AOD) and aerosol retrieved characteristics (https://aeronet.gsfc.nasa.gov/). Figure 6a compares the monthly 10 averaged AOD at 500 nm with the dust deposition rate for 2015. In a general sense the AOD and the deposition rates show comparable trends, both with maxima in spring and larger maxima in fall. However, the AOD reaches a first maximum in April, being one month later than that of the deposition rate. Also, the larger second AOD maximum occurred in August while the maximum deposition rate is broadly distributed over a three-month period, from August to October. The photometer measures light 15 attenuation by all aerosols along a column in the atmosphere, while deposition rate depends on dust at ground level only, the latter generally containing a relatively coarser dust fraction. The low-level dust particles are partly predominantly from local dust sources while the higher altitude dust could be transported from distal sources and chemically transformed, i.e., aged. As was pointed out by Yu et al. (2013) the differences between the deposition and AOD time series can in part be attributed to 20 modifications of the natural dust aerosol by anthropogenic activities, including petrochemical and other large industries along the Red Sea coast, as well as by entrainment of construction and road dust. The linear correlation coefficient between the monthly deposition rates and the monthly averaged AOD of 0.40 suggests a causal interrelationship between these two quantities.

Furthermore, a comparison between the deposition samples and the visibility is made with measurements taken in 2015 at the Jeddah airport meteorological station, approximately 70 km to the south of KAUST. Visibility is expressed as the frequency of dust events with reported weather codes 06-09, or 30–35, grouped as dusty or non-dusty days, for each month (Notaro et al., 2013; Anisimov et al., 2017), expressed as percentages. The bimodal monthly distributions seen with the deposition rates and AERONET monitoring are also mirrored by the visibility measurements collected at Jeddah (Fig. 6b). The linear correlation coefficient between the monthly deposition rates and monthly averaged visibility measurements is 0.48, clearly suggesting a causal relationship between the two variables. It has been suggested that visibility and visibility frequency be used as a metric of dust emission flux and near surface dust concentrations (Cowie et al., 2014; Anisimov et al., 2017; Yu et al., 2013; Norris et al., 2014; Shao and Dong, 2006; Notaro et al., 2013).

#### 5.4 Particle size distributions (PSD)

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Dust deposition rates depend on the meteorological conditions, and dust properties such as particle size distribution, their vertical distribution, and abundance.

Summary plots of results from SEM based individual particle analysis for each month of 2015, expressed by number are presented in the Supplement C to this paper. From these particle size and shape measurements, equivalent shape-dependent volumes for the particles were calculated, the summary

plots of which are shown in Appendix A. The volume of each particle is calculated from the measured maximum and minimum diameters, and assuming a prolate spheroid. Also, assuming a similar average density of e.g.  $2.65 \, \mathrm{g} \, \mathrm{cm}^{-3}$  for all minerals in the deposition samples, results in similar volume and mass distributions patterns. This was confirmed by XRD measurements and the abundance of quartz ( $2.65 \, \mathrm{g} \, \mathrm{cm}^{-1}$ ), feldspar ( $^{\sim} 2.65 \, \mathrm{g} \, \mathrm{cm}^{-1}$ ), micas ( $^{\sim} 2.83 \, \mathrm{g} \, \mathrm{cm}^{-1}$ ), and clays ( $^{\sim} 2.7 - 2.8 \, \mathrm{g} \, \mathrm{cm}^{-1}$ ) found in the deposition samples. The volume distributions were applied to assess the mass percentages and deposition rates of each size bin, e.g. the mass percentages and mass deposition rates of particles in bins less <  $10 \, \mu \mathrm{m}$  in average diameter, and similarly less than <  $2.5 \, \mu \mathrm{m}$  in average diameter, together with their uncertainties (Table 4). The contribution of particles <10 um to the total measured mass varies between about 4 and  $17 \, \%$  with an average of  $8.6 \, \%$  for the twelve months. Particles less than  $2.5 \, \mu \mathrm{m}$  range from about  $0.6 \, \mathrm{to}$  4 %, with an average of  $1.2 \, \%$  for the twelve month period. From these percentages and the total deposition rates, average deposition rates of  $1.2 \pm 0.7 \, \mathrm{g} \, \mathrm{m}^{-2}$  month<sup>-1</sup> for <  $10 \, \mu \mathrm{m}$  and  $0.1 \pm 0.1 \, \mathrm{g} \, \mathrm{m}^{-2}$  month<sup>-1</sup> for <  $2.5 \, \mu \mathrm{m}$  are estimated.

The average size distribution of the twelve deposition samples (Fig. 8a) is compared to that of the thirteen surface soils (Fig. 8b) from potential dust source regions along the Red Sea coastal plain (Prakash et al., 2016). The deposition samples with an average diameter of 0.9  $\mu$ m are much finer than the 3.9  $\mu$ m average diameter of the <38  $\mu$ m sieved soils. In addition the Frisbee sampler is biased towards the sampling of the coarser particles, as previously documented (Bergametti and Forêt, 2014; Goossens, 2005).

#### 5.5 Mineral analysis by XRD

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XRD analysis of the 27 samples (Fig. 7) show variable amounts of quartz (6–38 %, avg. 22 %) and feldspars (plagioclase, K-feldspar) (5-34 %, avg. 20 %), clays (10-18 %, avg. 13 %), micas (6-31 %, avg. 13 %), halite (1-53 %, avg. 7 %) with lesser amounts of gypsum (1-8 %, avg. 4 %), calcite (0-8 %, avg. 2 %), dolomite (0-7 %, avg. 3 %), hematite (0-8 %, avg. 3 %), and amphibole (and pyroxene) (0-4%, avg. 1 %).

From the XRD, four broad mineral assemblages are be distinguished, the first and major assemblage is comprised of feldspars, clays and micas as well as hematite and gypsum, the second group is of quartz, the third of halite, and the fourth of calcite.

There is an increase in the halite concentrations at sites DT1-DT3, from about 2 % (DT1) in December 2014 to about 53 % (DT2) in July 2015 (Fig. 7). From August onwards there is an abrupt decrease in halite content to less than 5 %, except for samples collected at the DT2 (CMOR, quay-side) site alongside the ocean. There was a simultaneous increase in the proportion of quartz to a maximum of 38 % in April (DT3), and decreasing to less than 25 % at all sites after July, 2015. The silicate mineral group decreased systematically from about 72 % (DT1) in December 2014 to about 25 % (DT2) in July. Except for two samples from the DT3 site collected in September and October 2015, the dominant minerals after July, 2015 included the silicate assemblage, with concentrations of up to 80 %. The variation in the proportions of the four mineral assemblages, especially the halite, is ascribed to seasonal fluctuations in wind, humidity and precipitation, as well as the proximity of the sea to the sampling sites.

#### **5.6** Chemistry (XRF and IC)

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As expected, the chemically analyzed deposition samples contain major amounts of  $SiO_2$  (Appendix A, Fig. 9a, b), varying between 12–53 % (avg. 31 %) in the sample subset, occurring as quartz, and together with  $Al_2O_3$ , (avg. 4 %) and CaO, (avg. 2.3 %) in plagioclase, and  $K_2O$  (avg. 0.6 %) in potassium feldspars.  $SiO_2$  together with  $Al_2O_3$ ,  $Fe_2O_3$ ,  $TiO_2$ , MnO, MgO, and some  $K_2O$  are also contained in the clays, micas and amphiboles, previously identified in these samples by optical microscopy and XRD. Lesser amounts of CaO are contained in gypsum and calcite, and together with MgO, in dolomite. The iron expressed here as  $Fe_2O_3$  can be contained in hematite ( $Fe_2O_3$ ), goethite FeO(OH) or in clay minerals such as illite, each with different solubility. It has been suggested that large fractions of iron in soils and dusts are contained as amorphous colloidal coatings on quartz and feldspars (Engelbrecht et al., 2016).

The water-soluble cations (Appendix A, Fig. 10a, b) account for 1-19 % and the anions for 1-30 % of the total mass, respectively. These account for variable amounts of halite (1-32 %), and gypsum (1-9 %), with lesser amounts of other chlorides and carbonates. Of importance as dust borne nutrients likely to be deposited in the Red Sea, are the low concentrations of both water soluble  $NO_3^-$  (avg. 0.8 %), and water soluble  $PO_4^{3-}$  (avg. 0.2 %) compared to the total  $P_2O_5$  (avg. 0.3 %) in the dust deposits. The phosphorus is contained in the largely insoluble mineral apatite (francolite), found in the sedimentary rocks underlying large parts of the Arabian Peninsula (Notholt et al., 2005).

The sum of chemical species, including elements expressed as oxides, and ion concentrations, vary from 35–78 %, with an average of 56 % of the measured chemical mass. The shortfall from 100 % is attributed in part to components not analyzed for, including  $H_2O$ , OH, carbon ( $CO_3^{2-}$ , organic carbon, elemental carbon) and artifacts of debris deposited onto the samplers.

The chemical abundances were recalculated as normative minerals (Fig. 11a, b), comparable in composition to those identified by XRD (Fig. 7) and optical microscopy. The relative normative mineral abundances (Fig. 11b) show variable amounts of quartz (avg. 52.4 %) feldspar (avg. 3.9 %), kaolinite (2.6 %), calcite (8.8 %) dolomite (0.2 %), hematite (8.0 %), as well as the evaporate minerals gypsum (12.1 %), halite (12.1 %), sylvite (0.2 %), and bischofite (0.2 %). There is also, as shown by XRD, an increase in halite content from about 7.8 % in January to about 25.9 % in July, followed by a sharp drop to about 4.6 % in August, with greater abundances at the CMOR quayside site in September (51.0 %) and October (31.6 %), ascribed to sea spray from stormy conditions during those two months.

Elemental mass ratios of the Frisbee deposition samples are compared to the <38 µm sieved soil samples from the Arabian Red Sea coastal plain (Prakash et al., 2016), and total suspended particulate (TSP) samples collected at other sites in the Middle East (Engelbrecht et al., 2009a) are compared in Table 5. The average Si/Al ratio of 6.86 of the Frisbee deposition samplers is intermediate to the 13.60 of the Arabian Red Sea coastal soils and the approximately unity of the Middle East samples. The Fe/Al ratios of the sample sets show similar relationships as the Si/Al ratios, being intermediate to the Red Sea coastal soils and four of the five other Middle East countries, excluding UAE to which it is similar. The difference is ascribed to the greater abundance of the minerals such as quartz in the coarser sieved soil samples, and less thereof in the finer TSP fractions. The Ca/Al ratio of 2.17 is similar to those of TSP samples from samples of Qatar (2.07) and UAE (2.16), ascribed to the regional carbonate-bearing soils in all three countries. The average Ti/Al, Mg/Al and K/Al ratios of the Frisbee deposition samples are substantially lower than those of the Red Sea coastal soils, which may be related to mineralogical

differences in the dust source regions. Differences can also be ascribed to larger percentages of Albearing minerals such as clays in the deposition samples from this study.

#### 6. Summary and conclusions

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This study provides new has as its goal the provision of mineralogical, physical and chemical information on deposition samples collected at the KAUST campus during 2015, as well as an assessment of the seasonal variability of the regional dust deposition rates onto Saudi Arabian coastal plain, and is meant to be used for validating dust mass balance in the meteorological models with the dust component.

Inverted Frisbee samplers with foam inserts are found to be robust, easy to use, and provided comparable results, for the collection of wet and dry deposits. Once a month the samples are retrieved by flushing the deposits into plastic flasks followed by freeze-drying of the slurry and recovery of all suspended particles and dissolved salts. The average deposition rate at KAUST for 2015 was 14 g m<sup>-2</sup> varying from 4 g m<sup>-2</sup> in December, to 20 g m<sup>-2</sup> in March, 5 g m<sup>-2</sup> in July, 28 g m<sup>-2</sup> in September and October, and down to 11 g m<sup>-2</sup> the following December. The changes are ascribed to seasonally variable meteorological conditions, including high humidity prevailing along the Arabian Red Sea coastal plain during the late summer and autumn months. The particle size distributions provide an assessment of <10  $\mu$ m and <2.5  $\mu$ m dust deposition rates, the former varying 0.6-2.8 g m<sup>-2</sup> and the latter 0.06-0.29 g m<sup>-2</sup>.per month We suggest these deposition rates as proxies for those of PM<sub>10</sub> (coarse) and PM<sub>2.5</sub> (fine), respectively.

Chemical analysis, confirmed by XRD, point to a consistent silicate mineral fractions for the deposition samples, at all sampling sites for the entire sampling period. The Si/Al, Fe/Al, and Ca/Al ratios of the deposition samples fall within the range of the soil samples previously collected along the Arabian Red Sea coastal plain as well as the TSP size fractions collected at several sites in the Middle East. It is therefore not feasible to explicitly relate the deposition samples to the coastal soils from chemical and mineralogical results on their own. It is proposed that the dust deposits along the Red Sea coast are a mixture of dust emissions from local soils, and soils imported from distal dust sources.

For 2015, there are marked similarities between monthly distribution patterns of the deposition samples and AOD measured at KAUST, as well as visibility measurements from Jeddah airport, 70 km to the south. This shows that both the AOD and visibility measurements mirror fluctuations in dust deposition, although it may not be justified to calculate quantitative interrelationships without further research.

30 Except for the variable halite fractions and local construction dust, there are small variations in the mineralogical content of the dust samples collected on the KAUST campus. To better represent model the dust being deposited in Red Sea, and coastal plain, the sampling campaign should be extended to sites beyond the KAUST campus. Such a sampling site was recently set up on an island off the coast from KAUST. Inclusion of particle size with mineralogical and chemical measurements provide more effective data for the modeling community.

The deposition samplers collect all particle sizes, however bin aerosol models usually consider only  $PM_{10}$ . The estimated  $PM_{10}$  deposition rates are lower than the total particulate deposition rates we observed. However, the size distribution of deposited particles shown in Figure 8a and Appendix A could be used to assess the contribution of  $PM_{10}$  in deposited mass, and reconcile models with observations.

Another would be to expand the calculated particle size range in the models to cover TSP. However, this could be computationally expansive.

#### 7. Data availability

The gravimetric, mineralogical and chemical data from this study are available upon request from Georgiy Stenchikov (Georgiy.Stenchikov@kaust.edu.sa).

Author Contributions. Johann Engelbrecht was responsible for the sample analysis and data compilation; Georgiy Stenchikov formulated the problem, designed the research project, and supported experimental activities; Jish Prakash collected, and conducted the freeze-drying of the samples, and performed part of the XRD analysis. Traci Lersch performed the SEM based individual particle analysis, Anatolii Anisimov assembled the meteorological and visibility data; Illia Shevchenko assembled the AERONET optical data; Engelbrecht, Stenchikov, Jish Prakash, Anisimov, and Shevchenko compiled different parts of the manuscript.

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#### **FIGURES**

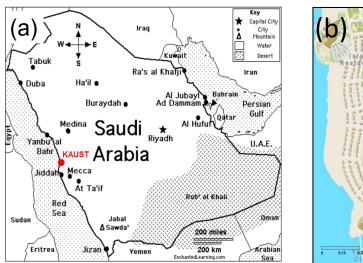




Figure 1. Position Locality-of (a) the King Abdullah University of Science and Technology (KAUST) campus
on the Arabian Peninsula (red marker), north of the coastal city of Jeddah, along the Saudi Arabian Red
Sea coast and (b) the Frisbee deposition sites (DT1-DT4) on the KAUST campus.

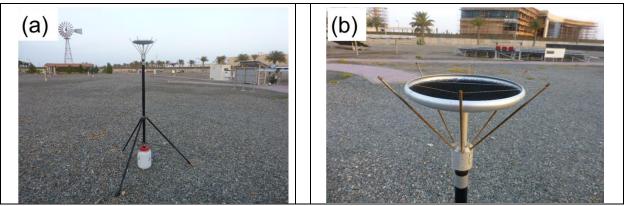


Figure 2. Inverted Frisbee type deposition sampler (a) on tripod and white plastic drainage bottle, (b) showing the foam insert in the collection dish to help retain the deposited dust particles, as well as the spikes with nylon thread to prevent birds from readily perching on the dish.

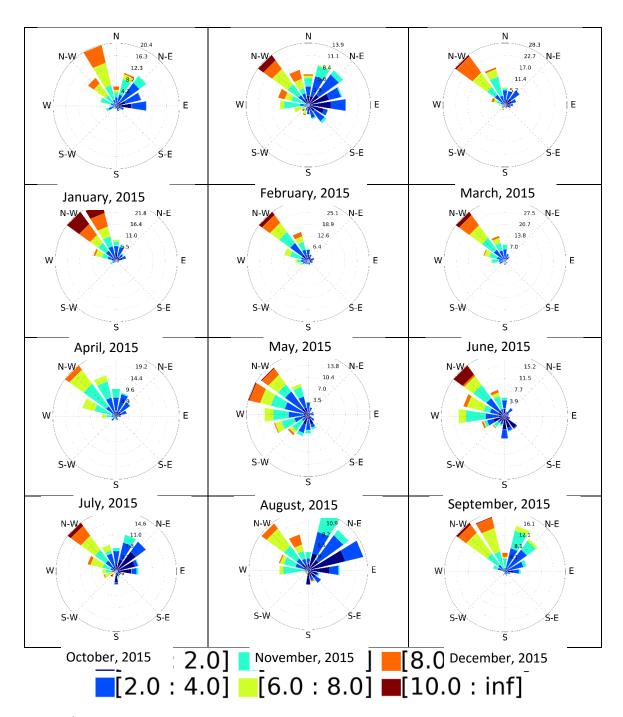


Figure 3. Wind\_(m s<sup>-1</sup>) roses <del>show prevailing north westerly Shamal winds at KAUST</del> for each month of 2015. <del>year with north-easterly winds (windspeed in m s<sup>-1</sup>) in the winter months of October through February.</del>

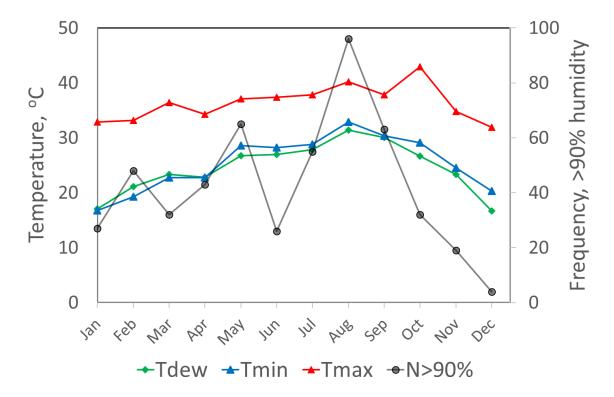


Figure 4. Monthly averaged minimum (———) and maximum (————) ambient temperatures as well as dewpoint (————) variations for KAUST during 2015. Also shown for each month is the frequency of hourly humidity measurements exceeding 90% (————).

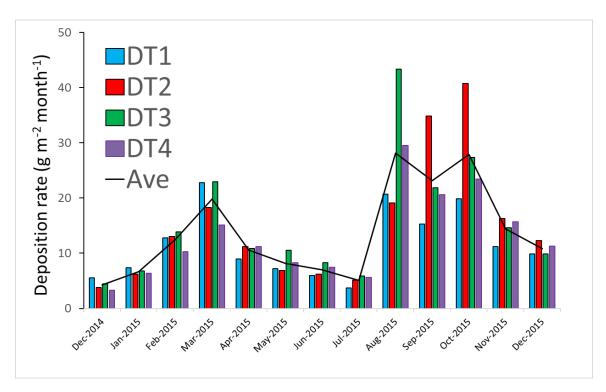


Figure 5. Monthly deposition rates (g m<sup>-2</sup>) from Frisbee samplers (DT1-DT4) at the KAUST campus. Also shown are the monthly averages for the four samplers.

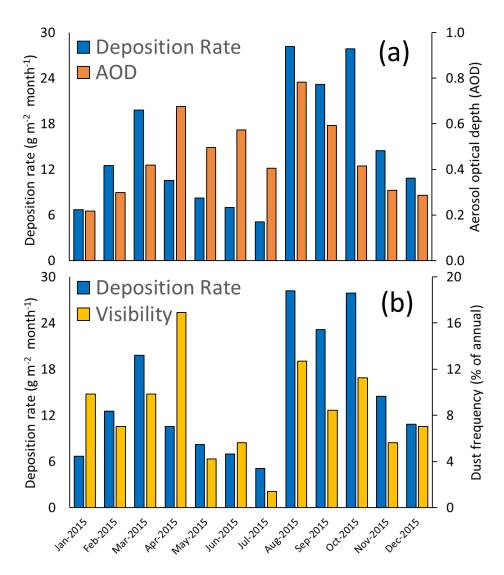


Figure 6. Average monthly deposition rates for all four samplers (DT1-DT4) on the KAUST campus, together with (a) monthly averaged AOD measurements from the KAUST AERONET site, and (b) monthly averaged visibility measurements collected from the Jeddah airport, for 2015.

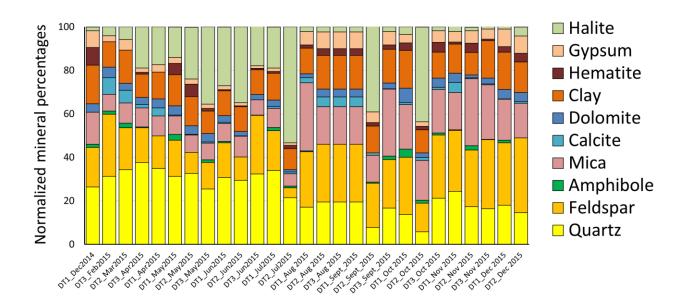


Figure 7. Semi-quantitative XRD mineral analyses of monthly Frisbee samples collected at the three sites DT1–DT3, for the period December, 2014 to December2015.

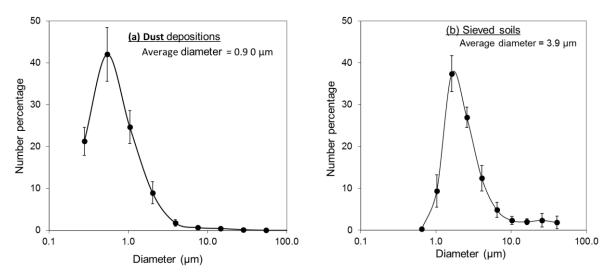


Figure 8. Average particle size distributions and standard deviations of (a) twelve deposition samples collected by Frisbee samplers on KAUST campus, and of (b) thirteen <38  $\mu$ m sieved soil samples from a previous study (Prakash et al., 2016), both measured by SEM.

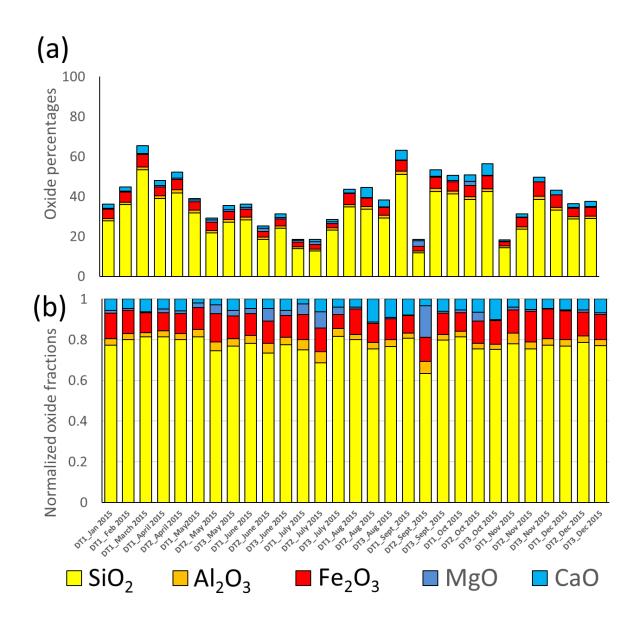


Figure 9 (a). Deposition sample elemental compositions, expressed as oxides and (b) fractions normalized to unity.

