Deanship of Graduate Studies
Al-Quds University

Removal of Selected Pharmaceuticals from water using Natural Jordanian Zeolite

Mahran Abdallah Deeb D'ana

M.Sc. Thesis

Jerusalem – Palestine

1438 / 2017

Removal of Selected Pharmaceuticals from water using Natural Jordanian Zeolite

Prepared by:

Mahran Abdallah Deeb D'ana

B.Sc: Chemistry, Hebron University, Palestine

Supervisor: Dr. Fuad Al-Rimawi

Co-supervisor: Dr. Mohannad Qurie

A thesis Submitted in Partial fulfillment of requirement for the degree of the Master of Applied and Industrial Technology, Al-Quds University

1438 / 2017

Al-Quds University

Deanship of Graduate Studies

Applied and Industrial Technology Program

Removal of Selected Pharmaceuticals from water using Natural Jordanian Zeolite

Prepared by: Mahran Abdallah Deeb D'ana

Registration No: 21312529

Supervisor: Dr. Fuad Al- Rimawi

Co supervisor: Dr.Mohannad Qurie

Master thesis submitted and accepted, Date: 06/05/2017

The names and signatures of the examining committee members are as follows:

1- Head of Committee/Dr. Fuad Al-Rimawi Signature……..
2- Co supervisor/Dr.Mohannad Qurie Signature……..
3- Internal Examiner/ Dr.Ziyad Shakhshir Signature……...
4- External Examiner/ Prof.Shehdeh Jodeh Signature……..

Jerusalem-Palestine

1438 / 2017

Declaration

I certify that this thesis submitted for the degree of master is the result of my own research, except where otherwise acknowledges, and that this thesis (or any part of the same) has not been submitted for the higher degree to any other university or institute.

Signed……

Mahran Abdallah Deeb D'ana

Date: 6 / 5 / 2017

Acknowledgements

Praise and endless thanks to God for completion, and providing patience in accomplishing my thesis. I am very pleased to express my gratitude to all people who assisted me to go forward in my studies, and to all people who offered me help and support.

Special thanks for my supervisors Dr. Fuad Al-Rimawi, Dr.Mohannad Qreia and Mr. Sameh Nusseibeh for their continuous help and supervision during my research work.

Finally, I am glad to thank my family who supported and encouraged me during this period.

Abstract

In this work, Removal of selected pharmaceutical compounds (Ibuprofen, Diclofenac sodium, Indomethacin, Chlorophenarimine maleate and Paracetamol) from water using natural Jordanian zeolite was studied. The influence of pH of solution, contact time, adsorbent dosage, and initial pharmaceutical concentration on the adsorption process were investigated using batch experiment in addition to column experiments.

The optimal pH for the removal of all selected pharmaceuticals was found to be 2 except for Diclofenac sodium where the optimal pH was 6. The adsorption time for all experiments was found to be 80 minutes. The percentage removal increased as the initial concentration of the pharmaceuticals increased from 10.0 to 50.0 ppm except for Indomethacin where the removal decrease as the initial concentration increased. After optimization of the parameters for these pharmaceuticals removal, the highest removal was found to be 88.3%, 30.1%, 59.0%, 85.8% and 12.7% for Ibuprofen, Diclofenac sodium, Indomethacin, Chlorophenarimine maleate and Paracetamol respectively.

The Langmuir and Freundlich isotherm models were used to evaluate the adsorption of the pharmaceuticals on natural Jordanian zeolite. Results demonstrated that Langmuir isotherm fit the experimental data for Diclofenac sodium, Indomethacin and Paracetamol, with adsorption capacity (Q_{max}) of 4.8, 26.6, and 55.6 mg/g for Diclofenac sodium, Indomethacin and Paracetamol, respectively. Freundlich isotherm fit the experimental data for Ibuprofen & Chloropheniramin maleate with adsorption capacity of 1.2 mg/g and 2.1 mg/g for Ibuprofen and Chloropheniramin maleate, respectively.