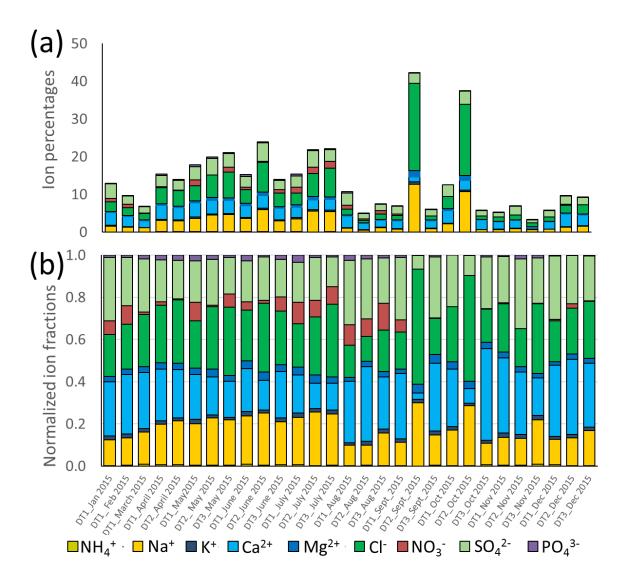


Figure 10 (a). Ion concentrations and (b) fractions totaled to unity.

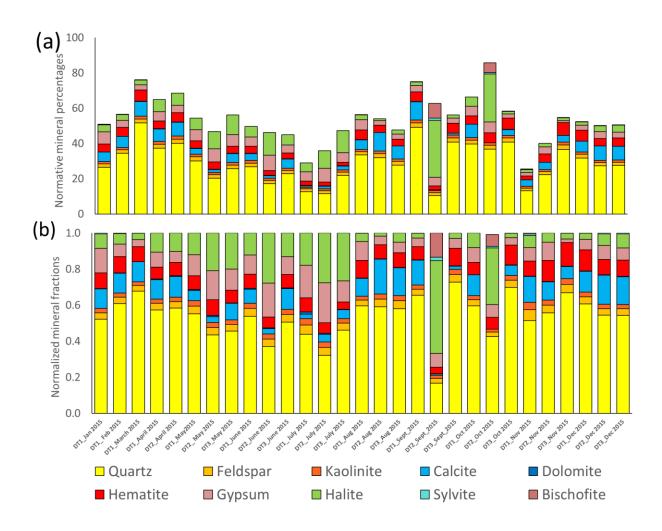


Figure 11 (a). Chemical abundances combined as normative minerals, (b) normalized to 100%.

# **TABLES**

Table 1. Locality of deposition samplers at six sites on the campus of KAUST.

|     | Site      | Latitude      | Longitude     | Elev.<br>m.a.s.l. | Start | End   |
|-----|-----------|---------------|---------------|-------------------|-------|-------|
| DT1 | NEO 1     | 22°18'16.12"N | 39°06'28.46"E | 1                 | Dec14 | Mar15 |
|     | Res G3705 | 22°18'59.06"N | 39°06'21.32"E | 12                | Apr15 | Dec15 |
| DT2 | NEO 2     | 22°18'16.84"N | 39° 6'29.33"E | 1                 | Dec14 | Mar15 |
|     | CMOR      | 22°18'16.60"N | 39° 6′7.91″E  | 1                 | Apr15 | Dec15 |
| DT3 | NEO 3     | 22°18'17.31"N | 39° 6'30.51"E | 1                 | Dec14 | Dec15 |
| DT4 | NEO 4     | 22°18'18.10"N | 39° 6'31.52"E | 1                 | Dec14 | Dec15 |

Table 2. Monthly averaged temperatures, humidity measurements, and calculated dewpoints at KAUST during 2015.

| Month |      | Temp | erature | 9     |   |      | Hu   | midity |       | Dewpoint |
|-------|------|------|---------|-------|---|------|------|--------|-------|----------|
|       | Avg. | Min. | Max.    | Range | _ | Avg. | Min. | Max.   | N>90% | Avg.     |
|       | °C   | °C   | °C      | °C    |   | %    | %    | %      | Count | °C       |
| Jan   | 25   | 17   | 33      | 16    |   | 61   | 10   | 99     | 27    | 17       |
| Feb   | 27   | 19   | 33      | 14    |   | 74   | 15   | 99     | 48    | 21       |
| Mar   | 28   | 23   | 36      | 14    |   | 76   | 29   | 99     | 32    | 23       |
| Apr   | 28   | 23   | 34      | 12    |   | 74   | 23   | 99     | 43    | 23       |
| May   | 32   | 29   | 37      | 9     |   | 77   | 21   | 99     | 65    | 27       |
| Jun   | 32   | 28   | 37      | 9     |   | 76   | 22   | 99     | 26    | 27       |
| Jul   | 33   | 29   | 38      | 9     |   | 75   | 26   | 99     | 55    | 28       |
| Aug   | 35   | 33   | 40      | 7     |   | 82   | 36   | 99     | 96    | 31       |
| Sep   | 34   | 30   | 38      | 8     |   | 80   | 26   | 99     | 63    | 30       |
| Oct   | 33   | 29   | 43      | 14    |   | 72   | 9    | 96     | 32    | 27       |
| Nov   | 30   | 25   | 35      | 10    |   | 69   | 25   | 99     | 19    | 23       |
| Dec   | 27   | 20   | 32      | 12    |   | 57   | 15   | 94     | 4     | 17       |

Table 3. Dust deposition measurements from the Middle East and other Global dust regions.

|     | Study                          | Locality             | Sampler type                        | Sampling period       | Average deposition rate (g m <sup>-2</sup> month <sup>-1</sup> ) | Range deposition rate (g m <sup>-2</sup> month <sup>-1</sup> ) |
|-----|--------------------------------|----------------------|-------------------------------------|-----------------------|------------------------------------------------------------------|----------------------------------------------------------------|
| (a) | This study (2017)              | Saudi Arabia, KAUST  | Frisbee with foam insert            | Dec 2014 - Dec 2015   | 14                                                               | 4 - 28                                                         |
| (b) | Modaihsh and Mahjoub (2013)    | Saudi Arabia, Riyadh | Dish with marbles                   | Jan - Mar ?           | 42                                                               | 20 - 140                                                       |
| (c) | Khalaf and Al-Hashash (1983)   | Kuwait, N-W Gulf     | Polyethelene cylinders with water   | Apr 1979 - Mar 1980   | 191                                                              | 10 - 1003                                                      |
| (d) | Al-Awadhi (2005)               | Kuwait, N-E Bay      | PVC bucket with marbles             | May 2002 - Apr 2003   | 28                                                               | 3 - 58                                                         |
| (e) | Al-Awadhi and AlShuaibi (2013) | Kuwait, City         | PVC bucket with marbles             | Mar 2011 - Feb 2012   | 53                                                               | 2 - 320                                                        |
| (f) | Offer and Goossens (2001)      | Israel, Negev        | Marble collectors                   | 1988 - 1997           | 17                                                               | 10 - 25                                                        |
| (g) | Goossens and Rajot (2008)      | Niger, Banizoumbou   | Frisbee with marbles, original data | 8 periods in 2005     | 13                                                               | 6 - 21                                                         |
| (h) | Smith and Twiss (1965)         | USA, Kansas          | Cylindrical rain gauge with screens | June 1963 - June 1964 | 6                                                                | 3 - 14                                                         |

Table 4. Monthly measured deposition rates, and assessments of < 10  $\mu$ m and < 2.5  $\mu$ m deposition rates from SEM based particle size measurements.

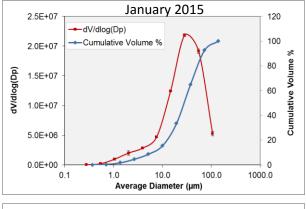
| Sample #     | Month (2015) |                                       | ſ             | Deposition Rate                       |               |                                       |
|--------------|--------------|---------------------------------------|---------------|---------------------------------------|---------------|---------------------------------------|
|              |              | Total                                 | < 1           | l0 μm                                 | < 2           | .5 μm                                 |
|              |              | g m <sup>-2</sup> month <sup>-1</sup> | % of Total    | g m <sup>-2</sup> month <sup>-1</sup> | % of Total    | g m <sup>-2</sup> month <sup>-1</sup> |
| DT3.1_012015 | January      | 7.34                                  | 16.5 ± 4.3    | 1.2 ± 0.3                             | 4.0 ± 1.6     | 0.29 ± 0.12                           |
| DT3.1_022015 | February     | 12.83                                 | 7.0 ± 3.3     | $0.9 \pm 0.4$                         | $0.9 \pm 0.4$ | $0.12 \pm 0.05$                       |
| DT3.4_032015 | March        | 15.11                                 | 12.0 ± 5.8    | $1.8 \pm 0.9$                         | 1.6 ± 0.6     | $0.24 \pm 0.09$                       |
| DT3.4_042015 | April        | 11.22                                 | 8.7 ± 2.8     | $1.0 \pm 0.3$                         | $0.8 \pm 0.3$ | $0.09 \pm 0.03$                       |
| DT3.3_052015 | May          | 10.51                                 | 7.3 ± 2.1     | $0.8 \pm 0.2$                         | $0.8 \pm 0.3$ | $0.08 \pm 0.03$                       |
| DT3.3_062015 | June         | 8.28                                  | 9.0 ± 2.8     | $0.7 \pm 0.2$                         | $0.9 \pm 0.3$ | $0.07 \pm 0.02$                       |
| DT3.3_072015 | July         | 5.86                                  | 9.7 ± 6.3     | $0.6 \pm 0.4$                         | 1.1 ± 0.7     | $0.06 \pm 0.04$                       |
| DT3.3_082015 | August       | 43.39                                 | $6.4 \pm 3.3$ | $2.8 \pm 1.4$                         | $0.6 \pm 0.4$ | 0.26 ± 0.17                           |
| DT3.3_092015 | September    | 21.90                                 | $4.3 \pm 7.1$ | $0.9 \pm 1.6$                         | 0.6 ± 1.0     | $0.13 \pm 0.22$                       |
| DT3.3_102015 | October      | 27.39                                 | 9.2 ± 7.3     | $2.5 \pm 2.0$                         | $0.8 \pm 0.6$ | $0.22 \pm 0.16$                       |
| DT3.3_112015 | November     | 14.59                                 | 6.1 ± 1.4     | $0.9 \pm 0.2$                         | $0.7 \pm 0.2$ | $0.10 \pm 0.03$                       |
| DT3.3_122015 | December     | 9.91                                  | 7.3 ± 2.5     | 0.7 ± 0.2                             | 1.1 ± 0.2     | 0.11 ± 0.02                           |

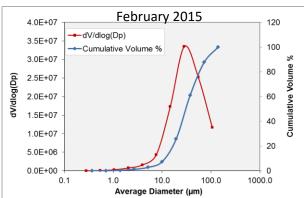
Table 5. Elemental mass ratios for the deposition samples from this study, compared to those of soils from the Red Sea coastal plain (Prakash et al., 2016) and TSP samples from other countries of the Middle East (Engelbrecht et al., 2009a). The TSP filter samples were collected by low-volume aerosol samplers without size selective inlets, for 24-hr sampling periods.

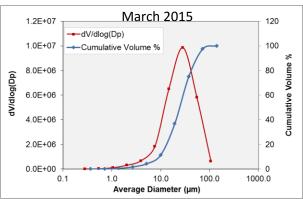
|             |              | Si/Al | Ti/Al | Fe/Al | Mg/Al | Ca/Al | K/Al |
|-------------|--------------|-------|-------|-------|-------|-------|------|
| Frisbee     | Deposition   | 6.86  | 0.14  | 1.47  | 0.11  | 2.17  | 0.34 |
| Saudi Soils | Sieved <38μm | 13.60 | 0.44  | 2.52  | 0.65  | 0.36  | 0.43 |
| Djibouti    | TSP          | 0.92  | 2.19  | 1.12  | 0.88  | 0.74  | 1.14 |
| Afghanistan | TSP          | 1.05  | 1.25  | 1.00  | 0.94  | 0.69  | 1.96 |
| Qatar       | TSP          | 1.02  | 0.24  | 0.98  | 1.40  | 2.07  | 0.93 |
| UAE         | TSP          | 1.29  | 0.28  | 1.52  | 2.85  | 2.16  | 1.02 |
| Iraq        | TSP          | 1.03  | 0.72  | 0.99  | 1.11  | 1.31  | 1.04 |
| Kuwait      | TSP          | 1.07  | 0.65  | 0.99  | 1.25  | 1.23  | 0.94 |

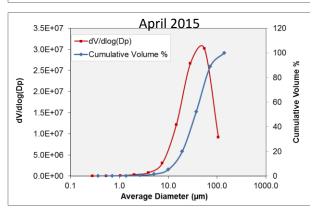
Appendix A

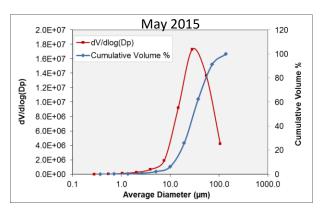
SEM based particle volume distribution curves for 12 months of 2015

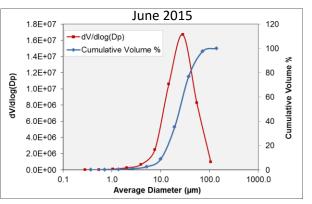




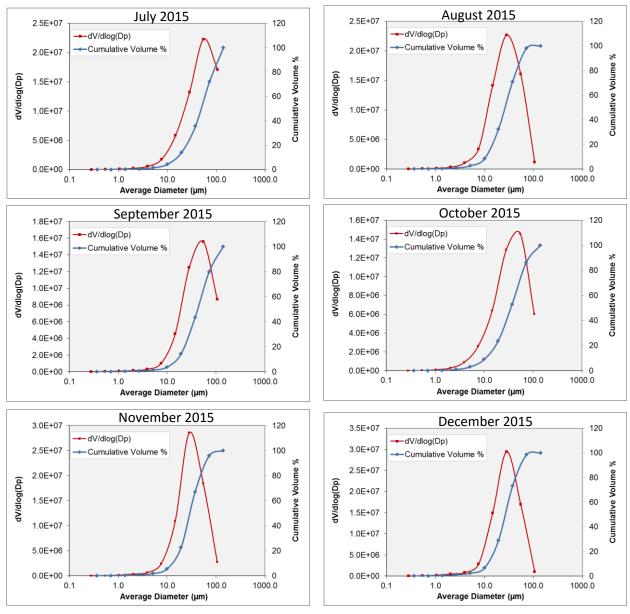








# Appendix A SEM based particle volume distribution curves for 12 months of 2015



Appendix B

| Sample                         | DT1_      | Jan | 2015  | DT1_   | Feb | 2015  | DT1_N  | 1ar | ch 2015 | DT1_/  | \pr | il 2015 | DT2_/  | \pr | il 2015 | DT1_   | Ma | y2015 | DT2_   | Ma | y 2015 |
|--------------------------------|-----------|-----|-------|--------|-----|-------|--------|-----|---------|--------|-----|---------|--------|-----|---------|--------|----|-------|--------|----|--------|
| Major and minor eleme          | nts as ox | ide | s (%) |        |     |       |        |     |         |        |     |         |        |     |         |        |    |       |        |    |        |
| SiO <sub>2</sub>               | 27.890    | ±   | 0.050 | 35.886 | ±   | 0.065 | 53.301 | ±   | 0.089   | 38.965 | ±   | 0.063   | 41.802 | ±   | 0.073   | 31.729 | ±  | 0.055 | 21.772 | ±  | 0.039  |
| TiO <sub>2</sub>               | 0.466     | ±   | 0.001 | 0.530  | ±   | 0.001 | 0.679  | ±   | 0.002   | 0.464  | ±   | 0.002   | 0.599  | ±   | 0.002   | 0.486  | ±  | 0.002 | 0.451  | ±  | 0.001  |
| Al <sub>2</sub> O <sub>3</sub> | 3.505     | ±   | 0.035 | 4.464  | ±   | 0.050 | 5.421  | ±   | 0.097   | 4.257  | ±   | 0.115   | 4.824  | ±   | 0.081   | 3.817  | ±  | 0.079 | 3.033  | ±  | 0.026  |
| Fe <sub>2</sub> O <sub>3</sub> | 4.480     | ±   | 0.007 | 5.119  | ±   | 0.008 | 6.312  | ±   | 0.013   | 4.309  | ±   | 0.013   | 5.091  | ±   | 0.011   | 4.150  | ±  | 0.010 | 4.104  | ±  | 0.006  |
| MnO                            | 0.080     | ±   | 0.001 | 0.082  | ±   | 0.002 | 0.109  | ±   | 0.004   | 0.075  | ±   | 0.005   | 0.091  | ±   | 0.003   | 0.077  | ±  | 0.003 | 0.066  | ±  | 0.001  |
| CaO*                           | 1.989     | ±   | 0.015 | 2.097  | ±   | 0.013 | 4.157  | ±   | 0.016   | 2.369  | ±   | 0.018   | 3.066  | ±   | 0.017   | 0.753  | ±  | 0.014 | 0.847  | ±  | 0.013  |
| K₂O*                           | 0.484     | ±   | 0.005 | 0.604  | ±   | 0.004 | 0.864  | ±   | 0.003   | 0.705  | ±   | 0.006   | 0.798  | ±   | 0.005   | 0.542  | ±  | 0.008 | 0.432  | ±  | 0.007  |
| P <sub>2</sub> O <sub>5</sub>  | 0.417     | ±   | 0.001 | 0.150  | ±   | 0.001 | 0.010  | ±   | 0.003   | 0.332  | ±   | 0.004   | 0.306  | ±   | 0.002   | 0.474  | ±  | 0.003 | 0.429  | ±  | 0.001  |
| Total (oxides)                 | 39.311    |     |       | 48.932 |     |       | 70.852 |     |         | 51.475 |     |         | 56.579 |     |         | 42.028 |    |       | 31.134 |    |        |
| Trace elements (ppm)           |           |     |       |        |     |       |        |     |         |        |     |         |        |     |         |        |    |       |        |    |        |
| V                              | 107       | ±   | 1     | 76     | ±   | 1     | 85     | ±   | 3       | 106    | ±   | 4       | 93     | ±   | 3       | 116    | ±  | 3     | 99     | ±  | 1      |
| Cr                             | 92        | ±   | 3     | 115    | ±   | 5     | 113    | ±   | 12      | 101    | ±   | 15      | 108    | ±   | 10      | 113    | ±  | 10    | 120    | ±  | 2      |
| Ni                             | 78        | ±   | 2     | 71     | ±   | 3     | 84     | ±   | 6       | 68     | ±   | 8       | 71     | ±   | 5       | 77     | ±  | 5     | 57     | ±  | 1      |
| Cu                             | 81        | ±   | 3     | 57     | ±   | 4     | 134    | ±   | 10      | 125    | ±   | 13      | 99     | ±   | 9       | 206    | ±  | 9     | 64     | ±  | 2      |
| Zn                             | 223       | ±   | 5     | 247    | ±   | 8     | 293    | ±   | 18      | 287    | ±   | 23      | 258    | ±   | 15      | 467    | ±  | 16    | 219    | ±  | 4      |
| As                             | 0         | ±   | 3     | 0      | ±   | 5     | 6      | ±   | 12      | 0      | ±   | 15      | 0      | ±   | 10      | 0      | ±  | 10    | 0      | ±  | 2      |
| Br                             | 29        | ±   | 3     | 26     | ±   | 4     | 42     | ±   | 10      | 59     | ±   | 13      | 45     | ±   | 9       | 34     | ±  | 9     | 76     | ±  | 2      |
| Rb                             | 28        | ±   | 1     | 24     | ±   | 1     | 16     | ±   | 3       | 28     | ±   | 4       | 34     | ±   | 3       | 30     | ±  | 3     | 27     | ±  | 1      |
| Sr                             | 333       | ±   | 3     | 392    | ±   | 5     | 514    | ±   | 11      | 386    | ±   | 13      | 422    | ±   | 9       | 303    | ±  | 9     | 341    | ±  | 2      |
| Υ                              | 1071      | ±   | 4     | 36     | ±   | 4     | 26     | ±   | 10      | 125    | ±   | 13      | 46     | ±   | 9       | 36     | ±  | 9     | 24     | ±  | 2      |
| Zr                             | 103       | ±   | 4     | 98     | ±   | 6     | 84     | ±   | 14      | 57     | ±   | 17      | 139    | ±   | 12      | 133    | ±  | 12    | 134    | ±  | 3      |
| Мо                             | 0         | ±   | 5     | 0      | ±   | 7     | 12     | ±   | 17      | 4      | ±   | 21      | 8      | ±   | 14      | 0      | ±  | 14    | 0      | ±  | 3      |
| Pb                             | 15        | ±   | 5     | 28     | ±   | 8     | 32     | ±   | 18      | 66     | ±   | 23      | 28     | ±   | 15      | 19     | ±  | 15    | 28     | ±  | 3      |
| Water soluble ions (%)         |           |     |       |        |     |       |        |     |         |        |     |         |        |     |         |        |    |       |        |    |        |
| NH <sub>4</sub> <sup>+</sup>   | 0.027     | ±   | 0.003 | 0.033  | ±   | 0.004 | 0.055  | ±   | 0.006   | 0.105  | ±   | 0.012   | 0.095  | ±   | 0.011   | 0.076  | ±  | 0.009 | 0.079  | ±  | 0.009  |
| Na <sup>+</sup>                | 1.585     | ±   | 0.012 | 1.267  | ±   | 0.010 | 1.053  | ±   | 0.009   | 2.952  | ±   | 0.021   | 2.897  | ±   | 0.021   | 3.519  | ±  | 0.025 | 4.473  | ±  | 0.031  |
| K <sup>+</sup>                 | 0.230     | ±   | 0.009 | 0.170  | ±   | 0.006 | 0.114  | ±   | 0.004   | 0.262  | ±   | 0.010   | 0.207  | ±   | 0.008   | 0.371  | ±  | 0.014 | 0.296  | ±  | 0.011  |
| Mg <sup>2+</sup>               | 0.338     | ±   | 0.005 | 0.265  | ±   | 0.004 | 0.188  | ±   | 0.003   | 0.488  | ±   | 0.007   | 0.400  | ±   | 0.005   | 0.558  | ±  | 0.008 | 0.744  | ±  | 0.010  |
| Ca <sup>2+</sup>               | 3.311     | ±   | 0.028 | 2.718  | ±   | 0.023 | 1.813  | ±   | 0.015   | 3.745  | ±   | 0.032   | 3.220  | ±   | 0.027   | 3.780  | ±  | 0.032 | 3.560  | ±  | 0.030  |
| Cl¯                            | 2.563     | ±   | 0.014 | 2.054  | ±   | 0.011 | 1.695  | ±   | 0.009   | 4.177  | ±   | 0.022   | 4.227  | ±   | 0.023   | 3.963  | ±  | 0.021 | 5.927  | ±  | 0.032  |
| NO <sub>3</sub>                | 0.830     | ±   | 0.030 | 0.843  | ±   | 0.031 | 0.082  | ±   | 0.004   | 0.269  | ±   | 0.010   | 0.051  | ±   | 0.004   | 1.578  | ±  | 0.057 | 0.107  | ±  | 0.005  |
| SO <sub>4</sub> <sup>2-</sup>  | 3.877     | ±   | 0.034 | 2.199  | ±   | 0.019 | 1.718  | ±   | 0.015   | 3.055  | ±   | 0.027   | 2.549  | ±   | 0.022   | 3.527  | ±  | 0.031 | 4.327  | ±  | 0.038  |
| Total (ions)                   | 12.761    |     |       | 9.548  |     |       | 6.719  |     |         | 15.052 |     |         | 13.647 |     |         | 17.372 |    |       | 19.513 | _  |        |
| Total (oxides + ions)          | 52.072    |     |       | 58.479 |     |       | 77.571 |     |         | 66.527 |     |         | 70.226 |     |         | 59.400 |    |       | 50.647 |    |        |

Appendix B

| Sample                         | DT3_I     | May | / 2015 | DT1_   | lun | e 2015 | DT2_J  | lun | e 2015 | DT3_J  | un | e 2015 | DT1_   | July | y 2015 | DT2_   | Jul | y 2015 | DT3_   | July | y 2015 |
|--------------------------------|-----------|-----|--------|--------|-----|--------|--------|-----|--------|--------|----|--------|--------|------|--------|--------|-----|--------|--------|------|--------|
| Major and minor eleme          | nts as ox | ide | s (%)  |        |     |        |        |     |        |        |    |        |        |      |        |        |     |        |        |      |        |
| SiO <sub>2</sub>               | 27.167    | ±   | 0.051  | 28.318 | ±   | 0.053  | 18.530 | ±   | 0.035  | 24.158 | ±  | 0.045  | 13.820 | ±    | 0.027  | 12.693 | ±   | 0.025  | 23.221 | ±    | 0.037  |
| TiO <sub>2</sub>               | 0.448     | ±   | 0.001  | 0.478  | ±   | 0.002  | 0.306  | ±   | 0.001  | 0.370  | ±  | 0.001  | 0.261  | ±    | 0.001  | 0.241  | ±   | 0.001  | 0.200  | ±    | 0.002  |
| Al <sub>2</sub> O <sub>3</sub> | 3.777     | ±   | 0.059  | 3.692  | ±   | 0.062  | 2.919  | ±   | 0.050  | 3.067  | ±  | 0.045  | 1.886  | ±    | 0.041  | 2.030  | ±   | 0.037  | 2.694  | ±    | 0.148  |
| Fe <sub>2</sub> O <sub>3</sub> | 3.972     | ±   | 0.008  | 3.906  | ±   | 0.008  | 2.782  | ±   | 0.006  | 3.346  | ±  | 0.006  | 2.287  | ±    | 0.005  | 2.154  | ±   | 0.005  | 1.935  | ±    | 0.014  |
| MnO                            | 0.083     | ±   | 0.002  | 0.077  | ±   | 0.002  | 0.050  | ±   | 0.002  | 0.068  | ±  | 0.002  | 0.050  | ±    | 0.002  | 0.044  | ±   | 0.002  | 0.050  | ±    | 0.007  |
| CaO*                           | 1.983     | ±   | 0.016  | 1.681  | ±   | 0.014  | 1.179  | ±   | 0.013  | 1.745  | ±  | 0.014  | 0.427  | ±    | 0.010  | 1.179  | ±   | 0.011  | 1.148  | ±    | 0.013  |
| K <sub>2</sub> O*              | 0.508     | ±   | 0.006  | 0.513  | ±   | 0.007  | 0.366  | ±   | 0.008  | 0.467  | ±  | 0.006  | 0.208  | ±    | 0.008  | 0.226  | ±   | 0.008  | 0.359  | ±    | 0.009  |
| P <sub>2</sub> O <sub>5</sub>  | 0.170     | ±   | 0.002  | 0.590  | ±   | 0.002  | 0.383  | ±   | 0.002  | 0.389  | ±  | 0.001  | 0.753  | ±    | 0.002  | 0.457  | ±   | 0.001  | 0.288  | ±    | 0.005  |
| Total (oxides)                 | 38.108    |     |        | 39.254 |     |        | 26.514 |     |        | 33.609 |    |        | 19.692 |      |        | 19.024 |     |        | 29.895 |      |        |
| Trace elements (ppm)           |           |     |        |        |     |        |        |     |        |        |    |        |        |      |        |        |     |        |        |      |        |
| V                              | 88        |     |        | 111    | ±   | 2      | 86     | ±   | 2      | 91     | ±  | 1      | 115    | ±    | 1      | 111    | ±   | 1      | 77     | ±    | 5      |
| Cr                             | 71        |     |        | 96     |     |        | 65     | ±   |        | 82     | ±  |        | 160    | ±    | 5      | 60     |     |        | 67     |      | 21     |
| Ni                             | 62        | ±   | 3      | 84     | ±   | 4      | 49     | ±   | 3      | 72     | ±  | 3      | 133    | ±    | 3      | 71     | ±   | 2      | 91     | ±    | 11     |
| Cu                             | 56        |     |        | 134    |     |        | 59     | ±   |        | 49     |    | 5      |        | ±    |        | 62     |     |        | 25     |      | 19     |
| Zn                             | 251       | ±   | 11     | 430    | ±   | 12     | 297    |     |        | 255    | ±  | 8      | 515    | ±    | 9      | 244    | ±   | 7      | 180    | ±    | 32     |
| As                             | 0         |     | 7      | 0      |     |        | 0      | ±   | 6      | 0      | ±  |        | 0      | ±    | 5      | 0      |     |        | 0      | ±    | 21     |
| Br                             | 58        |     |        | 62     |     |        | 100    |     | _      | 64     | ±  | 5      | 62     | ±    | 5      | 88     | ±   | 4      | 37     |      | 19     |
| Rb                             | 24        |     |        | 35     |     |        | 16     |     |        | 23     |    | 1      |        | ±    |        | 12     |     |        |        | ±    |        |
| Sr                             | 392       |     | _      | 322    |     |        | 276    |     | _      | 291    | ±  |        | 233    |      |        | 268    |     |        | 284    |      | 19     |
| Υ                              | 14        |     |        | 23     |     |        | 14     |     |        | 19     | ±  |        |        | ±    |        | 12     |     |        |        |      | 19     |
| Zr                             | 123       |     | 8      | 149    |     |        | 112    |     | _      | 82     | ±  |        |        | ±    |        | 83     |     |        |        |      | 24     |
| Мо                             | 0         |     | 10     |        |     | 10     | _      | ±   | _      | 0      |    | 7      |        | ±    |        | 0      |     |        |        |      | 29     |
| Pb                             | 18        | ±   | 10     | 12     | ±   | 11     | 15     | ±   | 9      | 17     | ±  | 8      | 22     | ±    | 8      | 30     | ±   | 7      | 45     | ±    | 32     |
| Water soluble ions (%)         |           |     |        |        |     |        |        |     |        |        |    |        |        |      |        |        |     |        |        |      |        |
| NH <sub>4</sub> <sup>+</sup>   | 0.091     | ±   | 0.010  | 0.125  | ±   | 0.014  | 0.120  | ±   | 0.013  | 0.100  | ±  | 0.011  | 0.088  | ±    | 0.010  | 0.032  | ±   | 0.004  | 0.109  | ±    | 0.012  |
| Na <sup>†</sup>                | 4.509     | ±   | 0.031  | 3.505  | ±   | 0.025  | 5.875  | ±   | 0.041  | 2.834  | ±  | 0.020  | 3.466  | ±    | 0.024  | 5.538  | ±   | 0.038  | 5.360  | ±    | 0.037  |
| K <sup>+</sup>                 | 0.276     | ±   | 0.010  | 0.312  | ±   | 0.012  | 0.352  | ±   | 0.013  | 0.256  | ±  | 0.010  | 0.339  | ±    | 0.013  | 0.360  | ±   | 0.013  | 0.390  | ±    | 0.015  |
| Mg <sup>2+</sup>               | 0.572     | ±   | 0.008  | 0.559  | ±   | 0.008  | 0.947  | ±   | 0.013  | 0.453  | ±  | 0.006  | 0.582  | ±    | 0.008  | 0.878  | ±   | 0.012  | 0.628  | ±    | 0.009  |
| Ca <sup>2+</sup>               | 3.582     |     |        |        |     | 0.026  |        |     | 0.028  |        |    | 0.026  |        |      | 0.024  |        |     | 0.022  | 2.833  |      |        |
| Cl                             | 6.828     |     |        |        |     | 0.020  |        |     | 0.042  |        |    | 0.019  |        |      | 0.017  |        |     | 0.032  |        |      | 0.041  |
| NO <sub>3</sub>                | 1.328     | ±   | 0.048  | 0.595  | ±   | 0.022  | 0.349  | ±   | 0.013  | 0.939  | ±  | 0.034  | 1.573  | ±    | 0.057  | 1.702  | ±   | 0.061  | 1.864  | ±    | 0.067  |
| SO <sub>4</sub> <sup>2-</sup>  | 3.649     | ±   | 0.032  | 2.965  | ±   | 0.026  | 4.878  | ±   | 0.043  | 2.483  | ±  | 0.022  | 2.923  | ±    | 0.026  | 4.424  | ±   | 0.039  | 3.087  | ±    | 0.027  |
| Total (ions)                   | 20.836    |     |        | 14.804 |     |        | 23.663 |     |        | 13.719 |    |        | 14.879 |      |        | 21.580 |     |        | 21.882 |      |        |
| Total (oxides + ions)          | 58.944    |     |        | 54.058 |     |        | 50.177 |     |        | 47.328 |    |        | 34.571 |      |        | 40.605 |     |        | 51.777 |      |        |

Appendix B

| Sample                         | DT1_      | Aug | g <b>201</b> 5 | DT2_   | <u>Au</u> | g 2015 | DT3_   | Au | g 2015 | DT1_9  | ept | t_2015 | DT2_5  | ept | t_2015 | DT3_9  | Sep | t_2015 | DT1_   | Oct | 2015  |
|--------------------------------|-----------|-----|----------------|--------|-----------|--------|--------|----|--------|--------|-----|--------|--------|-----|--------|--------|-----|--------|--------|-----|-------|
| Major and minor eleme          | nts as ox | ide | s (%)          |        |           |        |        |    |        |        |     |        |        |     |        |        |     |        |        |     |       |
| SiO <sub>2</sub>               | 34.862    | ±   | 0.061          | 33.619 | ±         | 0.058  | 29.244 | ±  | 0.054  | 50.971 | ±   | 0.083  | 11.690 | ±   | 0.024  | 42.544 | ±   | 0.074  | 41.270 | ±   | 0.067 |
| TiO <sub>2</sub>               | 0.591     | ±   | 0.001          | 0.469  | ±         | 0.002  | 0.422  | ±  | 0.002  | 0.605  | ±   | 0.003  | 0.206  | ±   | 0.001  | 0.609  | ±   | 0.002  | 0.496  | ±   | 0.002 |
| Al <sub>2</sub> O <sub>3</sub> | 4.021     | ±   | 0.033          | 3.773  | ±         | 0.078  | 3.514  | ±  | 0.068  | 5.057  | ±   | 0.124  | 3.628  | ±   | 0.053  | 5.114  | ±   | 0.081  | 4.374  | ±   | 0.115 |
| Fe <sub>2</sub> O <sub>3</sub> | 5.488     | ±   | 0.008          | 4.186  | ±         | 0.010  | 3.878  | ±  | 0.009  | 5.479  | ±   | 0.015  | 2.206  | ±   | 0.006  | 5.540  | ±   | 0.011  | 4.592  | ±   | 0.013 |
| MnO                            |           |     | 0.001          |        |           | 0.003  | 0.068  |    | 0.003  |        |     | 0.005  |        |     | 0.002  |        |     | 0.003  | 0.081  |     | 0.005 |
| CaO*                           | 1.772     |     |                |        |           | 0.017  |        |    | 0.014  |        |     | 0.019  |        |     | 0.006  |        |     | 0.014  | 2.690  |     | 0.018 |
| K <sub>2</sub> O*              | 0.598     | ±   | 0.003          | 0.850  | ±         | 0.002  | 0.709  | ±  | 0.003  | 0.988  | ±   | 0.002  | 0.000  | ±   | 0.014  |        |     | 0.003  | 0.756  | ±   | 0.004 |
| P <sub>2</sub> O <sub>5</sub>  | 0.249     | ±   | 0.001          | 0.158  | ±         | 0.002  | 0.076  | ±  | 0.002  |        |     | 0.004  | 0.000  | ±   | 0.001  |        |     | 0.002  |        | ±   | 0.003 |
| Total (oxides)                 | 47.676    |     |                | 48.204 |           |        | 41.334 |    |        | 68.110 |     |        | 21.051 |     |        | 55.547 |     |        | 54.306 |     |       |
| Trace elements (ppm)           |           |     |                |        |           |        |        |    |        |        |     |        |        |     |        |        |     |        |        |     |       |
| V                              | 122       | ±   | 1              | 110    | ±         | 3      | 42     | ±  | 2      | 51     | ±   | 4      | 19     | ±   | 2      | 93     | ±   | 2      | 0      | ±   | 4     |
| Cr                             | 116       | ±   | 3              | 59     |           | 10     | 76     | ±  | 9      | 96     |     | 16     | 46     | ±   | 6      | 93     |     | 10     | 152    | ±   | 15    |
| Ni                             | 91        | ±   | 1              | 71     | ±         | 5      | 54     | ±  | 4      | 61     | ±   | 8      | 29     | ±   | 3      | 76     | ±   | 5      | 73     | ±   | 8     |
| Cu                             | 55        | ±   | 2              | 76     | ±         | 9      | 41     | ±  | 7      | 55     | ±   | 14     | 32     | ±   | 5      | 86     | ±   | 8      | 40     | ±   | 13    |
| Zn                             | 194       | ±   | 4              | 226    | ±         | 15     | 127    | ±  | 13     | 157    | ±   | 24     | 107    | ±   | 9      | 339    | ±   | 15     | 185    | ±   | 23    |
| As                             | 0         | ±   | 3              | 0      | ±         | 10     | 0      | ±  | 9      | 0      | ±   | 16     | 0      | ±   | 6      | 0      | ±   | 10     | 0      | ±   | 15    |
| Br                             | 20        | ±   | 2              | 16     | ±         |        | 34     |    | 7      | 17     | ±   | 14     | 715    | ±   | 7      | 58     |     |        | 64     |     | 13    |
| Rb                             | 22        | ±   | 1              | 43     | ±         |        | 12     |    | 2      | 47     |     | 4      | 18     |     |        | 38     |     | 2      | 24     | ±   | _     |
| Sr                             | 441       |     |                | 369    | ±         |        | 257    |    | _      | 394    | ±   | 14     | 179    |     |        | 345    | ±   |        | 331    | ±   | 13    |
| Υ                              | 18        |     | _              | 9      |           | 9      | 25     |    | 7      | 23     |     | 14     |        | ±   |        | 14     |     |        | 18     |     | 13    |
| Zr                             | 141       |     |                | 133    |           |        | 125    |    | 10     | 123    |     | 20     | 62     |     |        | 127    |     | 12     | 99     |     | 17    |
| Mo                             |           | ±   |                | 0      |           | 14     | 0      |    | 12     | 0      |     | 23     |        | ±   |        | 3      |     | 13     | 17     |     | 21    |
| Pb                             | 4         | ±   | 4              | 58     | ±         | 15     | 63     | ±  | 13     | 36     | ±   | 24     | 12     | ±   | 9      | 28     | ±   | 14     | 46     | ±   | 23    |
| Water soluble ions (%)         |           |     |                |        |           |        |        |    |        |        |     |        |        |     |        |        |     |        |        |     |       |
| NH <sub>4</sub> <sup>+</sup>   | 0.025     | ±   | 0.003          | 0.018  | ±         | 0.002  | 0.021  | ±  | 0.002  | 0.021  | ±   | 0.003  | 0.027  | ±   | 0.003  | 0.024  | ±   | 0.003  | 0.013  | ±   | 0.002 |
| Na <sup>+</sup>                | 1.044     | ±   | 0.009          | 0.481  | ±         | 0.006  | 1.144  | ±  | 0.009  | 0.756  | ±   | 0.007  | 12.685 | ±   | 0.087  | 0.862  | ±   | 0.008  | 2.134  | ±   | 0.016 |
| K <sup>+</sup>                 | 0.116     | ±   | 0.004          | 0.094  | ±         | 0.004  | 0.133  | ±  | 0.005  | 0.106  | ±   | 0.004  | 0.625  | ±   | 0.023  | 0.123  | ±   | 0.005  | 0.192  | ±   | 0.007 |
| Mg <sup>2+</sup>               | 0.213     | ±   | 0.003          | 0.133  | ±         | 0.002  | 0.193  | ±  | 0.003  | 0.146  | ±   | 0.002  | 1.732  | ±   | 0.024  | 0.261  | ±   | 0.004  | 0.425  | ±   | 0.006 |
| Ca <sup>2+</sup>               | 3.101     | ±   | 0.026          | 1.772  | ±         | 0.015  | 1.840  |    | 0.016  | 2.143  | ±   | 0.018  | 1.280  | ±   | 0.011  | 1.928  | ±   | 0.016  | 3.415  | ±   | 0.029 |
| Cl                             |           |     | 0.009          |        |           | 0.004  | 1.460  |    | 0.008  |        |     | 0.007  | 23.054 |     | 0.123  |        |     | 0.006  | 3.267  |     | 0.018 |
| NO <sub>3</sub>                | 1.045     |     |                |        |           | 0.015  | 0.943  |    | 0.034  |        |     | 0.015  |        |     | 0.003  |        |     | 0.003  | 0.009  |     | 0.003 |
| SO <sub>4</sub> <sup>2</sup>   | 3.241     | ±   | 0.028          | 1.425  | ±         | 0.012  | 1.591  | ±  | 0.014  | 2.047  | ±   | 0.018  | 2.778  | ±   | 0.024  | 1.743  | ±   | 0.015  | 3.042  | ±   | 0.027 |
| Total (ions)                   | 10.410    |     |                | 4.930  |           |        | 7.324  |    |        | 6.831  |     |        | 42.187 |     |        | 5.980  |     |        | 12.496 |     |       |
| Total (oxides + ions)          | 58.085    |     |                | 53.134 |           |        | 48.658 |    |        | 74.941 |     |        | 63.238 |     |        | 61.527 |     |        | 66.802 |     |       |

Appendix B

| Sample                         | DT2_      | Oct | 2015    | DT3_                               | Oct  | 2015      | DT1_      | Nov | 2015  | DT2_   | No | v 2015 | DT3_   | No | v 2015 | DT1_   | De | 2015  | DT2_   | Dec | 2015  | DT3_   | De | c 2 | 015   |
|--------------------------------|-----------|-----|---------|------------------------------------|------|-----------|-----------|-----|-------|--------|----|--------|--------|----|--------|--------|----|-------|--------|-----|-------|--------|----|-----|-------|
| Major and minor eleme          | nts as ox | ide | s (%)   |                                    |      |           |           |     |       |        |    |        |        |    |        |        |    |       |        |     |       |        |    |     |       |
| SiO <sub>2</sub>               | 14.274    | ±   | 0.028   | 38.421                             | ±    | 0.068     | 42.440    | ±   | 0.076 | 23.596 | ±  | 0.044  | 38.454 | ±  | 0.070  | 33.248 | ±  | 0.059 | 28.716 | ±   | 0.053 | 28.860 | ±  | 0   | 0.052 |
| TiO <sub>2</sub>               | 0.206     | ±   | 0.001   | 0.587                              | ±    | 0.001     | 0.687     | ±   | 0.002 | 0.481  | ±  | 0.001  | 0.651  | ±  | 0.002  | 0.653  | ±  | 0.001 | 0.486  | ±   | 0.001 | 0.507  | ±  | 0   | 0.001 |
| Al <sub>2</sub> O <sub>3</sub> | 3.483     | ±   | 0.048   | 4.611                              | ±    | 0.045     | 5.114     | ±   | 0.052 | 3.202  | ±  | 0.046  | 5.353  | ±  | 0.069  | 4.657  | ±  | 0.040 | 3.472  | ±   | 0.055 | 3.603  | ±  | 0   | 0.038 |
| Fe <sub>2</sub> O <sub>3</sub> | 2.118     | ±   | 0.005   | 5.577                              | ±    | 0.008     | 6.509     | ±   | 0.010 | 4.736  | ±  | 0.008  | 7.170  | ±  | 0.012  | 6.180  | ±  | 0.009 | 4.276  | ±   | 0.008 | 4.597  | ±  | 0   | 0.007 |
| MnO                            | 0.041     |     |         | 0.103                              | ±    | 0.001     |           |     | 0.002 |        |    | 0.002  |        |    | 0.003  |        |    | 0.001 | 0.085  | ±   | 0.002 | 0.079  |    | _   |       |
| CaO*                           |           |     | 0.008   |                                    |      | 0.015     |           |     | 0.021 |        |    | 0.010  |        |    | 0.008  |        |    | 0.011 |        |     | 0.015 | 2.502  |    | _   |       |
| K <sub>2</sub> O*              | 0.090     | ±   | 0.010   | 0.774                              | ±    | 0.002     | 0.822     | ±   | 0.002 | 0.552  | ±  | 0.003  | 0.954  | ±  | 0.002  | 0.751  | ±  | 0.002 | 0.590  | ±   | 0.003 | 0.592  | ±  | 0   | 0.003 |
| P <sub>2</sub> O <sub>5</sub>  | 0.000     | ±   | 0.001   | 0.106                              | ±    | 0.001     | 0.000     | ±   | 0.001 |        |    | 0.001  |        |    | 0.002  |        |    | 0.001 |        |     | 0.002 | 0.102  |    | _   |       |
| Total (oxides)                 | 25.953    |     |         | 53.537                             |      |           | 56.429    |     |       | 34.418 |    |        | 55.254 |    |        | 48.064 |    |       | 39.745 | -   |       | 40.840 | _  | Ť   |       |
|                                |           |     |         |                                    |      |           |           |     |       |        |    |        |        |    |        |        |    |       |        |     |       |        |    |     |       |
| Trace elements (ppm)           |           |     |         |                                    |      |           |           |     |       |        |    |        |        |    |        |        |    |       |        |     |       |        |    |     |       |
| V                              | 67        | ±   | 1       | 125                                | ±    | 1         | 130       | ±   | 1     | 149    | ±  | 2      | 180    | ±  | 2      | 144    | ±  | 1     | 97     | ±   | 2     | 65     | ±  | 1   |       |
| Cr                             | 26        | ±   | 5       | 110                                | ±    | 4         | 119       | ±   | 5     | 300    | ±  | 6      | 135    | ±  | 8      | 218    | ±  | 4     | 103    | ±   | 7     | 71     | ±  | : 4 | ļ     |
| Ni                             | 33        | ±   | 3       | 84                                 | ±    | 2         | 68        | ±   | 2     | 971    | ±  | 3      | 121    | ±  | 4      | 211    | ±  | 2     | 134    | ±   | 3     | 64     | ±  | 2   | 2     |
| Cu                             | 28        | ±   | 5       | 111                                | ±    | 4         | 59        | ±   | 4     | 102    | ±  | 5      | 74     | ±  | 7      | 78     | ±  | 3     | 89     | ±   | 6     | 55     | ±  | : 3 | 3     |
| Zn                             | 82        | ±   | 8       | 250                                | ±    | 7         | 205       | ±   | 8     | 507    | ±  | 9      | 358    | ±  | 12     | 534    | ±  | 6     | 328    | ±   | 10    | 231    | ±  | 6   | 5     |
| As                             | 0         | ±   | 5       | 0                                  | ±    | 4         | 0         | ±   | 5     | 0      | ±  | 5      | 0      | ±  | 8      | 0      | ±  | 3     | 0      | ±   | 7     | 3      | ±  | 4   | ļ     |
| Br                             | 457       | ±   | 5       | 27                                 | ±    | 4         | 56        | ±   | 4     | 66     | ±  | 5      | 61     | ±  | 7      | 59     | ±  | 3     | 112    | ±   | 6     | 95     | ±  | : 4 | ļ     |
| Rb                             | 11        | ±   | 1       | 40                                 | ±    | 1         | 35        | ±   | 1     | 26     | ±  | 1      | 34     | ±  | 2      | 27     | ±  | 1     | 22     | ±   | 2     | 22     | ±  | : 1 |       |
| Sr                             | 227       | ±   | 5       | 422                                | ±    | 4         | 683       | ±   | 5     | 253    | ±  | 5      | 210    | ±  | 7      | 329    | ±  | 3     | 266    | ±   | 6     | 310    | ±  | : 4 | ļ     |
| Υ                              | 14        | ±   | 5       | 13                                 | ±    | 4         | 14        | ±   | 4     | 15     | ±  | 5      | 39     | ±  | 7      | 23     | ±  | 3     | 13     | ±   | 6     | 19     | ±  | 3   | 3     |
| Zr                             | 38        | ±   | 6       | 134                                | ±    | 5         | 143       | ±   | 6     | 103    | ±  | 6      | 192    | ±  | 10     | 139    | ±  | 4     | 113    | ±   | 8     | 95     | ±  | 5   | 5     |
| Мо                             | 0         | ±   | 7       | 1                                  | ±    | 6         | 0         | ±   | 7     | 3      | ±  | 7      | 0      | ±  | 10     | 1      | ±  | 5     | 3      | ±   | 9     | 0      | ±  | 5   | 5     |
| Pb                             | 8         | ±   | 8       | 36                                 | ±    | 6         | 17        | ±   | 7     | 20     | ±  | 8      | 24     | ±  | 11     | 22     | ±  | 5     | 30     | ±   | 10    | 40     | ±  | : 6 | 5     |
| Water soluble ions (%)         |           |     |         |                                    |      |           |           |     |       |        |    |        |        |    |        |        |    |       |        |     |       |        |    |     |       |
| NH <sub>4</sub> <sup>+</sup>   | 0.047     | +   | 0.005   | 0.043                              | +    | 0.005     | 0.028     | +   | 0 003 | 0.024  | +  | 0.003  | 0.025  | +  | 0.003  | 0.032  | +  | 0.004 | 0.011  | +   | 0.001 | 0.025  | +  |     | 003   |
| Na <sup>+</sup>                | 10.668    |     |         |                                    |      | 0.003     | 0.675     |     |       |        |    | 0.003  |        |    | 0.003  |        |    | 0.004 |        |     | 0.001 | 1.540  |    | -   |       |
| Na<br>K <sup>†</sup>           |           |     |         |                                    |      |           |           |     |       |        |    |        |        |    |        |        |    |       |        |     |       |        |    |     |       |
|                                | 0.447     |     |         | 0.086                              |      |           |           |     | 0.004 |        |    | 0.005  |        |    | 0.003  |        |    | 0.003 |        |     | 0.005 | 0.135  |    |     |       |
| Mg <sup>2+</sup>               | 1.315     |     |         |                                    |      | 0.002     |           |     | 0.002 |        |    | 0.003  |        |    | 0.001  |        |    | 0.001 |        |     | 0.003 | 0.225  |    |     |       |
| Ca <sup>2+</sup>               | 2.543     | ±   | 0.022   | 2.506                              | ±    | 0.021     | 1.851     | ±   | 0.016 | 2.059  | ±  | 0.018  | 0.573  | ±  | 0.005  | 1.949  | ±  | 0.017 | 3.422  | ±   | 0.029 | 2.815  | ±  | 0   | 0.024 |
| Cl                             | 18.796    | ±   | 0.101   | 0.910                              | ±    | 0.005     | 1.181     | ±   | 0.006 | 1.246  | ±  | 0.007  | 1.071  | ±  | 0.006  | 1.120  | ±  | 0.006 | 2.083  | ±   | 0.011 | 2.494  | ±  | 0   | 0.013 |
| NO <sub>3</sub>                | 0.009     | ±   | 0.003   | 0.007                              | ±    | 0.003     | 0.029     | ±   | 0.003 | 0.004  | ±  | 0.003  | 0.007  | ±  | 0.003  | 0.033  | ±  | 0.003 | 0.205  | ±   | 0.008 | 0.026  | ±  | 0   | 0.003 |
| SO <sub>4</sub> <sup>2-</sup>  | 3.581     | ±   | 0.031   | 1.411                              | ±    | 0.012     | 1.133     | ±   | 0.010 | 2.297  | ±  | 0.020  | 0.697  | ±  | 0.006  | 1.734  | ±  | 0.015 | 2.188  | ±   | 0.019 | 1.964  | ±  | 0   | 0.017 |
| Total (ions)                   | 37.407    |     |         | 5.704                              |      |           | 5.146     |     |       | 6.831  |    |        | 3.196  |    |        | 5.745  |    |       | 9.558  |     |       | 9.224  |    |     |       |
| Total (oxides + ions)          | 63.360    |     |         | 59.240                             |      |           | 61.574    |     |       | 41.249 |    |        | 58.450 |    |        | 53.810 |    |       | 49.303 |     |       | 50.065 |    |     |       |
| Note: CaO* and K2O* a          | re water  | ins | oluble. | P <sub>2</sub> O <sub>s</sub> cald | cula | ated from | m total P |     |       |        |    |        |        |    |        |        |    |       |        |     |       |        |    |     |       |