Table of Contents

	Page No.
Dedication	
Declaration	iv
Acknowledgements	iv
Abstract	iv
Table of Contents	iv
List of Tables	iv
List of Figures	iv
List of Appendices	xiv
1. Introduction	2-12
1.1 Back ground	2-3
1.2 Pharmaceutical contaminants in water	3-7
1.3 Ibuprofen	4
1.4 Diclofenac Sodium	5
1.5 Indomethacin	6
1.6 Chloropheniramine Maleate	7
1.7 Paracetamol	7
1.8 Zeolite	8-10
1.9 Ion Exchange	10
1.10 Adsorption	11

2. Literature review 13-15

2.1 Problems statement 14

2.2 Hypotheses and research questions 15

2.3 Objectives and aims 15

3. Materials and Methods 16-19

3.1 Instrumentation 17

3.1.1 pH meter 17

3.1.2 Shaker 17

3.1.3 Electronic Balance 17

3.1.4 UV-Visible Spectrophotometer 17

3.2Chemicals and Reagents 17

3.3 Methods 17

3.3. 1 Calibration curves 17

3.3.2 Batch adsorption isotherms 18

3.3.2.1 Effect of pH 18

3.3.2.2Effect of contact time 18

3.3.2. 3Effect of adsorbent dosage 19

3.3.2.4Effect of initial absorbent concentration 19

3.3.3Column Experiment 19

4. Results and discussion 21-51

4.1 Batch Experiment 21

4.1.1 Acidic Pharmaceuticals 21

4.1.1.1 Ibuprofen 21-26

4.1.1.1.1Calibration curve for Ibuprofen using UV-visible spectrophotometer 21

4.1.1.1.2Effects of different parameters on adsorption of Ibuprofen 22-26

4.1.1.2 Diclofenac Sodium 26-32

4.1.1.2.1Calibration curve for Diclofenac Sodium using UV-visible spectrophotometer 26

4.1.1.2.2Effects of different parameters on adsorption of Diclofenac Sodium 27-32

4.1.1.3 Indomethacin 32-37

4.1.1.3.1Calibration curve for Indomethacin using UV-visible spectrophotometer 32

4.1.1.3.2Effects of different parameters on adsorption of Indomethacin 33-37

4.1.2 Basic pharmaceuticals 37

4.1.2.1 chlorpheniramine maleate 37-42

4.1.2.1.1Calibration curve for chlorpheniramine maleate using UV-visible spectrophotometer 40

4.1.2.1.2Effects of different parameters on adsorption of chlorpheniramine Maleate 37-42

4.1.2.2 Paracetamol 42-47

4.1.2.2.1 Calibration curve for Paracetamol using UV-visible spectrophotometer 42

4.1.2.2.2 Effects of different parameters on adsorption of pracetamol 42-47

4.1.3 Comparison between Percentage removal and optimum pH for the selected pharmaceuticals. 47

4.2 Column Experiments 48-51

Conclusion 53

References 54-58

Appendices 59-66

Abstract in Arabic 67

List of Tables

Table No.	Table Name	Page No.
Table.1	Average chemical composition (%) of the bulk sample in the borehole B.	10
Table.2	λ max for the selected pharmaceuticals	18
Table.3	Freundlich isotherm for the adsorption of Ibuprofen on Zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g).	25
Table.4	Langmiur isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 6, contact time= 80 minutes, adsorbent dosage = 1.0 g).	30
Table.5	Freundlich isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 6, contact time= 80 minutes, adsorbent dosage = 1.0g).	30
Table.6	Langmiur isotherm for the adsorption of Indomethacin on Zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 2, contact time= 60 minutes, adsorbent dosage = 1.0 g).	36
Table.7	Freundlich isotherm for the adsorption of Chlorophenarimine Maleate on Zeolite usingUV-visible spectroscopy at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g).	41
Table.8	Langmiur isotherm for the adsorption of Paracetamol on zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0 g).	46
Table.9	Comparison between the results, Percentage removal and optimum pH for the selected pharmaceuticals.	47