#### Supplement A

#### Sampling Sites Meta-data

The NEO sampling site (DT3, DT4) is within a fenced area about 200 m in length and 50 m in width, situated along the southeastern border of the KAUST campus. The soil surface is covered by a layer of gravel and paved walkways, to contain local dust emissions. The Frisbee deposition samplers as well as several experimental photovoltaic (PV) and meteorological systems are installed at the NEO site. Besides the regional dust, the site is impacted by local emissions from vehicles traveling along the paved road to the south and in the paved parking lot to the north. Depending on the wind, the site was periodically exposed to dirt road and construction dust from building activities immediately outside the KAUST campus.

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The CMOR sampling site (DT3) is on a concrete paved quay about 200 m in length and 50 m wide, providing docking facilities for small and medium size boats in the KAUST harbor. The deposition sampler was set up close to the furthest edge of the quay, approximately 5m from the water's edge, about 1m above the water line, and approximately 65 m from the CMOR building entrance. The sampler is exposed to local emissions from cargo loading activities and other traffic, as well as sea spray during stormy conditions.

One Frisbee sampler (DT1) was installed on the pebble covered flat garage rooftop of a residential home (G3705), located approximately 1.5 km to the beach area in the west and about 1 km from the harbor to the south. The site is impacted by local paved road traffic, a nearby bus terminal, and activities at a local shopping center. The sampler was set about 5m above the street level, and to some extent above street level dust and local transport emissions.

#### Supplement B

#### **Dust Deposition on Solar Panels**

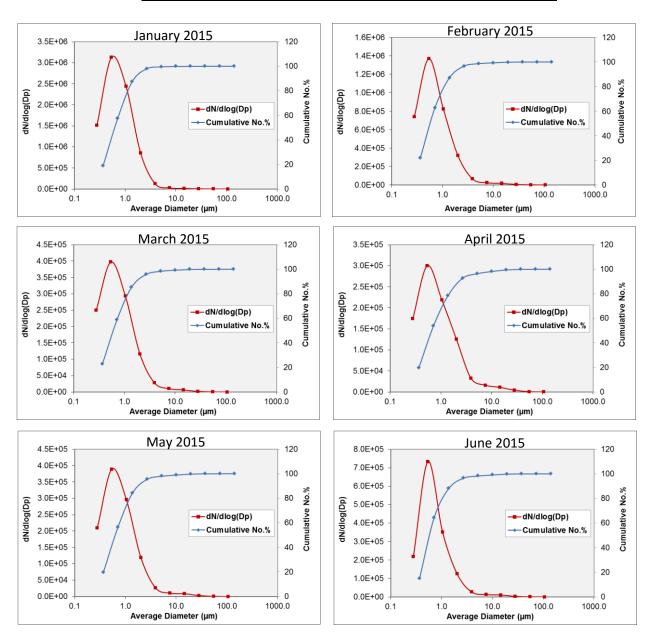
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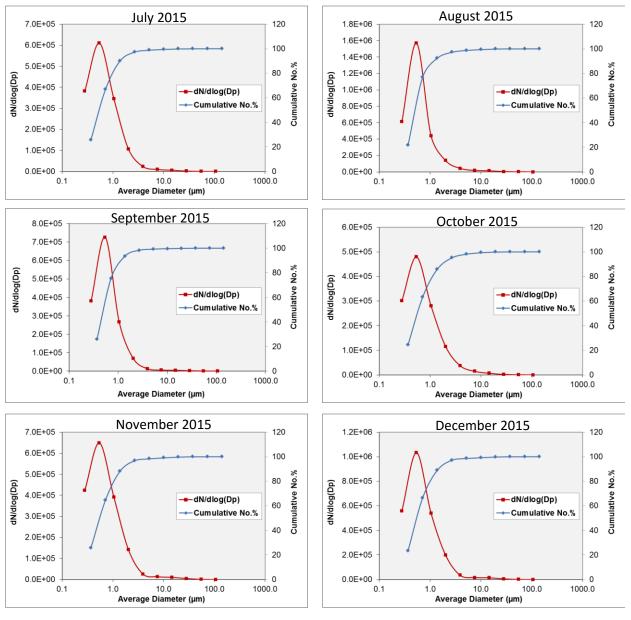
Dust deposits on solar panels are known to have a severe detrimental effect on the efficiency of photovoltaic systems (Goossens and Van Kerschaever, 1999; Hamou et al., 2014; Mejia et al., 2014; Rao et al., 2014; Sulaiman et al., 2014; Ilse et al., 2016). High humidity is experienced at KAUST throughout the year, more often during the late summer months of July to September. This is reflected by dew being formed on radiating cool surfaces such as solar panels, as well as coastal fog during the early morning hours. The gypsum component of dust being collected by the Frisbee samplers and on all exposed surfaces is being partly dissolved by the dew, and on drying being recrystallized. XRD measurements performed directly on dust collected on zero background silicon wafers exposed over a period of several months showed the dust surface to be hardened by the crystallization of blades of gypsum. These surface crusts of gypsum were shown to have a distinct preferred orientation, having their (010) crystal planes parallel to the surface of the silicon wafer, as will be the case on other flat surfaces. The cementation of dust on glass surfaces by gypsum encrustation increases the adhesion of dust on the solar panels, and a resultant attenuation of solar panel efficiency. Dust mineralogy and mineralogical interrelationships, together with climatic conditions are variables determining the nature of dust deposits on solar panels. This needs to be understood with the planning and placement of solar arrays.

# SEM based particle number distribution curves for 12 months of 2015



#### Supplement C

# SEM based particle number distribution curves for 12 months of 2015



# Co-Editor Decision: Publish subject to technical corrections (15 Aug 2017) by Yves Balkanski

#### Comments to the Author:

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- 1. Please adjust the following sentence in the abstract to make it easier for the reader to follow your thought: "The particle size distributions provide assessments of < 10  $\mu$ m and < 2.5  $\mu$ m dust deposition rates, and it is suggested that these furthermore represent proxies for PM10 (coarse) and PM2.5 (fine) particle size fractions, respectively." change to
- "The particle size distributions provide assessments of < 10  $\mu m$  and < 2.5  $\mu m$  dust deposition rates. Since more than xx to yy% of the aerosol composition is made up of dust, these represent proxies for PM10 (coarse) and PM2.5 (fine) particle size fractions, respectively."

**Authors Response:** Agreed. We rephrased the sentence to read "The particle size distributions measured on the resuspended samples provide assessments of  $<10 \, \mu m$  and  $<2.5 \, \mu m$  dust deposition rates. It is suggested that these represent proxies for  $PM_{10}$  (coarse) and  $PM_{2.5}$  (fine) particle size fractions in the dust deposits, respectively."

2. You response to the following point from Reviewer 1:
"27. Page 10, AERONET: You should either use the Angstrom exponent to screen out non-dusty days or use SDA coarse mode optical depth."
deserves to be treated in the manuscript. Please incorporate the most relevant part of your answer in the text of the paper.

**Authors Response**: Following the reviewer's suggestion, we compared the deposition rate with the AOD, screened by low Ångström exponent values. We have also examined the AERONET coarse mode AOD obtained using the Spectral Decomposition Algorithm (SDA). However, neither of these adjustments improved the correlation between the AOD and deposition rates for the reasons we briefly discuss. We have implemented this information in the revision and the corresponding portion of the text is read now as follows:

The Aerosol Optical Depth (AOD) is one of the best observed aerosol characteristics. It defines the aerosol radiative effect and reflects the abundance of aerosols in the atmosphere.

A CIMEL Robotic Sun Photometer is installed on the rooftop of the CMOR building on the campus of the KAUST and operated by our group since 2012, as a part of the NASA-AERONET, providing aerosol optical depth (AOD) and aerosol retrieved characteristics (https://aeronet.gsfc.nasa.gov/). Figure 6a compares the monthly averaged AOD at 500 nm with the dust deposition rate for 2015. In a general sense the AOD and the deposition rates show comparable trends, both with maxima in spring and larger maxima in fall. However, the AOD reaches a first maximum in April, being one month later than that of the deposition rate. Also, the larger second AOD maximum occurred in August while the maximum deposition rate is broadly distributed over a three-month period, from August to October, with October AOD being relatively low. The linear correlation coefficient between the monthly deposition rates and the monthly averaged AOD of 0.40 suggests a causal interrelationship between these two quantities, however, for a number of reasons it is relatively low. The photometer measures light attenuation by all aerosols along a column in the atmosphere, while deposition rate depends on dust at ground level only, the latter generally containing a relatively coarser dust fraction. The low-level dust particles are partly from local dust sources while the higher altitude dust could be transported from distal sources and chemically transformed, i.e., aged. As was pointed out by Yu et al. (2013) the differences between the deposition and AOD time series can in part be attributed to modifications of the natural dust aerosol by anthropogenic activities, including petrochemical and other large industries along the Red Sea coast, as well as by entrainment of construction and road dust. The substantial contamination of dust by anthropogenic species and sea salt is not likely in this area, as was suggested by observational (Osipov et al., 2015; Brindley et al., 2015) and modeling studies (Kalenderski and Stenchikov, 2016). To further test whether the non-dust fine aerosols (or remotely transported fine dust) significantly contribute to the interrelation between AOD and deposition rates, we tried two additional approaches. We screened the AODs by low Angstrom exponent value (<0.3), that is suggested to correspond to dust (Ginoux et., 2012), and considered the contribution to the total AOD from the SDA (Spectral De-Convolution Algorithm) fine mode aerosol product. Neither technique lead to increase of correlation coefficient. The discrepancy of high AOD – low deposition in summer (when Angstrom Exponent is low) and low AOD – high deposition

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in October still remained when AODs were screened. The fine mode AOD is almost constant throughout the year, and its contribution cannot explain the discrepancy as well.

3. What the reviewer implies from comment 28

("28. Page 10, Line 26: "dust particles are predominantly from local sources" but in

Abstract you wrote "dust deposits along the Red Sea coast are a mixture of dust emission

from local soils, and soils imported from distal sources." This is contradictory.) is that

dust particles are most more numerous in source regions than the ones that are

transported through medium and long-range transport, please just state this in your

sentence.

**Authors Response**: Added in the Abstract "The dust deposits along the Red Sea coast are considered to be a mixture of dust emissions from local soils, and soils imported from distal dust sources. Airborne mineral concentrations are greatest at or close to dust sources, compared to those through medium and long-range transport.

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4. You could include your answer to the first point raised by reviewer 3 at the end of your introduction. Namely you state that:

"Particle size distribution plots of 12 deposition samples collected monthly at the KAUST campus throughout the 2015 period are added as Appendix A and Supplement C. The mineralogical data are described under paragraph 5.5 and the normative mineralogy calculated from the chemistry in Fig. 11. The chemical data tables are renumbered as Appendix B."

Please contract this formulation and indicate it at the end of your introduction.

Authors Response: More appropriately added as a last paragraph to the Sampling and analysis. "Particle size distribution plots of 12 deposition samples collected monthly at the KAUST campus throughout the 2015 period are shown by volume in Appendix A and by number in Supplement C. The chemical abundance tables are in Appendix B. The mineralogical results from XRD are described under paragraph 5.5 and the normative mineralogy calculated from the chemistry, presented as histogram plots in Figure 11."

5. You do not clearly justify why you did not use the Angström exponent as 2 reviewers suggested. Could you make an argument about what you would have learned had you done it?

**Authors Response:** Please refer to the authors response to the Editor's comment 2.

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6. 3. Please write 1 or 2 sentences in the paper to summarize the answer you have to the following point raised by **Reviewer 3** 

"One of the main conclusions is that dust in the Red Sea is a mixture of dust from local soils and dust imported from distal sources, which is something that is already well-known; the paper makes no attempt to quantify the respective contributions. An additional concern that I have is the selection of the sites to measure dust deposition. It is not very clear what is the impact of local construction activities. The authors should make a clear statement in that respect.

**Authors Response**: Added at the end of paragraph 5.2 Gravimetric Analysis. "Source apportionment is considered to be a following step in the Red Sea dust research program. As an approximation of source contributions, the sampler with the lowest deposition rate can be considered to have negligible or contain the least amount of local dust and sea salt (Fig. 5). In the months of December 2014, January, April, March, June, July, and December 2015, the deposition rates at the four sampling sites were similar, and considered to have similar but negligible amounts of dust from local construction, roads, marine salt, or other particulates. In August, it is estimated that 24-56 g m<sup>-2</sup> month<sup>-1</sup> (10-35%) of the dust captured at sites DT3 and DT4 was from local construction and motor traffic. Similarly, it is estimated that the deposition rates of sea salt at DT2 varied from 20-21 g m<sup>-2</sup> month<sup>-1</sup> (51-56%) over the months of September and October."

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Also added at the end of paragraph 6. Summary and Conclusions. "As an approximation of source contributions, the sampler with the lowest deposition rate can be considered to have negligible or the least amount of local dust or sea salt (Fig.5). In the months of December 2014, January, April, March, June, July, and December 2015, the deposition rates at the four sites were similar, and considered to have no or negligible amounts of dust from local construction, campus roads, marine salt, or other particulates."

# **Second marked up revision**

# Physical and chemical properties of deposited airborne particulates over the Arabian Red Sea coastal plain

Johann P. Engelbrecht<sup>1,2</sup>, Georgiy Stenchikov<sup>1</sup>, P. Jish Prakash<sup>1</sup>, Traci Lersch<sup>3</sup>, Anatolii Anisimov<sup>1</sup> and Illia Shevchenko<sup>1</sup>

<sup>1</sup>King Abdullah University of Science and Technology (KAUST), Physical Science and Engineering Division (PSE), Thuwal, 23955-6900, Saudi Arabia.

<sup>2</sup>Desert Research Institute (DRI), Reno, Nevada 89512-1095, U.S.A.

<sup>3</sup> RJ Lee Group Inc., Monroeville, Pennsylvania 15146, U.S.A.

10 Correspondence to: Johann P. Engelbrecht (johann@dri.edu)

#### **Abstract**

Mineral dust is the most abundant aerosol, having a profound impact on the Global energy budget. This research continues our previous studies performed on surface soils in the Arabian Peninsula, focusing on the mineralogical, physical and chemical composition of dust deposits from the atmosphere at the Arabian Red Sea coast. For this purpose, aerosols deposited from the atmosphere are collected during 2015 at six sites on the campus of the King Abdullah University of Science and Technology (KAUST) situated on the Red Sea coastal plain of Saudi Arabia and subjected to the same chemical and mineralogical analysis we conducted on soil samples. Frisbee deposition samplers with foam inserts were used to collect dust and other deposits, for the period December 2014 to December 2015. The average deposition rate measured at KAUST for this period was 14 g m<sup>-2</sup> per month, with lowest values in winter and increased deposition rates in August to October. The particle size distributions provide assessments of <10  $\mu$ m and <2.5  $\mu$ m dust deposition rates, and it is suggested that these represent proxies for PM<sub>10</sub> (coarse) and PM<sub>2.5</sub> (fine) particle size fractions in the dust deposits.

XRD analysis of a subset of samples confirms variable amounts of quartz, feldspars, micas, and halite, with lesser amounts of gypsum, calcite, dolomite, hematite, and amphibole. Freeze-dried samples were re-suspended onto the Teflon® filters for elemental analysis by XRF, while splits from each sample were analyzed for water soluble cations and anions by Ion Chromatography. The dust deposits along the Red Sea coast are considered to be a mixture of dust emissions from local soils, and soils imported from distal dust sources. Airborne mineral concentrations are greatest at or close to dust sources, compared to those through medium and long-range transport. It is not possible to identify the exact origin of deposition samples from the mineralogical and chemical results alone. These aerosol data are the first of their kind from the Red Sea region. They will help assess their potential nutrient input into the Red Sea, as well the impact on human health, industry, and solar panel efficiency. These data will also support dust modeling in this important dust belt source area, by better quantifying dust mass balance and optical properties of airborne dust particles.

# Keywords

Dust mineralogy, chemistry, particle size distribution Frisbee deposition samplers

5 Freeze-dry dust retrieval
Dust deposition rates, AOD and visibility
Proxies for PM<sub>10</sub> and PM<sub>2.5</sub> deposition rates

#### 8. Introduction

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Dust emission and deposition modeling and measurements are required for the assessment of the dust mass budget. Both emission and deposition are under constrained in atmospheric dust models, leading to large uncertainties (Bergametti and Forêt, 2014; Schulz et al., 2012). To improve simulations, the above authors and others suggested the establishment of dust deposition networks in the vicinity of and away from dust source regions, operating throughout the year. In this paper we are presenting results from a network of dust deposition samplers located on the campus of the King Abdullah University of Science and Technology (KAUST) along the Red Sea coast of Saudi Arabia. This is an important dust source region (Ginoux et al., 2012; Prospero et al., 2002), the effect of which extends thousands of kilometers downwind. To better characterize optical, microphysical, and health effects of dust aerosols we conducted detailed chemical, mineralogical and particle size analysis of deposition samples collected from the air.

### 8.1 Importance of mineral dust

Mineral dust is the most abundant atmospheric aerosol, primarily from suspended soils in arid and semi-arid regions on Earth (Buseck et al., 2000; Washington and Todd, 2005; Goudie, 2006; Muhs et al., 2014), including deserts of the Arabian Peninsula (Edgell, 2006). Dust aerosols profoundly affect climate (Haywood and Boucher, 2000; Hsu et al., 2004; Kumar et al., 2014), cloud properties (Twomey et al., 1984; Wang et al., 2010; Huang et al., 2006), visibility (Kavouras et al., 2009; Moosmüller et al., 2005), air quality (Hagen and Woodruff, 1973), atmospheric chemistry and mineralogy (Sokolik and Toon, 1999; Kandler et al., 2007), biogeochemical cycles in the ocean and over land (Jickells et al., 2005; Mahowald, 2009), human health (Bennett et al., 2006; Bennion et al., 2007; De Longueville et al., 2010; Menéndez et al., 2017), and agriculture (Fryrear, 1981; Nihlen and Lund, 1995).

A further important implication of dust emission/deposition processes is associated with the harvesting of the solar renewable energy in the desert areas. Dust deposits on solar panels are known to have a severe detrimental effect on the efficiency of photovoltaic systems (Goossens and Van Kerschaever, 1999; Hamou et al., 2014; Mejia et al., 2014; Rao et al., 2014; Sulaiman et al., 2014; Ilse et al., 2016), with its adverse effects depending on mineral composition and atmospheric conditions.

#### 8.2 Importance of dust mineralogy

The importance of dust mineralogy has long been recognized (Engelbrecht et al., 2016), but only recently the explicit transport of different mineralogical species is implemented in climate models (Perlwitz et al., 2015a, b; Scanza et al., 2015)

The mineralogy and chemical composition of dust generated from the Red Sea coastal region remains uncertain. The Red Sea coastal plain is a narrow highly heterogeneous piedmont area, and existing soil databases do not have the spatial resolution to represent it adequately (Nickovic et al., 2012).

The specific objective of the present study is to examine mineralogical, chemical and morphological information of deposition samples collected on the KAUST campus. This will help to better quantify the ecological impacts, health effects, damage to property, and optical effects of dust blown across this area (Engelbrecht et al., 2009a, b; Weese and Abraham, 2009). Knowledge of the mineralogy of the dust deposits will provide information on refractive indices, which can be used to calculate dust optical

properties, providing input into radiative transfer models, and to assess the impact of dust events on the Red Sea and adjacent coastal plain.

### 8.3 Previous dust studies in the region

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This research complements our dust studies performed in the Arabian Peninsula (Engelbrecht et al., 2009a; Kalenderski et al., 2013; Prakash et al., 2015; Prakash et al., 2016) and globally (Engelbrecht et al., 2016).

The Arabian Peninsula is one of Earth's major sources of atmospheric dust, contributing as much as 11.8% (22–500 Mt  $a^{-1}$ ) of the total (1,877–4000 Mt  $a^{-1}$ ) global dust emissions (Tanaka and Chiba, 2006). The Red Sea, being enveloped by the Arabian and African deserts is strongly impacted by windborne mineral dust. Along with profound influence on the surface energy budget over land and the Red Sea (Kalenderski et al., 2013; Osipov et al., 2015; Brindley et al., 2015), dust is an important source of nutrients, more so for the oligotrophic northern Red Sea waters (Acosta et al., 2013). From preliminary assessments it is estimated that 5 to 6 major dust storms per year impact the Red Sea region, depositing about 6 Mt of mineral dust into the Red Sea (Prakash et al., 2015). Simulations and satellite observations suggest that the coastal dust contribution to the total deposition flux into the Red Sea could be substantial, even during fair weather conditions (Jiang et al., 2009; Anisimov et al., 2017). Therefore, the correct representation of the regional dust balance over the Red Sea coastal plain is especially important. Here we specifically focus on the dust deposition in this area, which helps to constrain the dust mass balance, as well as the dust mineralogy and chemical composition. Dust sources impacting on the Arabian Red Sea coastal region were shown to vary by season, coming from local haboobs and low level jets, delivered from the Tokar delta of Sudan in summer (Kalenderski and Stenchikov, 2016), and transported from the west coast of the Arabian Peninsula (Kalenderski et al., 2013).

Minerals previously identified in continental soils from Middle East dust generating regions include quartz, feldspars, calcite, dolomite, micas, chlorite, kaolinite, illite, smectite, palygorskite, mixed-layer clays, vermiculite, iron oxides, gypsum, hornblende and halite (Engelbrecht et al., 2009b; Engelbrecht et al., 2016; Goudie, 2006; Prakash et al., 2016; Pye, 1987; Scheuvens and Kandler, 2014). It could be expected that similar mineral assemblages would occur in variable proportions in the dust deposition samples collected in the region.

#### 9. Meteorology and climate

With the exception of the area around Jazan in the south, which is impacted by the Indian Ocean monsoon, the Red Sea coastal region has a desert climate characterized by extreme heat. Temperatures measured at the KAUST campus reach 43° C during the summer days, with a drop in night-time temperatures on average of more than 10° C. Although the extreme temperatures here are moderated by the proximity of the Red Sea, summer humidity is often 85 % or higher during periods of the northwesterly *Shamal* winds. Rainfall diminishes from an annual average of 133 mm at Jazan in the south to 56 mm at Jeddah, and 24 mm at Tabuk in the north. http://worldweather.wmo.int/en/city.html?cityId=699.

Vegetation is sparse, being restricted to semi-desert shrubs, and acacia trees along the ephemeral rivers (wadis), providing forage for small herds of goats, sheep and dromedary camels.

During infrequent but severe rainstorms, run-off from the escarpment along wadis produce flash floods in lowland areas. With such events, fine silt and clay deposits are formed on the coastal plain, which are transformed into dust sources during dry and windy periods of the year. The resultant dust is transported and deposited along the coastal plain itself and adjacent Red Sea, by prevailing northwesterly to southwesterly winds, with moderate breezes (wind speed >5.5 m s<sup>-1</sup>) at the coast (http://www.windfinder.com/weather-maps/report/saudiarabia#6/22.999/34.980).

## 10. Objectives

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This study is meant to complement the recently published papers by our research group that characterize the effect of dust storms (Prakash et al., 2015; Kalenderski et al., 2013), evaluate radiative effect of dust (Osipov et al., 2015), analyze soils from the Red Sea coastal plain (Prakash et al., 2016) and dust emissions in the same region (Anisimov et al., 2017). Mineralogical, physical and chemical results are presented of deposition samples collected largely during 2015 at six sites on the campus of KAUST, located approximately 80 km north of Jeddah, along the central part of the Red Sea coastal plain of Saudi Arabia, (Fig. 1).

#### 15 **10.1 Regional dust sources**

The coastal plains of the Arabian Peninsula along the Red Sea and Persian Gulf are among the most populated areas in this region, hosting several major industrial and residential centers. Airborne dust profoundly affects human activities, marine and land ecosystems, climate, air-quality, and human health. Satellite observations suggest that the narrow Red Sea coastal plain is an important dust source, augmented by fine sediment accumulations, scattered vegetation, and variable terrain. Airborne dust carries the mineralogical and chemical signature of a parent soil (Prakash et al., 2016). The purpose of a previous study on 13 soil samples from the Arabian Red Sea coastal area (Prakash et al., 2016) was to better characterize their mineralogical, chemical and physical properties, which in turn improve assessment of dust being deposited in the Red Sea and on land, affecting environmental systems and urban centers. It was found that the Red Sea coastal soils contain major components of quartz and feldspar, as well as lesser but variable amounts of amphibole, pyroxene, carbonate, clays, and micas, with traces of gypsum, halite, chlorite, epidote and oxides. The mineral assemblages in the soil samples were ascribed to the variety of igneous and metamorphic provenance rocks of the Arabian Shield forming the escarpment to the east of the coastal plain.

#### 11. Sampling and analysis

Anisimov et al. (2017) estimated that the eastern Red Sea coastal plain emits about 5–6 Mt of dust annually. Due to its close proximity, a significant portion of this dust is likely to be deposited into the Red Sea, which could be comparable in amount to the estimated annual deposition rate from remote sources during major dust events (Prakash et al., 2015). Therefore, we expect that the total dust deposition into the Red Sea is on the order of 10 Mt a<sup>-1</sup>, but this estimate still needs to be confirmed.

In the past few decades, wind tunnel and field tests have been performed on different designs of deposition samplers and sand traps to compare their efficiencies. The samplers and traps included marble dust collectors (MDCO), inverted Frisbees, and glass surfaces (Goossens and Rajot, 2008; Sow et al., 2006; Goossens et al., 2000; Goossens and Offer, 2000). Most of the experiments performed in wind tunnels failed to completely mimic the field conditions, which resulted in an underestimation of the dust

deposition, more so for the <10  $\mu$ m size fraction (Sow et al., 2006). Based on the field evaluations by Vallack (1995) and suggestions by Vallack and Shillito (1998) the decision was taken to deploy inverted Frisbee samplers with foam inserts.

At each sampling site the particulate deposits were collected into a 227 mm diameter inverted Frisbee dust deposit sampler, each with a polyester foam insert and bird strike preventers (Hall et al., 1993; Vallack and Chadwick, 1992, 1993; Vallack and Shillito, 1998) (Fig. 2). The purpose of the foam insert is to enhance the particulate collection capacity of the dust gauge (Vallack and Shillito, 1998) by better collecting and retaining wet (from fog, dew, rain) and dry, fine and coarse particles, under stable meteorological conditions, during severe dust events, northwesterly *Shamal* winds, and by daily coastal winds,

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For the period December 2014 to March 2015, four Frisbee samplers were located at the New Environmental Oasis (NEO) site, about 50 m apart. The gravimetric information from the four samplers were similar, with small variations amongst them ascribed to the impact from local construction activities. Due to the similarity of these gravimetric results, and to obtain a better representation of dust deposition onto the KAUST campus, two of the samplers (DT1 and DT2) were moved in March, the first (DT1) to a residential area and the other (DT2) to the quay adjacent to the Coastal & Marine Resources Core Lab (CMOR) (Table 1). (Site meta-data provided in the Supplement A).

The deposition samples were collected for intervals of a calendar month, starting in December 2014 and ending December 2015. At the end of each month, the samples are retrieved by flushing the dust deposit with distilled water from the foam insert and collection dish into the downpipe and plastic bottle. Both the insoluble particles and dissolved salts in the water suspension are retrieved in the laboratory by a freeze-drying (sublimation) procedure.

A total of 52 deposition samples were collected at the six sampling sites on the KAUST campus (Fig. 1b) over a period of 13 months, largely in 2015. Representative subsets of these samples were selected for X-ray diffraction (XRD), (27 samples) and chemical analysis (29 samples).

Freeze-dried sample splits were re-suspended in the laboratory onto Teflon® filters, for elemental analysis by X-ray Fluorescence (XRF) spectrometry using a miniaturized version of a dust entrainment facility (Engelbrecht et al., 2016), <a href="http://www.dri.edu/atmospheric-sciences/atms-laboratories/4185-dust-entrainment-and-characterization-facility">http://www.dri.edu/atmospheric-sciences/atms-laboratories/4185-dust-entrainment-and-characterization-facility</a>). With this modified system the dust sample is drawn into a vertically mounted tubular dilution chamber, and the re-suspended dust collected onto a 47 mm diameter Teflon® filter, for chemical analysis.

The samples re-suspended onto the Teflon® filters were chemically analyzed for elemental content by XRF, including for Si, Ti, Al, Fe, Mn, Ca, K, P, V, Cr, Ni, Cu, Zn, Rb, Sr, Y, Zr, and Pb (US EPA, 1999). Splits of about 2 mg from each freeze-dried sample were analyzed for water soluble cations of sodium (Na<sup>+</sup>), potassium (K<sup>+</sup>), calcium (Ca<sup>2+</sup>) and magnesium (Mg<sup>2+</sup>), and anions of sulfate (SO<sub>4</sub><sup>2-</sup>), chloride (Cl<sup>-</sup>), phosphate (PO<sub>4</sub><sup>3-</sup>) and nitrate (NO<sub>3</sub><sup>-</sup>), by Ion Chromatography (IC) (Chow and Watson, 1999).

A subset of 27 samples from the total of 52 samples, representing all months of the year, was selected for X-ray diffraction (XRD) analysis. XRD is a non-destructive technique particularly suited to identify and characterize minerals such as quartz, feldspars, calcite, dolomite, clay minerals and iron oxides, in fine soil and dust. Dust reactivity in seawater as well as optical properties depend on its mineralogy, e.g.

carbonates and sulfates are generally more soluble in water than silicates such as feldspars, amphiboles, pyroxenes or quartz. A Bruker D8° X-ray powder diffraction system was used to analyze the mineral content of the dust deposition samples. The diffractometer was operated at 40 kV and 40 mA, with Cu Kα radiation, scanning over a range of 4-50° 2θ. The Bruker Topas° software and relative intensity ratios (RIRs) were applied for semi-quantitative XRD analyses of the dust deposition samples (Rietveld, 1969; Chung, 1974; Esteve et al., 1997; Caquineau et al., 1997; Sturges et al., 1989).

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A likely bias in the results from applying the X-ray diffraction (XRD) technique, together with the RIR method is widely recognized, and therefore our methodology is considered to be semi-quantitative at best. Chung (1974) recognized that if the RIRs of all the crystalline phases in a mineral mixture are known, the sum of all the fractions should add to 100%. However, XRD is effective at measuring crystalline phases such as quartz, calcite, and feldspars, and less so for partly crystalline and amorphous phases, including some layered silicates such as clays as well as many hydrous minerals. This could lead to an overestimation of the abundance of the crystalline mineral species in the dust, compared to partly crystalline and amorphous phases (Formenti et al., 2008; Kandler et al., 2009). Other discrepancies could occur from preferred orientation of layered silicates in the sample mounts. To minimize this effect, the dust samples were loaded into side-mount holders.

Electron microscopy provided information on the individual particle size and shape of micron-size particles, important for determining the optical parameters for modeling of dust (Moosmüller et al., 2012). The scanning electron microscope (SEM) based individual particle analysis was performed on a subset of twelve deposition samples collected for each month of 2015. For each sample, a portion of the deposition sample was suspended in isopropanol and dispersed by sonication. The suspension was vacuum filtered onto a 0.2 µm pore size polycarbonate substrate. A section of the substrate was mounted onto a metal SEM stub with colloidal graphite adhesive. The sample mounts were sputtercoated with carbon to dissipate the negative charge induced on the sample by the electron beam. The automated analysis was conducted on a Tescan MIRA 3° field emission scanning electron microscope (FE-SEM) by rastering the electron beam over the sample while monitoring the resultant combined backscattered electron (BE) and secondary electron (SE) signals. Based on the grayscale levels, preset threshold values segmented the image into particles of interest and background. The system was configured to automatically measure the size and shape of anywhere from 5,000 to 15,000 particles per sample measuring > 0.2 µm in average diameter. A digital image was acquired of each particle, for measurement, and stored for subsequent review. Size measurements were based on Feret diameters obtained from the projected area of each particle, by tracing their outer edges. This information was used to calculate the shape-dependent particle volumes. The particles were grouped into "bins" by their size. The field emission electron source allows for high magnifications and sharp secondary electron images (SEI), as well as for the detailed study of particle size distributions.

Particle size distribution plots of 12 deposition samples collected monthly at the KAUST campus throughout the 2015 period are shown by volume in Appendix A and by number in Supplement C. The chemical abundance tables are in Appendix B. The mineralogical results from XRD are described under paragraph 5.5 and the normative mineralogy calculated from the chemistry, presented as histogram plots in Figure 11.

#### 12. Results

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#### 12.1 Meteorology

Northwesterly *Shamal* winds prevailed during all twelve months of 2015 (Fig. 3). Four to five severe dust storms lasting three to five days each, contributed to hot humid conditions during the summer months. Weaker northeasterly winds were experienced in October and November of that year. Although the northeasterly winds were more frequent in November, they did not reach the maximum strength of the northwesterly winds.

The first four months in the second half of 2015 experienced the highest ambient temperatures (Table 2), with an average temperature of 35° C for August followed by 34° C for September, bracketed by 33° C for both July and October. The highest single temperature was 43° C, recorded in October, with the coolest temperature of 17° C in January of that year. The range of temperatures was the greatest through fall, winter and spring, with large diurnal temperature fluctuations during these seasons. The humidity at KAUST is consistently high (Table 2), with averages varying from 57 % for December and 61% for January, to as high as 82 % for August and 80% for September. Dewpoints were calculated for each set of hourly measurements, applying the August-Roche-Magnus approximation (Alduchov and Eskridge, 1996; August, 1828; Magnus, 1844). The highest dewpoint temperatures were calculated in August (31° C) and September (30° C) while the month with the greatest frequency of humidity measurements (96) in excess of 90 % was also recorded in August (Table 2, Fig. 4). The lowest monthly frequency (4) for humidity exceeding 90 % was December. In 2015, there were only a few light rainfall events at KAUST, and as such not of much importance to our measurements.

### 12.2 Gravimetric analysis

With a few exceptions, the monthly gravimetric measurements from the four samplers (DT1 – DT4) are comparable (Fig. 5), changing similarly by month and season. The deposition rates were at their lowest for December 2014 (avg. 4 g m<sup>-2</sup>), increasing steadily for four months to a peak value for March, 2015 (avg. 20 g m<sup>-2</sup>) before decreasing over the subsequent four months to a low for July (avg. 5 g m<sup>-2</sup>). The deposition rates increased sharply for August (28 g m<sup>-2</sup>), September (23 g m<sup>-2</sup>) and October (28 g m<sup>-2</sup>), before diminishing in November (14 g m<sup>-2</sup>) and December (11 g m<sup>-2</sup>). The NEO terrain is close to several building construction sites, about 400 m to the east and southeast of the installed deposition samplers, which periodically created substantial amounts of local airborne dust. This, together with the windy conditions are held responsible for elevated dust concentrations measured at the two NEO sites (DT3, DT4). The higher deposition rate at DT3 for August, compared to DT4, is ascribed to the fact that the former sampler is about 100m closer to construction material handling activities during that month. Wind-blown sea spray during stormy conditions was responsible for elevated deposition levels of sea salt at the CMOR (DT2) quay-side site, for the months of September and October 2015.

35 Source apportionment is considered to be a following step in the Red Sea dust research program. As an approximation of source contributions, the sampler with the lowest deposition rate can be considered to have negligible or contain the least amount of local dust and sea salt (Fig. 5). In the months of December 2014, January, April, March, June, July, and December 2015, the deposition rates at the four sampling sites were similar, and considered to have similar but negligible amounts of dust from local construction, roads, marine salt, or other particulates. In August, it is estimated that 24-56 g m<sup>-2</sup> month<sup>-1</sup>

(10-35%) of the dust captured at sites DT3 and DT4 was from local construction and motor traffic. Similarly, it is estimated that the deposition rates of sea salt at DT2 varied from 20-21 g m<sup>-2</sup> month<sup>-1</sup> (51-56%) over the months of September and October.

Bearing in mind that the dust deposition samplers, sampling procedures, as well as conditions and sampling periods were different to those of this study, some comparisons to similar studies in desert regions are listed in Table 3. The deposition rates from this study, both on average (14 g m<sup>-2</sup> month<sup>-1</sup>) and in range (4-28 g m<sup>-2</sup> month<sup>-1</sup>), were found to be similar to those previously recorded by Offer and Goossens (2001) in the Negev Desert, Israel (average 17 g m<sup>-2</sup> month<sup>-1</sup>, range 10-25 g m<sup>-2</sup> month<sup>-1</sup>), and West Niger (Goossens and Rajot, 2008) (average 13 g m<sup>-2</sup> month<sup>-1</sup>, range 6-21 g m<sup>-2</sup> month<sup>-1</sup>). A campaign in the Saudi Arabian capital of Riyadh (Modaihsh, 1997; Modaihsha and Mahjoub, 2013) during the dusty months of January to March showed average monthly deposition rates of 42 g m<sup>-2</sup>, and a range of 20-140 g m<sup>-2</sup>. The dust deposition measured in Kuwait on the other hand, varies substantially between sites due to the contribution from disturbed soils in lowlands during periods of north-westerly *Shamal* winds.

## 15 **12.3 AERONET** and visibility measurements

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The Aerosol Optical Depth (AOD) is one of the best observed aerosol characteristics. It defines the aerosol radiative effect and reflects the abundance of aerosols in the atmosphere. A CIMEL Robotic Sun Photometer is installed on the rooftop of the CMOR building on the campus of the KAUST and operated by our group since 2012, as a part of the NASA- AERONET, providing aerosol optical depth (AOD) and aerosol retrieved characteristics (https://aeronet.gsfc.nasa.gov/). Figure 6a compares the monthly averaged AOD at 500 nm with the dust deposition rate for 2015. In a general sense the AOD and the deposition rates show comparable trends, both with maxima in spring and larger maxima in fall. However, the AOD reaches a first maximum in April, being one month later than that of the deposition rate. Also, the larger second AOD maximum occurred in August while the maximum deposition rate is broadly distributed over a three-month period, from August to October, with AOD in October being relatively low. The linear correlation coefficient between the monthly deposition rates and the monthly averaged AOD of 0.40 suggests a causal interrelationship between these two quantities. However, for a number of reasons it is relatively low. The photometer measures light attenuation by all aerosols along a column in the atmosphere, while deposition rate depends on dust at ground level only, the latter generally containing a relatively coarser dust fraction. The low-level dust particles are partly from local dust sources while the higher altitude dust could be transported from distal sources and chemically transformed, i.e., aged. As was pointed out by Yu et al. (2013) the differences between the deposition and AOD time series can in part be attributed to modifications of the natural dust aerosol by anthropogenic activities, including petrochemical and other large industries along the Red Sea coast, as well as by entrainment of construction and road dust. However, the substantial contamination of dust by anthropogenic species and sea salt is not likely in this area, as was suggested by both observational (Osipov et al., 2015; Brindley et al., 2015) and modeling studies (Kalenderski and Stenchikov, 2016). To further test whether the non-dust fine aerosols (or remotely transported fine dust) significantly contribute to the interrelation between AOD and deposition rates, we followed two additional approaches. We screened the AODs by low Angström Exponent value (<0.3), suggested to correspond to dust (Ginoux et., 2012), and considered the contribution to the total AOD from the Spectral De-Convolution Algorithm (SDA) fine mode aerosol product. Neither technique contributed to an increase

of correlation coefficient. The discrepancy of high AOD – low deposition rate in summer (low Ångström Exponent), and low AOD – high deposition rate in October still remained when AODs were screened. The fine mode AOD remains almost constant throughout the year, and its contribution cannot explain the discrepancy.

Furthermore, a comparison between the deposition samples and the visibility is made with measurements taken in 2015 at the Jeddah airport meteorological station, approximately 70 km to the south of KAUST. Visibility is expressed as the frequency of dust events with reported weather codes 06-09, or 30–35, grouped as dusty or non-dusty days, for each month (Notaro et al., 2013; Anisimov et al., 2017), expressed as percentages. The bimodal monthly distributions seen with the deposition rates and AERONET monitoring are also mirrored by the visibility measurements collected at Jeddah (Fig. 6b). The linear correlation coefficient between the monthly deposition rates and monthly averaged visibility measurements is 0.48, clearly suggesting a causal relationship between the two variables.

#### **12.4** Particle size distributions (PSD)

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Dust deposition rates depend on the meteorological conditions, and dust properties such as particle size distribution, their vertical distribution, and abundance.

Summary plots of results from SEM based individual particle analysis for each month of 2015, expressed by number are presented in the Supplement C to this paper. From these particle size and shape measurements, equivalent shape-dependent volumes for the particles were calculated, the summary plots of which are shown in Appendix A. The volume of each particle is calculated from the measured maximum and minimum diameters, and assuming a prolate spheroid. Also, assuming a similar average density of e.g. 2.65 g cm<sup>-3</sup> for all minerals in the deposition samples, results in similar volume and mass distributions patterns. This was confirmed by XRD measurements and the abundance of quartz (2.65 g cm<sup>-1</sup>), feldspar (~ 2.65 g cm<sup>-1</sup>), micas (~ 2.83 g cm<sup>-1</sup>), and clays (~ 2.7–2.8 g cm<sup>-1</sup>) found in the deposition samples. The volume distributions were applied to assess the mass percentages and deposition rates of each size bin, e.g. the mass percentages and mass deposition rates of particles in bins less <10 µm in average diameter, and similarly less than <2.5 μm in average diameter, together with their uncertainties (Table 4). The contribution of particles <10 um to the total measured mass varies between about 4 and 17 % with an average of 8.6 % for the twelve months. Particles less than 2.5 µm range from about 0.6 to 4 %, with an average of 1.2 % for the twelve-month period. From these percentages and the total deposition rates, average deposition rates of 1.2±0.7 g m<sup>-2</sup> month<sup>-1</sup> for <10 μm and 0.1±0.1 g m<sup>-2</sup> month<sup>-1</sup> for  $<2.5 \mu m$  are estimated.

The average size distribution of the twelve deposition samples (Fig. 8a) is compared to that of the thirteen surface soils (Fig. 8b) from potential dust source regions along the Red Sea coastal plain (Prakash et al., 2016). The deposition samples with an average diameter of 0.9  $\mu$ m are much finer than the 3.9  $\mu$ m average diameter of the <38  $\mu$ m sieved soils. In addition the Frisbee sampler is biased towards the sampling of the coarser particles, as previously documented (Bergametti and Forêt, 2014; Goossens, 2005).