List of Figures

Figure No.	Figure Name	Page No.
Figure.1	Chemical structure of Ibuprofen.	5
Figure.2	Chemical structure of Diclofenac Soduim.	5
Figure.3	Chemical structure of Indomethacin.	6
Figure.4	Chemical structure of Chloropheniramine maleate.	7
Figure.5	Chemical structure of Paracetamol.	8
Figure.6	Chemical structure of zeolite.	8
Figure.7	Calibration curve of Ibuprofen obtained by UV-visible spectrophotometer at λ = 224nm.	21
Figure.8	Percentage removal of Ibuprofen on Zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 224nm).	22
Figure.9	Percentage removal of Ibuprofen on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 224nm).	23
Figure.10	Percentage removal of Ibuprofen on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 224nm).	23
Figure.11	Percentage removal of ibuprofen on zeolite as a function of initial Ibuprofen concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 224nm).	24
Figure.12	Freundlich isotherm for the adsorption of Ibuprofen on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g and λ= 224nm).	24
Figure.13	Calibration curve of Diclofenac sodium obtained by UV-visible spectrophotometer at λ= 276nm.	26

Figure.14	Percentage removal of Diclofenac sodium on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 276 nm).	27
Figure.15	Percentage removal of Diclofenac sodium on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 276 nm).	28
Figure.16	Percentage removal of Diclofinac sodium on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, contact time= 80 minutes and λ= 276 nm).	28
Figure.17	Percentage removal of Diclofenac sodium on Zeolite as a function of initial Diclofenac sodium concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 6, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 276 nm).	29
Figure.18	Langmiur isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 6, contact time= 80 minutes, adsorbent dosage = 1.0 g and λ= 276 nm).	29
Figure.19	Freundlich isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 6, contact time= 40 minutes, adsorbent dosage = 1.0g and λ= 276 nm).	30
Figure.20	Calibration curve of Indomethacin obtained by UV-visible spectrophotometer at λ= 225 nm.	32
Figure.21	Percentage removal of indomethacin on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).	33
Figure.22	Percentage removal of Indomethacin on zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).	34
Figure.23	Percentage removal of Indomethacin on zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 40 ppm, contact time= 60 minutes and λ= 225 nm).	34

Figure.24	Percentage removal of Indomethacin on zeolite as a function of initial Indomethacin concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 60 minutes and λ= 225 nm).	35
Figure.25	Langmiur isotherm for the adsorption of Indomethacin on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 60 minutes, adsorbent dosage = 1.0 g and λ= 225 nm).	35
Figure.26	Calibration curve of chlorpheniramine maleate obtained by UV-visible spectrophotometer at λ= 261 nm.	37
Figure.27	Percentage removal of chlorophenarimine maleate on Zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 261 nm).	38
Figure.28	Percentage removal of chlorophenarimine maleate on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 261 nm).	39
Figure.29	Percentage removal of Chlorophenarimine maleate on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 261 nm).	39
Figure.30	Percentage removal of Chlorophenarimine maleate on Zeolite as a function of initial Chlorophenarimine maleate concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.5 g, contact time= 80 minutes and λ= 261 nm).	40
Figure.31	Freundlich isotherm for the adsorption of Chlorophenarimine maleate on Zeolite usingUV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g and λ= 261 nm).	40
Figure.32	Calibration curve of Paracetamol obtained by UV-visible spectrophotometer at λ= 243 nm.	42
Figure.33	Percentage removal of Paracetamol on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).	43