#### 12.5 Mineral analysis by XRD

XRD analysis of the 27 samples (Fig. 7) show variable amounts of quartz (6–38 %, avg. 22 %) and feldspars (plagioclase, K-feldspar) (5-34 %, avg. 20 %), clays (10-18 %, avg. 13 %), micas (6-31 %, avg. 13

%), halite (1-53 %, avg. 7 %) with lesser amounts of gypsum (1-8 %, avg. 4 %), calcite (0-8 %, avg. 2 %), dolomite (0-7 %, avg. 3 %), hematite (0-8 %, avg. 3 %), and amphibole (and pyroxene) (0-4%, avg. 1 %).

From the XRD, four broad mineral assemblages are distinguished, the first and major assemblage is comprised of feldspars, clays and micas as well as hematite and gypsum, the second group is of quartz, the third of halite, and the fourth of calcite.

There is an increase in the halite concentrations at sites DT1-DT3, from about 2 % (DT1) in December 2014 to about 53 % (DT2) in July 2015 (Fig. 7). From August onwards there is an abrupt decrease in halite content to less than 5 %, except for samples collected at the DT2 (CMOR, quay-side) site alongside the ocean. There was a simultaneous increase in the proportion of quartz to a maximum of 38 % in April (DT3), and decreasing to less than 25 % at all sites after July, 2015. The silicate mineral group decreased systematically from about 72 % (DT1) in December 2014 to about 25 % (DT2) in July. Except for two samples from the DT3 site collected in September and October 2015, the dominant minerals after July, 2015 included the silicate assemblage, with concentrations of up to 80 %. The variation in the proportions of the four mineral assemblages, especially the halite, is ascribed to seasonal fluctuations in wind, humidity and precipitation, as well as the proximity of the sea to the sampling sites.

SEM based SEIs of individual dust particles show that the larger particles being composed of mineral aggregates and coatings on other mineral particles. Examples (Supplement D) include particles composed of coatings of clay minerals on quartz and feldspar; clusters of clay minerals, calcite, gypsum and halite; and of iron oxides and clay minerals. Similar coatings and aggregates in re-suspended soil samples are reported by Engelbrecht et al. (2016).

### 12.6 Chemistry (XRF and IC)

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As expected, the chemically analyzed deposition samples contain major amounts of  $SiO_2$  (Appendix A, Fig. 9a, b), varying between 12–53 % (avg. 31 %) in the sample subset, occurring as quartz, and together with  $Al_2O_3$ , (avg. 4 %) and CaO, (avg. 2.3 %) in plagioclase, and  $K_2O$  (avg. 0.6 %) in potassium feldspars.  $SiO_2$  together with  $Al_2O_3$ ,  $Fe_2O_3$ ,  $TiO_2$ , MnO, MgO, and some  $K_2O$  are also contained in the clays, micas and amphiboles, previously identified in these samples by optical microscopy and XRD. Lesser amounts of CaO are contained in gypsum and calcite, and together with MgO, in dolomite. The iron expressed here as  $Fe_2O_3$  can be contained in hematite ( $Fe_2O_3$ ), goethite FeO(OH) or in clay minerals such as illite, each with different solubility. It has been suggested that large fractions of iron in soils and dusts are contained as amorphous colloidal coatings on quartz and feldspars (Engelbrecht et al., 2016).

The water-soluble cations (Appendix A, Fig. 10a, b) account for 1-19 % and the anions for 1-30 % of the total mass, respectively. These account for variable amounts of halite (1-32 %), and gypsum (1-9 %), with lesser amounts of other chlorides and carbonates. Of importance as dust borne nutrients likely to be deposited in the Red Sea, are the low concentrations of both water soluble  $NO_3^-$  (avg. 0.8 %), and water soluble  $PO_4^{3-}$  (avg. 0.2 %) compared to the total  $P_2O_5$  (avg. 0.3 %) in the dust deposits. The phosphorus is contained in the largely insoluble mineral apatite (francolite), found in the sedimentary rocks underlying large parts of the Arabian Peninsula (Notholt et al., 2005).

The sum of chemical species, including elements expressed as oxides, and ion concentrations, vary from 35–78 %, with an average of 56 % of the measured chemical mass. The shortfall from 100 % is

attributed in part to components not analyzed for, including  $H_2O$ , OH, carbon ( $CO_3^{2-}$ , organic carbon, elemental carbon) and artifacts of debris deposited onto the samplers.

The chemical abundances were recalculated as normative minerals (Fig. 11a, b), comparable in composition to those identified by XRD (Fig. 7) and optical microscopy. The relative normative mineral abundances (Fig. 11b) show variable amounts of quartz (avg. 52.4 %) feldspar (avg. 3.9 %), kaolinite (2.6 %), calcite (8.8 %) dolomite (0.2 %), hematite (8.0 %), as well as the evaporate minerals gypsum (12.1 %), halite (12.1 %), sylvite (0.2 %), and bischofite (0.2 %). There is also, as shown by XRD, an increase in halite content from about 7.8 % in January to about 25.9 % in July, followed by a sharp drop to about 4.6 % in August, with greater abundances at the CMOR quayside site in September (51.0 %) and October (31.6 %), ascribed to sea spray from stormy conditions during those two months.

Elemental mass ratios of the Frisbee deposition samples are compared to the <38 μm sieved soil samples from the Arabian Red Sea coastal plain (Prakash et al., 2016), and total suspended particulate (TSP) samples collected at other sites in the Middle East (Engelbrecht et al., 2009a) are compared in Table 5. The average Si/Al ratio of 6.86 of the Frisbee deposition samplers is intermediate to the 13.60 of the Arabian Red Sea coastal soils and the approximately unity of the Middle East samples. The Fe/Al ratios of the sample sets show similar relationships as the Si/Al ratios, being intermediate to the Red Sea coastal soils and four of the five other Middle East countries, excluding UAE to which it is similar. The difference is ascribed to the greater abundance of the minerals such as quartz in the coarser sieved soil samples, and less thereof in the finer TSP fractions. The Ca/Al ratio of 2.17 is similar to those of TSP samples from samples of Qatar (2.07) and UAE (2.16), ascribed to the regional carbonate-bearing soils in all three countries. The average Ti/Al, Mg/Al and K/Al ratios of the Frisbee deposition samples are substantially lower than those of the Red Sea coastal soils, which may be related to mineralogical differences in the dust source regions. Differences can also be ascribed to larger percentages of Albearing minerals such as clays in the deposition samples from this study.

#### 25 13. Summary and conclusions

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This study provides new mineralogical, physical and chemical information on deposition samples collected at the KAUST campus during 2015, as well as an assessment of the seasonal variability of the regional dust deposition rates onto Saudi Arabian coastal plain.

Inverted Frisbee samplers with foam inserts are found to be robust, easy to use, and provided comparable results, for the collection of wet and dry deposits. Once a month the samples are retrieved by flushing the deposits into plastic flasks followed by freeze-drying of the slurry and recovery of all suspended particles and dissolved salts. The average deposition rate at KAUST for 2015 was 14 g m<sup>-2</sup> varying from 4 g m<sup>-2</sup> in December, to 20 g m<sup>-2</sup> in March, 5 g m<sup>-2</sup> in July, 28 g m<sup>-2</sup> in September and October, and down to 11 g m<sup>-2</sup> the following December. The changes are ascribed to seasonally variable meteorological conditions, including high humidity prevailing along the Arabian Red Sea coastal plain during the late summer and autumn months. The particle size distributions provide an assessment of <10  $\mu$ m and <2.5  $\mu$ m dust deposition rates, the former varying 0.6-2.8 g m<sup>-2</sup> and the latter 0.06-0.29 g m<sup>-2</sup>.per month We suggest these deposition rates as proxies for those of PM<sub>10</sub> (coarse) and PM<sub>2.5</sub> (fine), respectively.

40 Chemical analysis, confirmed by XRD, point to a consistent silicate mineral fractions for the deposition samples, at all sampling sites for the entire sampling period. The Si/Al, Fe/Al, and Ca/Al ratios of the

deposition samples fall within the range of the soil samples previously collected along the Arabian Red Sea coastal plain as well as the TSP size fractions collected at several sites in the Middle East. It is proposed that the dust deposits along the Red Sea coast are a mixture of dust emissions from local soils, and soils imported from distal dust sources. Airborne mineral concentrations are greatest at or close to dust sources, compared to dusts, due to medium and long-range transport.

For 2015, there are marked similarities between monthly distribution patterns of the deposition samples and AOD measured at KAUST, as well as visibility measurements from Jeddah airport, 70 km to the south. This shows that both the AOD and visibility measurements mirror fluctuations in dust deposition, although it may not be justified to calculate quantitative interrelationships without further research.

Except for the variable halite fractions and local construction dust, there are small variations in the mineralogical content of the dust samples collected on the KAUST campus. To better model the dust being deposited in Red Sea, and coastal plain, the sampling campaign should be extended to sites beyond the KAUST campus. Such a sampling site was recently set up on an island off the coast from KAUST. Inclusion of particle size with mineralogical and chemical measurements provide more effective data for the modeling community.

The deposition samplers collect all particle sizes, however bin aerosol models usually consider only  $PM_{10}$ . The estimated  $PM_{10}$  deposition rates are lower than the total particulate deposition rates we observed. However, the size distribution of deposited particles shown in Figure 8a and Appendix A could be used to assess the contribution of  $PM_{10}$  in deposited mass, and reconcile models with observations. Alternatively, the calculated particle size range in the models can be potentially be extended to cover TSP. However, this could be computationally expansive.

As an approximation of source contributions, the sampler with the lowest deposition rate can be considered to have negligible or the least amount of local dust or sea salt (Fig.5). In the months of December 2014, January, April, March, June, July, and December 2015, the deposition rates at the four sites were similar, and considered to have no or negligible amounts of dust from local construction, campus roads, marine salt, or other particulates.

#### 14. Data availability

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The gravimetric, mineralogical and chemical data from this study are available upon request from Georgiy Stenchikov (Georgiy.Stenchikov@kaust.edu.sa).

Author Contributions. Johann Engelbrecht was responsible for the sample analysis and data compilation; Georgiy Stenchikov formulated the problem, designed the research project, and supported experimental activities; Jish Prakash collected, and conducted the freeze-drying of the samples, and performed part of the XRD analysis. Traci Lersch performed the SEM based individual particle analysis, Anatolii Anisimov assembled the meteorological and visibility data;
 Illia Shevchenko assembled the AERONET optical data; Engelbrecht, Stenchikov, Jish Prakash, Anisimov, and Shevchenko compiled different parts of the manuscript.

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## **FIGURES**

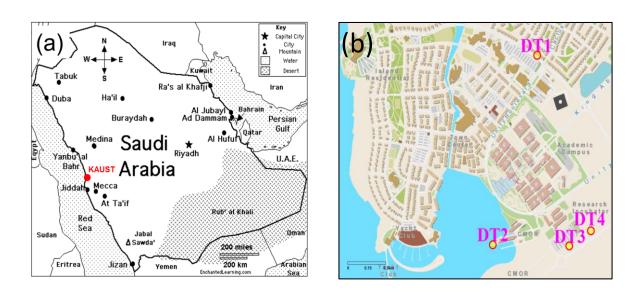


Figure 1. Position of (a) the King Abdullah University of Science and Technology (KAUST) campus on the Arabian Peninsula (red marker), north of the coastal city of Jeddah and (b) the Frisbee deposition sites (DT1-DT4) on the KAUST campus.

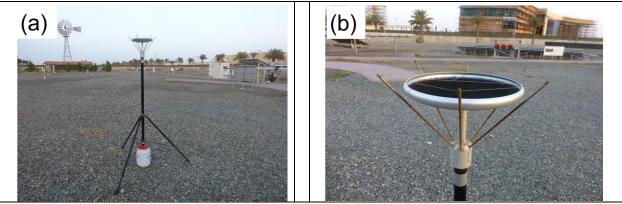


Figure 2. Inverted Frisbee type deposition sampler (a) on tripod and white plastic drainage bottle, (b) showing the foam insert in the collection dish to help retain the deposited dust particles, as well as the spikes with nylon thread to prevent birds from readily perching on the dish.

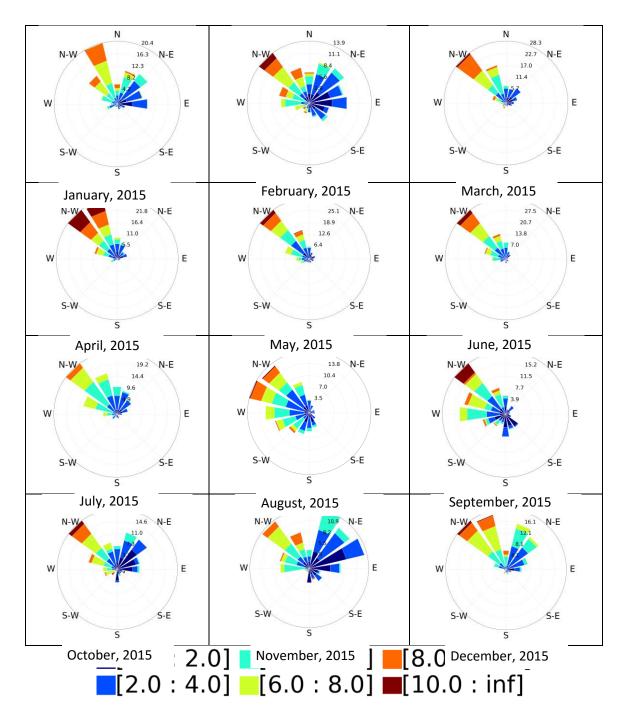


Figure 3. Wind (m s<sup>-1</sup>) roses for each month of 2015.

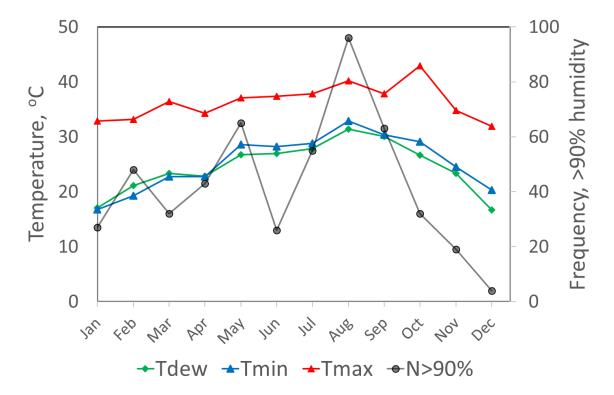


Figure 4. Monthly averaged minimum (———) and maximum (————) ambient temperatures as well as dewpoint (————) variations for KAUST during 2015. Also shown for each month is the frequency of hourly humidity measurements exceeding 90% (————).

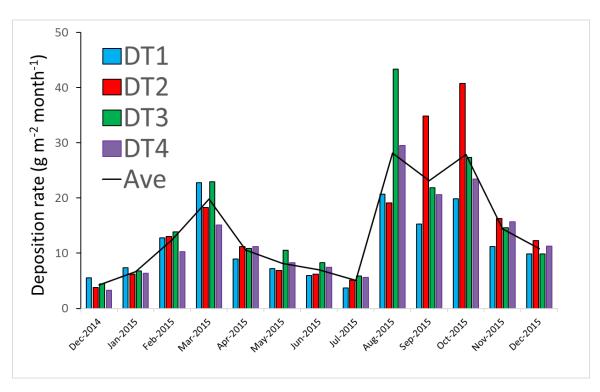


Figure 5. Monthly deposition rates (g m<sup>-2</sup>) from Frisbee samplers (DT1-DT4) at the KAUST campus. Also shown are the monthly averages for the four samplers.

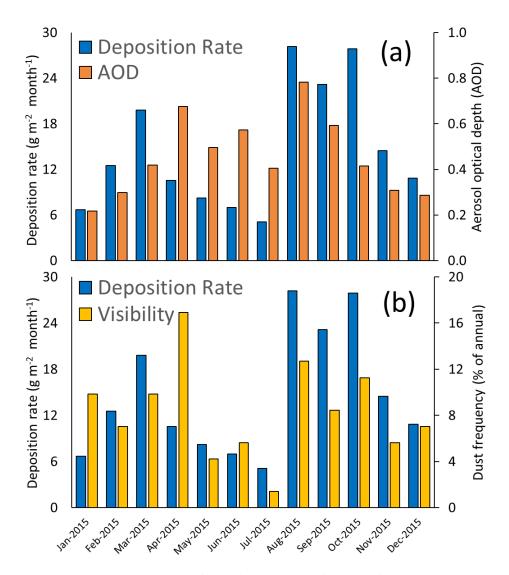


Figure 6. Average monthly deposition rates for all four samplers (DT1-DT4) on the KAUST campus, together with (a) monthly averaged AOD measurements from the KAUST AERONET site, and (b) monthly averaged visibility measurements collected from the Jeddah airport, for 2015.

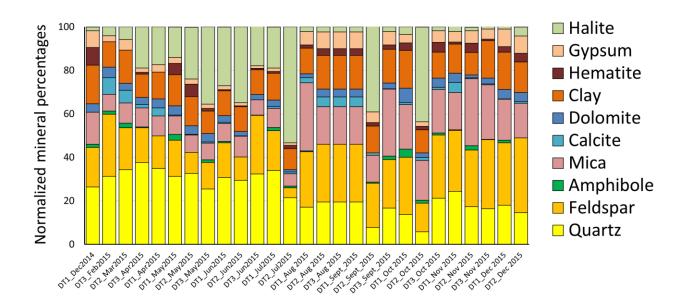


Figure 7. Semi-quantitative XRD mineral analyses of monthly Frisbee samples collected at the three sites DT1–DT3, for the period December, 2014 to December2015.

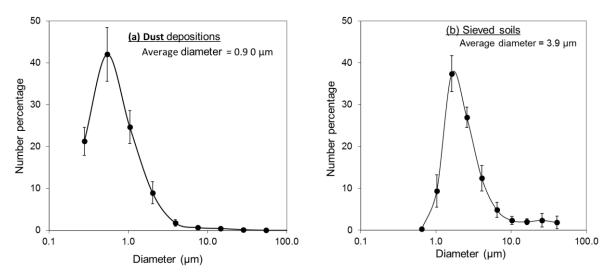


Figure 8. Average particle size distributions and standard deviations of (a) twelve deposition samples collected by Frisbee samplers on KAUST campus, and of (b) thirteen <38  $\mu$ m sieved soil samples from a previous study (Prakash et al., 2016), both measured by SEM.

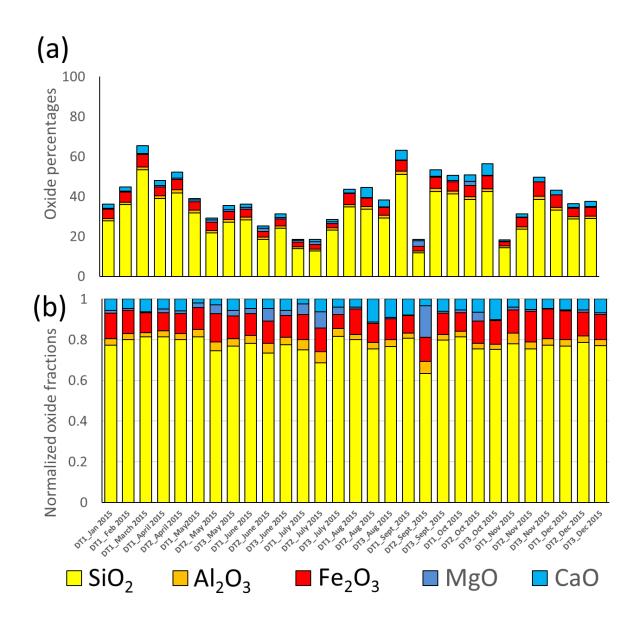


Figure 9 (a). Deposition sample elemental compositions, expressed as oxides and (b) fractions normalized to unity.

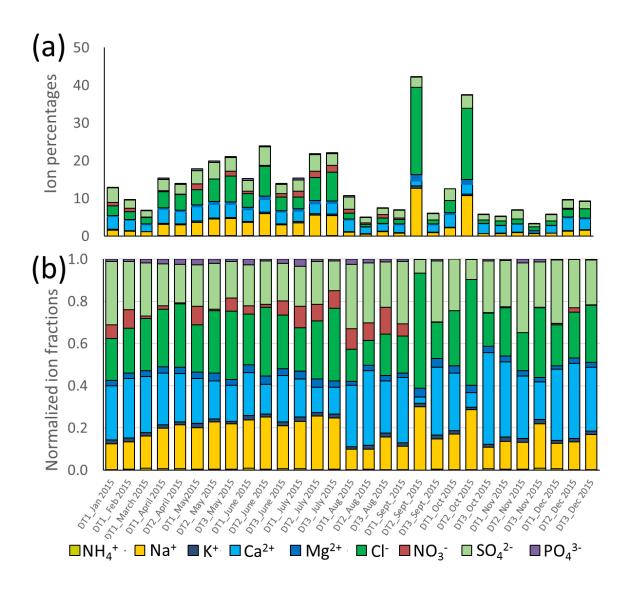


Figure 10 (a). Ion concentrations and (b) fractions totaled to unity.

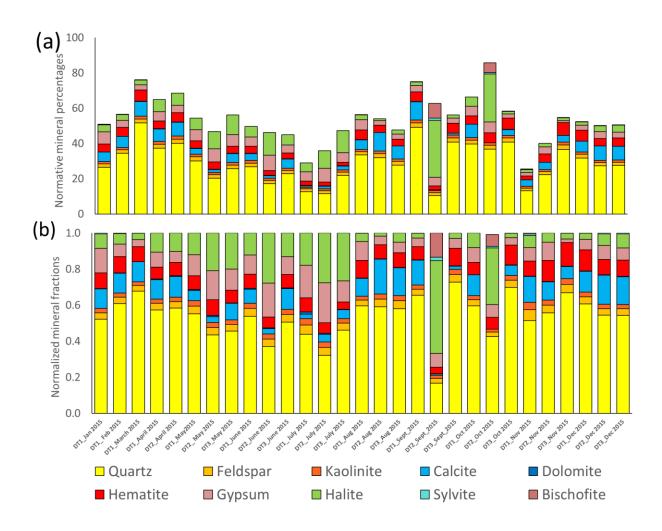


Figure 11 (a). Chemical abundances combined as normative minerals, (b) normalized to 100%.

## **TABLES**

Table 1. Locality of deposition samplers at six sites on the campus of KAUST.

|     | Site      | Latitude      | Longitude     | Elev.<br>m.a.s.l. | Start | End   |
|-----|-----------|---------------|---------------|-------------------|-------|-------|
| DT1 | NEO 1     | 22°18'16.12"N | 39°06'28.46"E | 1                 | Dec14 | Mar15 |
|     | Res G3705 | 22°18'59.06"N | 39°06'21.32"E | 12                | Apr15 | Dec15 |
| DT2 | NEO 2     | 22°18'16.84"N | 39° 6'29.33"E | 1                 | Dec14 | Mar15 |
|     | CMOR      | 22°18'16.60"N | 39° 6'7.91"E  | 1                 | Apr15 | Dec15 |
| DT3 | NEO 3     | 22°18'17.31"N | 39° 6'30.51"E | 1                 | Dec14 | Dec15 |
| DT4 | NEO 4     | 22°18'18.10"N | 39° 6'31.52"E | 1                 | Dec14 | Dec15 |

Table 2. Monthly averaged temperatures, humidity measurements, and calculated dewpoints at KAUST during 2015.

| Month |      | Temp | erature | <u>;</u> |      | Hu   | midity |       | Dewpoint |
|-------|------|------|---------|----------|------|------|--------|-------|----------|
|       | Avg. | Min. | Max.    | Range    | Avg. | Min. | Max.   | N>90% | Avg.     |
|       | °C   | °C   | °C      | °C       | %    | %    | %      | Count | °C       |
| Jan   | 25   | 17   | 33      | 16       | 61   | 10   | 99     | 27    | 17       |
| Feb   | 27   | 19   | 33      | 14       | 74   | 15   | 99     | 48    | 21       |
| Mar   | 28   | 23   | 36      | 14       | 76   | 29   | 99     | 32    | 23       |
| Apr   | 28   | 23   | 34      | 12       | 74   | 23   | 99     | 43    | 23       |
| May   | 32   | 29   | 37      | 9        | 77   | 21   | 99     | 65    | 27       |
| Jun   | 32   | 28   | 37      | 9        | 76   | 22   | 99     | 26    | 27       |
| Jul   | 33   | 29   | 38      | 9        | 75   | 26   | 99     | 55    | 28       |
| Aug   | 35   | 33   | 40      | 7        | 82   | 36   | 99     | 96    | 31       |
| Sep   | 34   | 30   | 38      | 8        | 80   | 26   | 99     | 63    | 30       |
| Oct   | 33   | 29   | 43      | 14       | 72   | 9    | 96     | 32    | 27       |
| Nov   | 30   | 25   | 35      | 10       | 69   | 25   | 99     | 19    | 23       |
| Dec   | 27   | 20   | 32      | 12       | 57   | 15   | 94     | 4     | 17       |

Table 3. Dust deposition measurements from the Middle East and other Global dust regions.

|     | Study                          | Locality             | Sampler type                        | Sampling period       | Average deposition rate (g m <sup>-2</sup> month <sup>-1</sup> ) | Range deposition rate (g m <sup>-2</sup> month <sup>-1</sup> ) |
|-----|--------------------------------|----------------------|-------------------------------------|-----------------------|------------------------------------------------------------------|----------------------------------------------------------------|
| (a) | This study (2017)              | Saudi Arabia, KAUST  | Frisbee with foam insert            | Dec 2014 - Dec 2015   | 14                                                               | 4 - 28                                                         |
| (b) | Modaihsh and Mahjoub (2013)    | Saudi Arabia, Riyadh | Dish with marbles                   | Jan - Mar ?           | 42                                                               | 20 - 140                                                       |
| (c) | Khalaf and Al-Hashash (1983)   | Kuwait, N-W Gulf     | Polyethelene cylinders with water   | Apr 1979 - Mar 1980   | 191                                                              | 10 - 1003                                                      |
| (d) | Al-Awadhi (2005)               | Kuwait, N-E Bay      | PVC bucket with marbles             | May 2002 - Apr 2003   | 28                                                               | 3 - 58                                                         |
| (e) | Al-Awadhi and AlShuaibi (2013) | Kuwait, City         | PVC bucket with marbles             | Mar 2011 - Feb 2012   | 53                                                               | 2 - 320                                                        |
| (f) | Offer and Goossens (2001)      | Israel, Negev        | Marble collectors                   | 1988 - 1997           | 17                                                               | 10 - 25                                                        |
| (g) | Goossens and Rajot (2008)      | Niger, Banizoumbou   | Frisbee with marbles, original data | 8 periods in 2005     | 13                                                               | 6 - 21                                                         |
| (h) | Smith and Twiss (1965)         | USA, Kansas          | Cylindrical rain gauge with screens | June 1963 - June 1964 | 6                                                                | 3 - 14                                                         |

Table 4. Monthly measured deposition rates, and assessments of <10  $\mu$ m and <2.5  $\mu$ m deposition rates from SEM based particle size measurements.

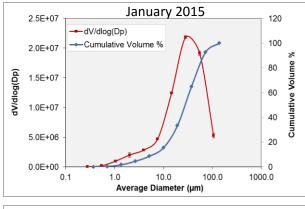
| Sample #     | Month (2015) |                                       | I             | Deposition Rate                       |               |                                       |
|--------------|--------------|---------------------------------------|---------------|---------------------------------------|---------------|---------------------------------------|
|              |              | Total                                 | < 1           | .0 μm                                 | < 2.          | .5 μm                                 |
|              |              | g m <sup>-2</sup> month <sup>-1</sup> | % of Total    | g m <sup>-2</sup> month <sup>-1</sup> | % of Total    | g m <sup>-2</sup> month <sup>-1</sup> |
| DT3.1_012015 | January      | 7.34                                  | 16.5 ± 4.3    | 1.2 ± 0.3                             | 4.0 ± 1.6     | 0.29 ± 0.12                           |
| DT3.1_022015 | February     | 12.83                                 | $7.0 \pm 3.3$ | $0.9 \pm 0.4$                         | $0.9 \pm 0.4$ | $0.12 \pm 0.05$                       |
| DT3.4_032015 | March        | 15.11                                 | 12.0 ± 5.8    | $1.8 \pm 0.9$                         | 1.6 ± 0.6     | $0.24 \pm 0.09$                       |
| DT3.4_042015 | April        | 11.22                                 | 8.7 ± 2.8     | $1.0 \pm 0.3$                         | $0.8 \pm 0.3$ | $0.09 \pm 0.03$                       |
| DT3.3_052015 | May          | 10.51                                 | $7.3 \pm 2.1$ | $0.8 \pm 0.2$                         | $0.8 \pm 0.3$ | $0.08 \pm 0.03$                       |
| DT3.3_062015 | June         | 8.28                                  | 9.0 ± 2.8     | $0.7 \pm 0.2$                         | $0.9 \pm 0.3$ | $0.07 \pm 0.02$                       |
| DT3.3_072015 | July         | 5.86                                  | 9.7 ± 6.3     | $0.6 \pm 0.4$                         | 1.1 ± 0.7     | $0.06 \pm 0.04$                       |
| DT3.3_082015 | August       | 43.39                                 | 6.4 ± 3.3     | $2.8 \pm 1.4$                         | $0.6 \pm 0.4$ | 0.26 ± 0.17                           |
| DT3.3_092015 | September    | 21.90                                 | $4.3 \pm 7.1$ | $0.9 \pm 1.6$                         | $0.6 \pm 1.0$ | $0.13 \pm 0.22$                       |
| DT3.3_102015 | October      | 27.39                                 | 9.2 ± 7.3     | 2.5 ± 2.0                             | $0.8 \pm 0.6$ | $0.22 \pm 0.16$                       |
| DT3.3_112015 | November     | 14.59                                 | 6.1 ± 1.4     | $0.9 \pm 0.2$                         | $0.7 \pm 0.2$ | $0.10 \pm 0.03$                       |
| DT3.3_122015 | December     | 9.91                                  | 7.3 ± 2.5     | $0.7 \pm 0.2$                         | 1.1 ± 0.2     | 0.11 ± 0.02                           |

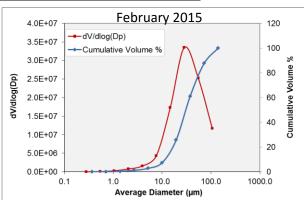
Table 5. Elemental mass ratios for the deposition samples from this study, compared to those of soils from the Red Sea coastal plain (Prakash et al., 2016) and TSP samples from other countries of the Middle East (Engelbrecht et al., 2009a). The TSP filter samples were collected by low-volume aerosol samplers without size selective inlets, for 24-hr sampling periods.

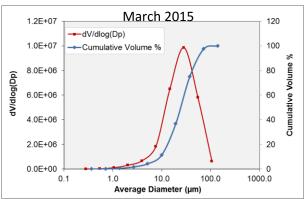
|             |              | Si/Al | Ti/Al | Fe/Al | Mg/Al | Ca/Al | K/Al |
|-------------|--------------|-------|-------|-------|-------|-------|------|
| Frisbee     | Deposition   | 6.86  | 0.14  | 1.47  | 0.11  | 2.17  | 0.34 |
| Saudi Soils | Sieved <38μm | 13.60 | 0.44  | 2.52  | 0.65  | 0.36  | 0.43 |
| Djibouti    | TSP          | 0.92  | 2.19  | 1.12  | 0.88  | 0.74  | 1.14 |
| Afghanistan | TSP          | 1.05  | 1.25  | 1.00  | 0.94  | 0.69  | 1.96 |
| Qatar       | TSP          | 1.02  | 0.24  | 0.98  | 1.40  | 2.07  | 0.93 |
| UAE         | TSP          | 1.29  | 0.28  | 1.52  | 2.85  | 2.16  | 1.02 |
| Iraq        | TSP          | 1.03  | 0.72  | 0.99  | 1.11  | 1.31  | 1.04 |
| Kuwait      | TSP          | 1.07  | 0.65  | 0.99  | 1.25  | 1.23  | 0.94 |

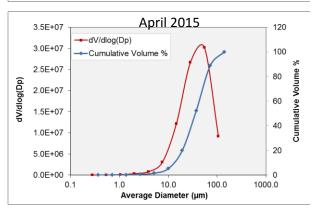
Appendix A

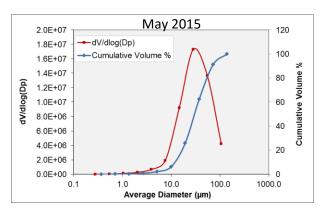
SEM based particle volume distribution curves for 12 months of 2015

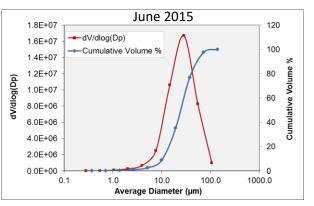




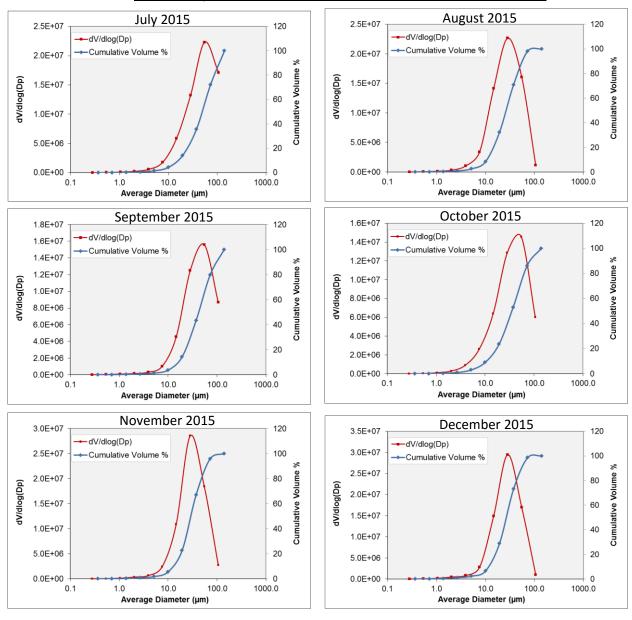








## Appendix A SEM based particle volume distribution curves for 12 months of 2015



Appendix B

| Sample                             | DT1_             | Jan | 2015  | DT1_            | Fel | 2015  | DT1_N           | lar | ch 2015 | DT1_/            | ۱pr | il 2015 | DT2_/            | ٩pr | il 2015 | DT1_             | Ma | y2015 | DT2_             | Ma       | y 2015 |
|------------------------------------|------------------|-----|-------|-----------------|-----|-------|-----------------|-----|---------|------------------|-----|---------|------------------|-----|---------|------------------|----|-------|------------------|----------|--------|
| Major and minor eleme              | ents as ox       | ide | s (%) |                 |     |       |                 |     |         |                  |     |         |                  |     |         |                  |    |       |                  |          |        |
| SiO <sub>2</sub>                   | 27.890           | ±   | 0.050 | 35.886          | ±   | 0.065 | 53.301          | ±   | 0.089   | 38.965           | ±   | 0.063   | 41.802           | ±   | 0.073   | 31.729           | ±  | 0.055 | 21.772           | ±        | 0.039  |
| TiO <sub>2</sub>                   | 0.466            | ±   | 0.001 | 0.530           | ±   | 0.001 | 0.679           | ±   | 0.002   | 0.464            | ±   | 0.002   | 0.599            | ±   | 0.002   | 0.486            | ±  | 0.002 | 0.451            | ±        | 0.003  |
| Al <sub>2</sub> O <sub>3</sub>     | 3.505            | ±   | 0.035 | 4.464           | ±   | 0.050 | 5.421           | ±   | 0.097   | 4.257            | ±   | 0.115   | 4.824            | ±   | 0.081   | 3.817            | ±  | 0.079 | 3.033            | ±        | 0.02   |
| Fe <sub>2</sub> O <sub>3</sub>     | 4.480            | ±   | 0.007 | 5.119           | ±   | 0.008 | 6.312           | ±   | 0.013   | 4.309            | ±   | 0.013   | 5.091            | ±   | 0.011   | 4.150            | ±  | 0.010 | 4.104            | ±        | 0.00   |
| MnO                                | 0.080            | ±   | 0.001 | 0.082           | ±   | 0.002 | 0.109           | ±   | 0.004   | 0.075            | ±   | 0.005   | 0.091            | ±   | 0.003   | 0.077            | ±  | 0.003 | 0.066            |          | _      |
| CaO*                               | 1.989            | ±   | 0.015 | 2.097           | ±   | 0.013 | 4.157           | ±   | 0.016   | 2.369            | ±   | 0.018   | 3.066            | ±   | 0.017   | 0.753            | ±  | 0.014 | 0.847            | ±        | 0.01   |
| K <sub>2</sub> O*                  | 0.484            | ±   | 0.005 | 0.604           | ±   | 0.004 | 0.864           | ±   | 0.003   | 0.705            | ±   | 0.006   | 0.798            | ±   | 0.005   | 0.542            | ±  | 0.008 | 0.432            | ±        | 0.00   |
| P <sub>2</sub> O <sub>5</sub>      | 0.417            | ±   | 0.001 | 0.150           | ±   | 0.001 | 0.010           | ±   | 0.003   | 0.332            | ±   | 0.004   | 0.306            | ±   | 0.002   | 0.474            | ±  | 0.003 | 0.429            | ±        | 0.00   |
| Total (oxides)                     | 39.311           |     |       | 48.932          |     |       | 70.852          |     |         | 51.475           |     |         | 56.579           |     |         | 42.028           |    |       | 31.134           |          |        |
| Trace elements (ppm)               |                  |     |       |                 |     |       |                 |     |         |                  |     |         |                  |     |         |                  |    |       |                  |          |        |
| V                                  | 107              | +   | 1     | 76              | ±   | 1     | 85              | ±   | 3       | 106              | ±   | 4       | 93               | ±   | 3       | 116              | ±  | 3     | 99               | <u>+</u> | 1      |
| Cr                                 | 92               |     |       | 115             |     | 5     | 113             |     | 12      | 101              |     | 15      | 108              |     | 10      | 113              |    | 10    | 120              | ±        | _      |
| Ni                                 | 78               |     | -     | 71              | ±   | -     | 84              |     | 6       | 68               |     |         |                  | ±   | -       | 77               |    | 5     | 57               |          | _      |
| Cu                                 | 81               |     |       | 57              |     | 4     | 134             |     | 10      | 125              |     | 13      | 99               |     | 9       | 206              |    | 9     | 64               | ±        | _      |
| Zn                                 | 223              |     |       | 247             | ±   | 8     | 293             | ±   | 18      | 287              | ±   | 23      | 258              | ±   | 15      | 467              | ±  | 16    | 219              | ±        | 4      |
| As                                 | 0                | ±   | 3     | 0               | ±   | 5     | 6               | ±   | 12      | 0                | ±   | 15      | 0                | ±   | 10      | 0                | ±  | 10    | 0                | ±        | 2      |
| Br                                 | 29               | ±   | 3     | 26              | ±   | 4     | 42              | ±   | 10      | 59               | ±   | 13      | 45               | ±   | 9       | 34               | ±  | 9     | 76               | ±        | 2      |
| Rb                                 | 28               | ±   | 1     | 24              | ±   | 1     | 16              | ±   | 3       | 28               | ±   | 4       | 34               | ±   | 3       | 30               | ±  | 3     | 27               | ±        | 1      |
| Sr                                 | 333              | ±   | 3     | 392             | ±   | 5     | 514             | ±   | 11      | 386              | ±   | 13      | 422              | ±   | 9       | 303              | ±  | 9     | 341              | ±        | 2      |
| Υ                                  | 1071             | ±   | 4     | 36              | ±   | 4     | 26              | ±   | 10      | 125              | ±   | 13      | 46               | ±   | 9       | 36               | ±  | 9     | 24               | ±        | 2      |
| Zr                                 | 103              | ±   | 4     | 98              | ±   | 6     | 84              | ±   | 14      | 57               | ±   | 17      | 139              | ±   | 12      | 133              | ±  | 12    | 134              | ±        | 3      |
| Мо                                 | 0                | ±   | 5     | 0               | ±   | 7     | 12              | ±   | 17      | 4                | ±   | 21      | 8                | ±   | 14      | 0                | ±  | 14    | 0                | ±        | 3      |
| Pb                                 | 15               | ±   | 5     | 28              | ±   | 8     | 32              | ±   | 18      | 66               | ±   | 23      | 28               | ±   | 15      | 19               | ±  | 15    | 28               | ±        | 3      |
| Water soluble ions (%)             |                  |     |       |                 |     |       |                 |     |         |                  |     |         |                  |     |         |                  |    |       |                  |          |        |
| NH <sub>4</sub> <sup>+</sup>       | 0.027            | ±   | 0.003 | 0.033           | ±   | 0.004 | 0.055           | ±   | 0.006   | 0.105            | ±   | 0.012   | 0.095            | ±   | 0.011   | 0.076            | ±  | 0.009 | 0.079            | ±        | 0.00   |
| Na <sup>†</sup>                    | 1.585            | ±   | 0.012 | 1.267           | ±   | 0.010 | 1.053           | ±   | 0.009   | 2.952            | ±   | 0.021   | 2.897            | ±   | 0.021   | 3.519            | ±  | 0.025 | 4.473            | ±        | 0.03   |
| K <sup>+</sup>                     | 0.230            | ±   | 0.009 | 0.170           | ±   | 0.006 | 0.114           | ±   | 0.004   | 0.262            | ±   | 0.010   | 0.207            | ±   | 0.008   | 0.371            | ±  | 0.014 | 0.296            | ±        | 0.01   |
| Mg <sup>2+</sup>                   | 0.338            | ±   | 0.005 | 0.265           | ±   | 0.004 | 0.188           | ±   | 0.003   |                  |     | 0.007   | 0.400            | ±   | 0.005   |                  |    | 0.008 | 0.744            | ±        | 0.01   |
| Ca <sup>2+</sup>                   |                  |     | 0.028 |                 |     | 0.023 |                 |     | 0.015   |                  |     | 0.032   |                  |     | 0.027   |                  |    | 0.032 | 3.560            |          |        |
| Cl <sup>-</sup>                    |                  |     | 0.014 |                 |     | 0.011 |                 |     | 0.009   |                  |     | 0.022   |                  |     | 0.023   |                  |    | 0.021 | 5.927            |          |        |
| NO <sub>3</sub>                    |                  |     | 0.030 |                 |     | 0.011 |                 |     | 0.003   |                  |     | 0.010   |                  |     | 0.004   |                  |    | 0.057 | 0.107            |          |        |
| SO <sub>4</sub> <sup>2-</sup>      |                  |     | -     |                 |     |       |                 |     |         |                  |     |         |                  |     |         |                  |    |       |                  |          |        |
|                                    |                  | ±   | 0.034 |                 | ±   | 0.019 |                 | ±   | 0.015   |                  | ±   | 0.027   |                  | ±   | 0.022   |                  | ±  | 0.031 | 4.327            | ±        | 0.03   |
| Total (ions) Total (oxides + ions) | 12.761<br>52.072 |     |       | 9.548<br>58.479 |     |       | 6.719<br>77.571 |     |         | 15.052<br>66.527 |     |         | 13.647<br>70.226 |     |         | 17.372<br>59.400 |    |       | 19.513<br>50.647 |          |        |