Figure.34	percentage removal of Paracetamol on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).	44
Figure.35	Percentage removal of Paracetamol on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial, conc. = 50 ppm, contact time= 80 minutes and λ= 243 nm).	44
Figure.36	Percentage removal of Paracetamol on zeolite as a function of initial Paracetamol concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 243 nm).	45
Figure.37	Langmiur isotherm for the adsorption of Paracetamol on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0 g and λ= 243 nm).	45
Figure.38	Percentage removal of Ibuprofen on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH =2, concentration of Ibuprofen 50 ppm and λ= 224 nm).	48
Figure.39	Percentage removal of Diclofenac sodium on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH =6, concentration of Diclofenac sodium 50 ppm and λ= 276 nm).	49
Figure.40	Percentage removal of chlorophenarimine maleate on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH=2, concentration of chlorophenarimine maleate 50 ppm and λ= 261 nm).	49
Figure.41	Percentage removal of Paracetamol on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH =2, concentration of Paracetamol 50 ppm and λ= 243 nm).	50

List of Appendices

Appendix	Appendix Name	Page No.
Appendix A	Percentage removal of ibuprofen on zeolite as a function of pH by uv-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and at and λ= 224 nm).	60
Appendix B	Percentage removal of ibuprofen on zeolite as a function of Contact time by uv-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 and λ= 224 nm).	60
Appendix C	Percentage removal of ibuprofen on zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 224 nm).	60
Appendix D	Percentage removal of ibuprofen on zeolite as a function of initial Ibuprofen concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 224 nm).	61
Appendix E	Percentage Diclofenac sodium on zeolite as a function of pH by UV-visible spectrophotometer at T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 276 nm).	61
Appendix F	Percentage removal of Diclofenac sodium on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 276 nm).	61
Appendix G	Percentage removal of Diclofinac sodium on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, contact time= 80 minutes and λ= 276 nm).	62

Appendix H	Percentage removal of Diclofenac sodium on zeolite as a function of initial Ibuprofen concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 6, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 276 nm).	62
Appendix I	Percentage removal of Indomethacin on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).	62
Appendix J	Percentage removal of Indomethacin on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).	63
Appendix K	Percentage removal of Indomethacin on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 40 ppm, contact time= 60 minutes and λ= 225 nm).	63
Appendix L	Percentage removal of Indomethacin on zeolite as a function of initial Indomethacin concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 60 minutes and λ= 225 nm).	63
Appendix M	Percentage chlorophenarimine maleate on Zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 261 nm).	64
Appendix N	Percentage removal of Chlorophenarimine maleate on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 261 nm).	64
Appendix O	Percentage removal of Chlorophenarimine maleate on Zeolite as afunction of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 261 nm)	64

Appendix P	Percentage removal of Chlorophenarimine maleate on Zeolite as a function of initial Chlorophenarimine maleate concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.5 g, contact time= 80 minutes and λ= 261 nm).	65
Appendix Q	Percentage Paracetamol on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).	65
Appendix R	Percentage removal of Paracetamol on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).	65
Appendix S	Percentage removal of Paracetamol on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at(T= 25˚C, pH= 2, initial, conc. = 50 ppm, contact time= 80 minutes and λ= 243 nm).	66
Appendix T	Percentage removal of Paracetamol on zeolite as a function of initial Paracetamol concentration by UV-visible spectrophotometer at(T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 243 nm).	66

Chapter One

Introduction

Introduction

1.1 Back ground

Water has a broad impact on all aspects of human life including but not limited to health, food, energy, and economy. In addition to environmental, economic, and social impacts of poor water supply and sanitation (Mara, 2003).Water resources in Palestine are mainly the Jordan River, Wadi flows and groundwater (utilized mainly through wells and springs) (Aliewi, 2007). Water quality in the West Bank is considered acceptable in general; there are no serious indications of pollution in the deep aquifers. However, there is contamination of water in the shallow aquifer wells and springs (Nazer, 2010).Water resources in the West Bank / Palestine are limited. The expected increase in the Palestinian population and development in social, commercial, industrial and environmental sectors will increase the pressure on the already shortage water resources (Nazer, 2010).

The increasing consumption of water will result in an increase of wastewater production. The five public wastewater treatment plants in the West Bank are largely malfunctioning (Nazer, 2010). In most cases the wastewater is discharged into wadis without any type of treatment which increase the environmental problems. Moreover, increasing the percentage of the population connected to the sewer system, also in the West Bank - only increases the environmental deterioration when treatment capacity is not increased proportionally (Nazer, 2010).

With an increase in the standard of living there has been an increased dependence on different pharmaceuticals. There are many concerns that this results in increase of drugs which can enter the environment after passing through wastewater treatment facilities .That effluents from such treatment facilities feed into bodies of water that can end up in water supplies, which drinking water is taken from.

Among the methods used for wastewater treatment are the following: (1) Biodegradation: biological degradation (aerobic/anaerobic by micro-organisms) of drug substances leading to a reduction of the parent compounds and/or their metabolites during wastewater treatment. (2) deconjugation: conjugates of organic compounds such as steroid hormones have been shown

to be readily deconjugated in domestic wastewater and within sewage treatment plants (STPs) due to the large amounts of β-glucuridase enzyme present (produced by the fecal bacterium Escherichia coif). It seems probable that gluconoride and sulfate conjugates of drug compounds will be degraded by the same process. The effect will increase the excreted contribution of the active drugs to sewage and effluents. (3) Partitioning: partitioning between the aqueous and organic biomass phases is a key component in determining the ultimate concentrations of organic pollutants. Compounds with high log P (lipophilic molecules) values are known to sorb to sludge, while substances with lower values are more likely to stay in the aquatic phase, depending on the individual compound, and substances sorbing to solids may also be remobilized if they are not strongly bound. (4) Removal during sludge treatment: drugs may also be degraded during sewage treatment processes. Many pharmaceuticals are not thermally stable and so might be expected to break down during processes such as composting due to heat (as well as chemical and biodegradation). (5) Photo degradation: several pharmaceutical compounds have been shown to degrade due to the action of sunlight. The most extensively studied of these compounds is the analgesic/anti-inflammatory drug diclofenac, which has been shown to degrade in the aquatic environment due to ultraviolet (UV) light (Jones et al, 2005).

In practice there are various conventional methods available for removal of pharmaceuticals such as reverse osmosis, precipitation, chemical reduction etc., but these methods require high experimental set-up, more expensive and also less effective. Removal of pharmaceuticals by adsorption is one of the most promising techniques which are low cost, more effective and eco-friendly (Wang et al, 2009). Acidic pharmaceuticals, for example Ibuprofen, Diclofenac Sodium and Indomethacin, with pKa values from 4.1 to 4.9 occur as ions at neutral pH are, therefore, not readily adsorbed by sludge, and remain in the aqueous phase (Foye, 2008).

The aim of the current study is to study removal of acidic, basic and neutral selected pharmaceuticals from polluted water by using Natural Jordanian Zeolite.

1.2 Pharmaceutical contaminants in water

The contamination of water bodies in simplest words means water pollution. Thereby the abuse of lakes, ponds, oceans, rivers, reservoirs etc is water pollution. Pollution of water

occurs when substances that will modify the water in negative fashion are discharged in it. This discharge of pollutants can be direct as well as indirect.

Traces of pharmaceuticals, typically at levels in the micrograms to low nanograms per litre range, have been reported in the water cycle, including surface waters, wastewater, and groundwater and, to a lesser extent, drinking-water. Advances in analytical technology have been a key factor driving their increased detection. Their presence in water, even at these very low concentrations, has raised concerns among stakeholders, such as drinking-water regulators, governments, water suppliers and the public, regarding the potential risks to human health from exposure to traces of pharmaceuticals via drinking-water (World health organization, 2001).