Appendix B

| Sample                         |           | _   | y 2015 | DT1_J  | lun | e 2015 | DT2_   | un | e 2015 | DT3_J  | un | e 2015 | DT1_   | July | y 2015 | DT2_   | Jul | y 2015 | DT3_   | Jul | y 2015 |
|--------------------------------|-----------|-----|--------|--------|-----|--------|--------|----|--------|--------|----|--------|--------|------|--------|--------|-----|--------|--------|-----|--------|
| Major and minor eleme          | nts as ox | ide | s (%)  |        |     |        |        |    |        |        |    |        |        |      |        |        |     |        |        |     |        |
| SiO <sub>2</sub>               | 27.167    | ±   | 0.051  | 28.318 | ±   | 0.053  | 18.530 | ±  | 0.035  | 24.158 | ±  | 0.045  | 13.820 | ±    | 0.027  | 12.693 | ±   | 0.025  | 23.221 | ±   | 0.037  |
| TiO <sub>2</sub>               | 0.448     | ±   | 0.001  | 0.478  | ±   | 0.002  | 0.306  | ±  | 0.001  | 0.370  | ±  | 0.001  | 0.261  | ±    | 0.001  | 0.241  | ±   | 0.001  | 0.200  | ±   | 0.002  |
| Al <sub>2</sub> O <sub>3</sub> | 3.777     | ±   | 0.059  | 3.692  | ±   | 0.062  | 2.919  | ±  | 0.050  | 3.067  | ±  | 0.045  | 1.886  | ±    | 0.041  | 2.030  | ±   | 0.037  | 2.694  | ±   | 0.148  |
| Fe <sub>2</sub> O <sub>3</sub> | 3.972     | ±   | 0.008  | 3.906  | ±   | 0.008  | 2.782  | ±  | 0.006  | 3.346  | ±  | 0.006  | 2.287  | ±    | 0.005  | 2.154  | ±   | 0.005  | 1.935  | ±   | 0.014  |
| MnO                            | 0.083     | ±   | 0.002  | 0.077  | ±   | 0.002  | 0.050  | ±  | 0.002  | 0.068  | ±  | 0.002  | 0.050  | ±    | 0.002  | 0.044  | ±   | 0.002  | 0.050  | ±   | 0.007  |
| CaO*                           | 1.983     | ±   | 0.016  | 1.681  | ±   | 0.014  | 1.179  | ±  | 0.013  | 1.745  | ±  | 0.014  | 0.427  | ±    | 0.010  | 1.179  | ±   | 0.011  | 1.148  | ±   | 0.013  |
| K <sub>2</sub> O*              | 0.508     | ±   | 0.006  | 0.513  | ±   | 0.007  | 0.366  | ±  | 0.008  | 0.467  | ±  | 0.006  | 0.208  | ±    | 0.008  | 0.226  | ±   | 0.008  | 0.359  | ±   | 0.009  |
| P <sub>2</sub> O <sub>5</sub>  | 0.170     | ±   | 0.002  | 0.590  | ±   | 0.002  | 0.383  | ±  | 0.002  | 0.389  | ±  | 0.001  | 0.753  | ±    | 0.002  | 0.457  | ±   | 0.001  | 0.288  | ±   | 0.005  |
| Total (oxides)                 | 38.108    |     |        | 39.254 |     |        | 26.514 |    |        | 33.609 |    |        | 19.692 |      |        | 19.024 |     |        | 29.895 |     |        |
| Trace elements (ppm)           |           |     |        |        |     |        |        |    |        |        |    |        |        |      |        |        |     |        |        |     |        |
| V                              | 88        |     |        | 111    | ±   | 2      | 86     | ±  | 2      | 91     | ±  | 1      | 115    | ±    | 1      | 111    | ±   | 1      | 77     | ±   | 5      |
| Cr                             | 71        |     |        | 96     | ±   | 7      | 65     |    |        | 82     | ±  |        | 160    | ±    | 5      | 60     |     |        | 67     | ±   | 21     |
| Ni                             | 62        | ±   | 3      | 84     | ±   | 4      | 49     | ±  | 3      | 72     | ±  | 3      | 133    | ±    | 3      | 71     | ±   | 2      | 91     | ±   | 11     |
| Cu                             | 56        | ±   | 6      | 134    | ±   | 7      | 59     | ±  | 5      | 49     |    | 5      | 209    | ±    |        | 62     | ±   |        | 25     | ±   | 19     |
| Zn                             | 251       |     | 11     | 430    |     | 12     | 297    | ±  | 9      | 255    | ±  | 8      | 515    | ±    | 9      | 244    | ±   | 7      | 180    | ±   | 32     |
| As                             | 0         |     |        | 0      |     |        | 0      |    | 6      | 0      |    | 5      |        | ±    |        | 0      |     |        | 0      |     | 21     |
| Br                             | 58        |     |        | 62     |     | _      | 100    | ±  | 5      | 64     | ±  | 5      | 62     | ±    | 5      | 88     | ±   | 4      | 37     | ±   | 19     |
| Rb                             | 24        |     |        | 35     |     |        | 16     |    |        | 23     | ±  |        | 17     | ±    | 1      | 12     | ±   | 1      | 8      | ±   |        |
| Sr                             | 392       |     |        | 322    |     | _      | 276    | ±  | 6      | 291    | ±  | 5      | 233    |      |        | 268    | ±   | 4      | 284    | ±   | 19     |
| Υ                              | 14        | ±   | 6      | 23     | ±   | 7      |        | ±  |        | 19     | ±  | 5      | 7      | ±    | 5      | 12     | ±   | 4      | 23     | ±   | 19     |
| Zr                             | 123       |     | 8      | 149    | ±   | 9      | 112    |    |        | 82     | ±  |        | 76     |      |        | 83     | ±   |        | 88     |     | 24     |
| Мо                             | 0         | ±   | 10     | 0      | ±   | 10     | 0      | ±  | 8      | 0      | ±  | 7      | 0      | ±    | 7      | 0      | ±   | 6      |        |     | 29     |
| Pb                             | 18        | ±   | 10     | 12     | ±   | 11     | 15     | ±  | 9      | 17     | ±  | 8      | 22     | ±    | 8      | 30     | ±   | 7      | 45     | ±   | 32     |
| Water soluble ions (%)         |           |     |        |        |     |        |        |    |        |        |    |        |        |      |        |        |     |        |        |     |        |
| NH <sub>4</sub> <sup>+</sup>   | 0.091     | ±   | 0.010  | 0.125  | ±   | 0.014  | 0.120  | ±  | 0.013  | 0.100  | ±  | 0.011  | 0.088  | ±    | 0.010  | 0.032  | ±   | 0.004  | 0.109  | ±   | 0.012  |
| Na <sup>+</sup>                | 4.509     | ±   | 0.031  | 3.505  | ±   | 0.025  | 5.875  | ±  | 0.041  | 2.834  | ±  | 0.020  | 3.466  | ±    | 0.024  | 5.538  | ±   | 0.038  | 5.360  | ±   | 0.037  |
| K <sup>+</sup>                 | 0.276     | ±   | 0.010  | 0.312  | ±   | 0.012  | 0.352  | ±  | 0.013  | 0.256  | ±  | 0.010  | 0.339  | ±    | 0.013  | 0.360  | ±   | 0.013  | 0.390  | ±   | 0.015  |
| Mg <sup>2+</sup>               | 0.572     | ±   | 0.008  | 0.559  | ±   | 0.008  | 0.947  | ±  | 0.013  | 0.453  | ±  | 0.006  | 0.582  | ±    | 0.008  | 0.878  | ±   | 0.012  | 0.628  | ±   | 0.009  |
| Ca <sup>2+</sup>               | 3.582     | ±   | 0.030  | 3.094  | ±   | 0.026  | 3.341  | ±  | 0.028  | 3.092  | ±  | 0.026  | 2.767  | ±    | 0.024  | 2.642  | ±   | 0.022  | 2.833  | ±   | 0.024  |
| Cl                             | 6.828     |     |        | 3.648  | ±   | 0.020  | 7.799  | ±  | 0.042  | 3.562  | ±  | 0.019  | 3.141  | ±    | 0.017  | 6.005  | ±   | 0.032  | 7.612  |     |        |
| NO <sub>3</sub>                | 1.328     | ±   | 0.048  | 0.595  | ±   | 0.022  | 0.349  | ±  | 0.013  | 0.939  | ±  | 0.034  | 1.573  | ±    | 0.057  | 1.702  | ±   | 0.061  | 1.864  | ±   | 0.067  |
| SO <sub>4</sub> <sup>2-</sup>  | 3.649     | ±   | 0.032  | 2.965  | ±   | 0.026  | 4.878  | ±  | 0.043  | 2.483  | ±  | 0.022  | 2.923  | ±    | 0.026  | 4.424  | ±   | 0.039  | 3.087  | ±   | 0.02   |
| Total (ions)                   | 20.836    |     |        | 14.804 |     |        | 23.663 |    |        | 13.719 |    |        | 14.879 |      |        | 21.580 |     |        | 21.882 |     |        |
| Total (oxides + ions)          | 58.944    |     |        | 54.058 |     |        | 50.177 |    |        | 47.328 |    |        | 34.571 |      |        | 40.605 |     |        | 51.777 |     |        |

Appendix B

| Sample                         | DT1_      | Aug | g <b>201</b> 5 | DT2_   | <u>Au</u> | g 2015 | DT3_   | Au | g 2015 | DT1_9  | ept | t_2015 | DT2_5  | ept | t_2015 | DT3_9  | Sep | t_2015 | DT1_   | Oct | 2015  |
|--------------------------------|-----------|-----|----------------|--------|-----------|--------|--------|----|--------|--------|-----|--------|--------|-----|--------|--------|-----|--------|--------|-----|-------|
| Major and minor eleme          | nts as ox | ide | s (%)          |        |           |        |        |    |        |        |     |        |        |     |        |        |     |        |        |     |       |
| SiO <sub>2</sub>               | 34.862    | ±   | 0.061          | 33.619 | ±         | 0.058  | 29.244 | ±  | 0.054  | 50.971 | ±   | 0.083  | 11.690 | ±   | 0.024  | 42.544 | ±   | 0.074  | 41.270 | ±   | 0.067 |
| TiO <sub>2</sub>               | 0.591     | ±   | 0.001          | 0.469  | ±         | 0.002  | 0.422  | ±  | 0.002  | 0.605  | ±   | 0.003  | 0.206  | ±   | 0.001  | 0.609  | ±   | 0.002  | 0.496  | ±   | 0.002 |
| Al <sub>2</sub> O <sub>3</sub> | 4.021     | ±   | 0.033          | 3.773  | ±         | 0.078  | 3.514  | ±  | 0.068  | 5.057  | ±   | 0.124  | 3.628  | ±   | 0.053  | 5.114  | ±   | 0.081  | 4.374  | ±   | 0.115 |
| Fe <sub>2</sub> O <sub>3</sub> | 5.488     | ±   | 0.008          | 4.186  | ±         | 0.010  | 3.878  | ±  | 0.009  | 5.479  | ±   | 0.015  | 2.206  | ±   | 0.006  | 5.540  | ±   | 0.011  | 4.592  | ±   | 0.013 |
| MnO                            |           |     | 0.001          |        |           | 0.003  | 0.068  |    | 0.003  |        |     | 0.005  |        |     | 0.002  |        |     | 0.003  | 0.081  |     | 0.005 |
| CaO*                           | 1.772     |     |                |        |           | 0.017  |        |    | 0.014  |        |     | 0.019  |        |     | 0.006  |        |     | 0.014  | 2.690  |     | 0.018 |
| K <sub>2</sub> O*              | 0.598     | ±   | 0.003          | 0.850  | ±         | 0.002  | 0.709  | ±  | 0.003  | 0.988  | ±   | 0.002  | 0.000  | ±   | 0.014  |        |     | 0.003  | 0.756  | ±   | 0.004 |
| P <sub>2</sub> O <sub>5</sub>  | 0.249     | ±   | 0.001          | 0.158  | ±         | 0.002  | 0.076  | ±  | 0.002  |        |     | 0.004  | 0.000  | ±   | 0.001  |        |     | 0.002  |        | ±   | 0.003 |
| Total (oxides)                 | 47.676    |     |                | 48.204 |           |        | 41.334 |    |        | 68.110 |     |        | 21.051 |     |        | 55.547 |     |        | 54.306 |     |       |
| Trace elements (ppm)           |           |     |                |        |           |        |        |    |        |        |     |        |        |     |        |        |     |        |        |     |       |
| V                              | 122       | ±   | 1              | 110    | ±         | 3      | 42     | ±  | 2      | 51     | ±   | 4      | 19     | ±   | 2      | 93     | ±   | 2      | 0      | ±   | 4     |
| Cr                             | 116       | ±   | 3              | 59     |           | 10     | 76     | ±  | 9      | 96     |     | 16     | 46     | ±   | 6      | 93     |     | 10     | 152    | ±   | 15    |
| Ni                             | 91        | ±   | 1              | 71     | ±         | 5      | 54     | ±  | 4      | 61     | ±   | 8      | 29     | ±   | 3      | 76     | ±   | 5      | 73     | ±   | 8     |
| Cu                             | 55        | ±   | 2              | 76     | ±         | 9      | 41     | ±  | 7      | 55     | ±   | 14     | 32     | ±   | 5      | 86     | ±   | 8      | 40     | ±   | 13    |
| Zn                             | 194       | ±   | 4              | 226    | ±         | 15     | 127    | ±  | 13     | 157    | ±   | 24     | 107    | ±   | 9      | 339    | ±   | 15     | 185    | ±   | 23    |
| As                             | 0         | ±   | 3              | 0      | ±         | 10     | 0      | ±  | 9      | 0      | ±   | 16     | 0      | ±   | 6      | 0      | ±   | 10     | 0      | ±   | 15    |
| Br                             | 20        | ±   | 2              | 16     | ±         |        | 34     |    | 7      | 17     | ±   | 14     | 715    | ±   | 7      | 58     |     |        | 64     |     | 13    |
| Rb                             | 22        | ±   | 1              | 43     | ±         |        | 12     |    | 2      | 47     |     | 4      | 18     |     |        | 38     |     | 2      | 24     | ±   | _     |
| Sr                             | 441       |     |                | 369    | ±         |        | 257    |    | _      | 394    | ±   | 14     | 179    |     |        | 345    | ±   |        | 331    | ±   | 13    |
| Υ                              | 18        |     | _              | 9      |           | 9      | 25     |    | 7      | 23     |     | 14     |        | ±   |        | 14     |     |        | 18     |     | 13    |
| Zr                             | 141       |     |                | 133    |           |        | 125    |    | 10     | 123    |     | 20     | 62     |     |        | 127    |     | 12     | 99     |     | 17    |
| Mo                             |           | ±   |                | 0      |           | 14     | 0      |    | 12     | 0      |     | 23     |        | ±   |        | 3      |     | 13     | 17     |     | 21    |
| Pb                             | 4         | ±   | 4              | 58     | ±         | 15     | 63     | ±  | 13     | 36     | ±   | 24     | 12     | ±   | 9      | 28     | ±   | 14     | 46     | ±   | 23    |
| Water soluble ions (%)         |           |     |                |        |           |        |        |    |        |        |     |        |        |     |        |        |     |        |        |     |       |
| NH <sub>4</sub> <sup>+</sup>   | 0.025     | ±   | 0.003          | 0.018  | ±         | 0.002  | 0.021  | ±  | 0.002  | 0.021  | ±   | 0.003  | 0.027  | ±   | 0.003  | 0.024  | ±   | 0.003  | 0.013  | ±   | 0.002 |
| Na <sup>+</sup>                | 1.044     | ±   | 0.009          | 0.481  | ±         | 0.006  | 1.144  | ±  | 0.009  | 0.756  | ±   | 0.007  | 12.685 | ±   | 0.087  | 0.862  | ±   | 0.008  | 2.134  | ±   | 0.016 |
| K <sup>+</sup>                 | 0.116     | ±   | 0.004          | 0.094  | ±         | 0.004  | 0.133  | ±  | 0.005  | 0.106  | ±   | 0.004  | 0.625  | ±   | 0.023  | 0.123  | ±   | 0.005  | 0.192  | ±   | 0.007 |
| Mg <sup>2+</sup>               | 0.213     | ±   | 0.003          | 0.133  | ±         | 0.002  | 0.193  | ±  | 0.003  | 0.146  | ±   | 0.002  | 1.732  | ±   | 0.024  | 0.261  | ±   | 0.004  | 0.425  | ±   | 0.006 |
| Ca <sup>2+</sup>               | 3.101     | ±   | 0.026          | 1.772  | ±         | 0.015  | 1.840  |    | 0.016  | 2.143  | ±   | 0.018  | 1.280  | ±   | 0.011  | 1.928  | ±   | 0.016  | 3.415  | ±   | 0.029 |
| Cl                             |           |     | 0.009          |        |           | 0.004  | 1.460  |    | 0.008  |        |     | 0.007  | 23.054 |     | 0.123  |        |     | 0.006  | 3.267  |     | 0.018 |
| NO <sub>3</sub>                | 1.045     |     |                |        |           | 0.015  | 0.943  |    | 0.034  |        |     | 0.015  |        |     | 0.003  |        |     | 0.003  | 0.009  |     | 0.003 |
| SO <sub>4</sub> <sup>2</sup>   | 3.241     | ±   | 0.028          | 1.425  | ±         | 0.012  | 1.591  | ±  | 0.014  | 2.047  | ±   | 0.018  | 2.778  | ±   | 0.024  | 1.743  | ±   | 0.015  | 3.042  | ±   | 0.027 |
| Total (ions)                   | 10.410    |     |                | 4.930  |           |        | 7.324  |    |        | 6.831  |     |        | 42.187 |     |        | 5.980  |     |        | 12.496 |     |       |
| Total (oxides + ions)          | 58.085    |     |                | 53.134 |           |        | 48.658 |    |        | 74.941 |     |        | 63.238 |     |        | 61.527 |     |        | 66.802 |     |       |

Appendix B

| Sample                         | DT2_O       | ct 2       | 2015  | DT3_   | 00 | t 2015 | DT1_   | Nov | 2015  | DT2_   | Nov | 2015  | DT3_   | No | v 2015 | DT1_   | De | 2015  | DT2_   | De | 2015  | DT3_   | De | c 2015 |
|--------------------------------|-------------|------------|-------|--------|----|--------|--------|-----|-------|--------|-----|-------|--------|----|--------|--------|----|-------|--------|----|-------|--------|----|--------|
| Major and minor eleme          | nts as oxic | les        | (%)   |        |    |        |        |     |       |        |     |       |        |    |        |        |    |       |        |    |       |        |    |        |
| SiO <sub>2</sub>               | 14.274      | ± (        | 0.028 | 38.421 | ±  | 0.068  | 42.440 | ±   | 0.076 | 23.596 | ±   | 0.044 | 38.454 | ±  | 0.070  | 33.248 | ±  | 0.059 | 28.716 | ±  | 0.053 | 28.860 | ±  | 0.052  |
| TiO <sub>2</sub>               | 0.206       | ± (        | 0.001 | 0.587  | ±  | 0.001  | 0.687  | ±   | 0.002 | 0.481  | ±   | 0.001 | 0.651  | ±  | 0.002  | 0.653  | ±  | 0.001 | 0.486  | ±  | 0.001 | 0.507  | ±  | 0.001  |
| Al <sub>2</sub> O <sub>3</sub> | 3.483       | <u>+</u> ( | 0.048 | 4.611  | ±  | 0.045  | 5.114  | ±   | 0.052 | 3.202  | ±   | 0.046 | 5.353  | ±  | 0.069  | 4.657  | ±  | 0.040 | 3.472  | ±  | 0.055 | 3.603  | ±  | 0.038  |
| Fe <sub>2</sub> O <sub>3</sub> | 2.118       | ± (        | 0.005 | 5.577  | ±  | 0.008  | 6.509  | ±   | 0.010 | 4.736  | ±   | 0.008 | 7.170  | ±  | 0.012  | 6.180  | ±  | 0.009 | 4.276  | ±  | 0.008 | 4.597  | ±  | 0.007  |
| MnO                            | 0.041       | <u>+</u> ( | 0.002 | 0.103  | ±  | 0.001  | 0.125  | ±   | 0.002 | 0.119  | ±   | 0.002 | 0.121  | ±  | 0.003  | 0.121  | ±  | 0.001 | 0.085  | ±  | 0.002 | 0.079  | ±  | 0.001  |
| CaO*                           | 5.742       | ± (        | 0.008 | 3.357  | ±  | 0.015  | 0.731  | ±   | 0.021 | 1.586  |     |       | 2.370  | ±  | 0.008  | 2.321  | ±  | 0.011 | 1.965  |    |       | 2.502  | ±  | 0.014  |
| K <sub>2</sub> O*              | 0.090       | ± (        | 0.010 | 0.774  | ±  | 0.002  | 0.822  | ±   | 0.002 | 0.552  | ±   | 0.003 | 0.954  | ±  | 0.002  | 0.751  | ±  | 0.002 | 0.590  | ±  | 0.003 | 0.592  | ±  | 0.003  |
| P <sub>2</sub> O <sub>5</sub>  | 0.000       | ± (        | 0.001 | 0.106  | ±  | 0.001  | 0.000  | ±   | 0.001 | 0.145  | ±   | 0.001 | 0.181  | ±  | 0.002  | 0.133  | ±  | 0.001 | 0.155  | ±  | 0.002 | 0.102  | ±  | 0.001  |
| Total (oxides)                 | 25.953      |            |       | 53.537 |    |        | 56.429 |     |       | 34.418 |     |       | 55.254 |    |        | 48.064 |    |       | 39.745 |    |       | 40.840 |    |        |
| Trace elements (ppm)           |             | +          |       |        |    |        |        |     |       |        |     |       |        |    |        |        |    |       |        |    |       |        |    |        |
| V                              | 67          | ±          | 1     | 125    | ±  | 1      | 130    | ±   | 1     | 149    | ±   | 2     | 180    | ±  | 2      | 144    | ±  | 1     | 97     | ±  | 2     | 65     | ±  | 1      |
| Cr                             | 26          | ± !        | 5     | 110    | ±  | 4      | 119    | ±   | 5     | 300    | ±   | 6     | 135    | ±  | 8      | 218    | ±  | 4     | 103    | ±  | 7     | 71     | ±  | 4      |
| Ni                             | 33          | ± :        | 3     | 84     | ±  | 2      | 68     | ±   | 2     | 971    | ±   | 3     | 121    | ±  | 4      | 211    | ±  | 2     | 134    | ±  | 3     | 64     | ±  | 2      |
| Cu                             | 28          | ± !        | 5     | 111    | ±  | 4      | 59     | ±   | 4     | 102    | ±   | 5     | 74     | ±  | 7      | 78     | ±  | 3     | 89     | ±  | 6     | 55     | ±  | 3      |
| Zn                             | 82          | ±          | 8     | 250    | ±  | 7      | 205    | ±   | 8     | 507    | ±   | 9     | 358    | ±  | 12     | 534    | ±  | 6     | 328    | ±  | 10    | 231    | ±  | 6      |
| As                             | 0 :         | ± !        | 5     | 0      | ±  | 4      | 0      | ±   | 5     | 0      | ±   | 5     |        |    | 8      | 0      | ±  | 3     | 0      | ±  | 7     |        | ±  |        |
| Br                             | 457         | ± !        | 5     | _      | ±  |        |        | ±   |       | 66     |     |       | 61     |    |        | 59     | ±  | 3     | 112    | ±  | 6     | 95     |    |        |
| Rb                             | 11 :        | -          |       |        | ±  |        |        |     |       | 26     |     |       | 34     |    |        | 27     |    |       |        | ±  |       | 22     |    |        |
| Sr                             | 227         |            |       | 422    |    |        | 683    |     |       | 253    |     |       | 210    |    |        | 329    |    |       | 266    |    |       | 310    |    |        |
| Υ                              | 14          |            |       |        | ±  |        | 14     |     |       | 15     |     |       | 39     |    |        | 23     |    |       | 13     |    |       | 19     |    |        |
| Zr                             | 38          |            |       | 134    |    |        | 143    |     |       | 103    |     |       | 192    |    |        | 139    |    |       | 113    |    |       | 95     |    |        |
| Мо                             | 0           |            |       |        |    | 6      |        | ±   |       |        | ±   |       |        |    | 10     |        | ±  |       |        | ±  |       |        | ±  |        |
| Pb                             | 8           | ±          | 8     | 36     | ±  | 6      | 17     | ±   | 7     | 20     | ±   | 8     | 24     | ±  | 11     | 22     | ±  | 5     | 30     | ±  | 10    | 40     | ±  | 6      |
| Water soluble ions (%)         |             |            |       |        |    |        |        |     |       |        |     |       |        |    |        |        |    |       |        |    |       |        |    |        |
| NH <sub>4</sub> <sup>+</sup>   | 0.047       | ± (        | 0.005 | 0.043  | ±  | 0.005  | 0.028  | ±   | 0.003 | 0.024  | ±   | 0.003 | 0.025  | ±  | 0.003  | 0.032  | ±  | 0.004 | 0.011  | ±  | 0.001 | 0.025  | ±  | 0.003  |
| Na <sup>†</sup>                | 10.668      | <u>+</u> ( | 0.073 | 0.577  | ±  | 0.006  | 0.675  | ±   | 0.007 | 0.890  | ±   | 0.008 | 0.684  | ±  | 0.007  | 0.699  | ±  | 0.007 | 1.279  | ±  | 0.010 | 1.540  | ±  | 0.012  |
| K <sup>+</sup>                 | 0.447       | <u>+</u> ( | 0.017 | 0.086  | ±  | 0.003  | 0.106  | ±   | 0.004 | 0.125  | ±   | 0.005 | 0.072  | ±  | 0.003  | 0.081  | ±  | 0.003 | 0.136  | ±  | 0.005 | 0.135  | ±  | 0.005  |
| Mg <sup>2+</sup>               | 1.315       | <u>+</u> ( | 0.018 | 0.164  | ±  | 0.002  | 0.143  | ±   | 0.002 | 0.185  | ±   | 0.003 | 0.066  | ±  | 0.001  | 0.095  | ±  | 0.001 | 0.232  | ±  | 0.003 | 0.225  | ±  | 0.003  |
| Ca <sup>2+</sup>               | 2.543       |            |       |        |    | 0.021  |        |     | 0.016 |        |     | 0.018 |        |    | 0.005  |        |    | 0.017 |        |    | 0.029 |        |    | 0.024  |
| Cl                             | 18.796      |            |       |        |    | 0.005  |        |     | 0.006 | 1.246  |     |       |        |    | 0.006  |        |    | 0.006 |        |    | 0.011 |        |    | 0.013  |
| NO <sub>3</sub>                | 0.009       |            |       |        |    | 0.003  |        |     | 0.003 | 0.004  |     |       |        |    | 0.003  |        |    | 0.003 |        |    | 0.008 |        |    | 0.003  |
| SO <sub>4</sub> <sup>2</sup> - | 3.581       | <u>+</u> ( | 0.031 | 1.411  | ±  | 0.012  | 1.133  | ±   | 0.010 | 2.297  | ±   | 0.020 | 0.697  | ±  | 0.006  | 1.734  | ±  | 0.015 | 2.188  | ±  | 0.019 | 1.964  | ±  | 0.017  |
| Total (ions)                   | 37.407      |            |       | 5.704  |    |        | 5.146  |     |       | 6.831  |     |       | 3.196  |    |        | 5.745  |    |       | 9.558  |    |       | 9.224  |    |        |
| Total (oxides + ions)          | 63.360      |            |       | 59.240 |    |        | 61.574 |     |       | 41.249 |     |       | 58.450 |    |        | 53.810 |    |       | 49.303 |    |       | 50.065 |    |        |