The occurrence of pharmaceuticals in the environment and the water cycle at trace levels (in the range of micrograms to low nanograms per litre) has been widely discussed and published in literature in the past decade. The increase in detection is largely attributable to the advances in analytical techniques and instrumentation. Many surveys and studies have confirmed the presence of pharmaceuticals in municipal wastewater and effluents, and these have been identified as a major source of pharmaceuticals in drinking-water.

When the patients take the drugs their bodies absorb some of the medication, but the rest of it passes through and is flushed down the wastewater. The wastewater is treated before it is discharged into reservoirs, rivers or lakes. Then, some of the water is treated again to be used as drinking. But most treatments do not remove all drug residues (Grosvenor et al, 2004).

1.3 Ibuprofen

Ibuprofen is a common nonsteroidal anti-inflammatory drug widely used in the treatment of pain and fever. Its IUPAC name is 2-[4-(2-methylpropyl) phenyl] propanoic acid. It is marketed under the trade names of Advil and Motrin, among others. It is one of many chemicals included in a general group referred to as “pharmaceuticals and personal care products (PPCPs)”. Concern has stemmed from the low-level detection of PPCPs in surface waters and drinking water sources worldwide. (Kim et al, 2007), Fig .1 Show the Structure of Ibuprofen.

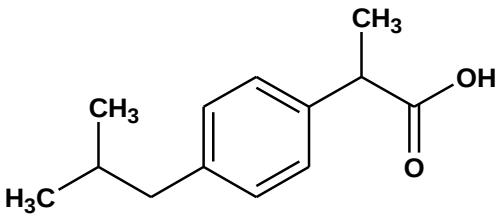

Figure.1 Chemical structure of Ibuprofen.

Ibuprofen is a chiral, propionic acid derivative which exhibits analgesic, fever-reducing, and anti-inflammatory action, and even surpassing that of acetaminophen (Vardanyan et al, 2006). Ibuprofen is an important nonprescription drug, and is the third-most popular drug in the world (Jones et al, 2005).

Ibuprofen's physiochemical properties (i.e. high water solubility, low volatility) with pka of 5.2 and logp of 3.97 suggest a high mobility in the aquatic environment, and consequently, it is a commonly detected PPCP in the environment [(Jones et al, 2005), (Vieno et al, 2005), (Zweiner et al, 2005)]. However, it is not very persistent and behaves differently in comparison to some other pharmaceutical compounds (Jones et al, 2005).

1.4 Diclofenac soduim

An important non-steroidal anti-inflammatory drug (NSAID) and widely used to reduce inflammation and as an analgesic in conditions such as in arthritis or acute injury. Its IUPAC name is Sodium 2-[(2, 6-dichlorophenyl) amino] phenyl] acetate (Scheurell et al, 2009). Fig .2 Show the Structure of Diclofenac soduim.

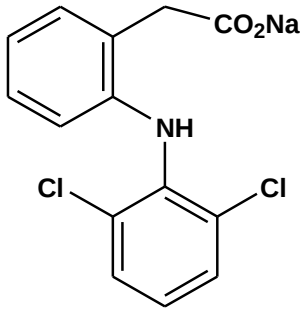

Figure.2 Chemical structure of Diclofenac Soduim.

Due to its extensive use, Diclofenac soduim has been considered as one of the most frequently detected pharmaceutical residues in water bodies thus far. It has been detected in influents and effluents from water treatment plants at concentrations up to mg/L level (Mompelat et al, 2009).

Municipal wastewater treatment plants are unable to cause a degradation of this compound. Ozonation, UV radiation, and activated carbon adsorption are potential treatments that might improve the effectiveness of Diclofenac soduim removal in municipal wastewater treatment plants (Beltran et al, 2009).

Adsorption to activated carbon is efficient only for hydrophobic contaminants (Ternes et al, 2002) and is considerably impacted by the presence of interfering substances such as humic acid and surfactants (Ternes et al, 2002).

Diclofenac's physiochemical properties (i.e. sparingly soluble in water, freely soluble in methanol, soluble in ethanol (96 per cent), slightly soluble in acetone.) with pka of 4.0 and logp of 4.26 suggest a high mobility in the aquatic environment (Pharmacopoeia, 2016).

1.5 Indomethacin

Indomethacin (IDM), is a known nonsteroidal anti-inflammatory drug (NSAID)broadly used to calm down acute joins and backbone pain and for the treatment of degenerative diseases of the joints and ligaments, Its IUPAC name is [1-(4-chlorobenzoyl)-5-methoxy-2-methylindol-3-yl]acetic acid, Fig . 3 shows the structure of Indomethacin.

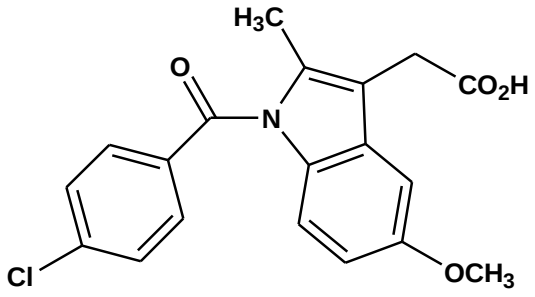

Figure.3 Chemical structure of Indomethacin.

During both human and veterinary usage, a significant proportion of the IDM can pass through the body unmetabolized and is thus released into water systems.

IDM is typically considered stable in the environment. While acute effects of the exposure of aquatic animals to IDM or other NSAIDs may not appear immediately, the long-term presence of such a xenobiotic micro contaminant in aquatic systems may lead to chronic toxicity and subtle effects such as endocrine disruption, growth inhibition and cytotoxicity to aquatic animals (Fent et al, 2006).

The physiochemical properties of the indomethacin are: practically insoluble in water, sparingly soluble in alcohol with pka of 3.8. (Pharmacopoeia, 2016).

1.6 Chloropheniramine Maleate

Chlorpheniramine maleate (CPM) has been used extensively as an antihistaminic drug for symptomatic relief of common cold and allergy. CPM is typically administered as 4 mg dose, 2 to 3 times daily. The absolute bioavailability of Chloropheniramine maleate is 25 to 50 % (Scheurell et al, 2009), Its IUPAC name is (3*RS*)-3-(4-Chlorophenyl)-*N*, *N*-dimethyl-3-(pyridin-2-yl) propan-1-amine hydrogen (*Z*) – butenedioate. Fig.5 shows the structure of Chloropheniramine maleate.

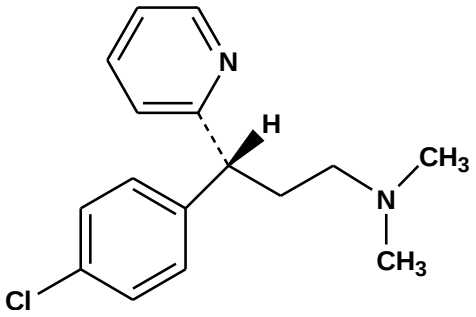

Figure. 4 Chemical structure of Chloropheniramine maleate

Chloropheniramine maleate is a white or almost white, crystalline powder, freely soluble in water and soluble in ethanol (Pharmacopoeia, 2016), it is a basic drug with pka of 9.3 and logp of 3.38.

1.7 Paracetamol:

Paracetamol has been used extensively as analgesic; antipyretic drug. Paracetamol is crystalline powder white or almost white, it's sparingly soluble in water, freely soluble in alcohol, very slightly soluble in methylene chloride, its IUPAC name is *N*-(4-Hydroxyphenyl)

acetamide. Fig.6 Show the Structure of Paracetamol (Pharmacopoeia, 2016). It is neutral drug with pka of 9.5 and logp of 0.31. Figure.5 shows chemical structure of Paracetamol.

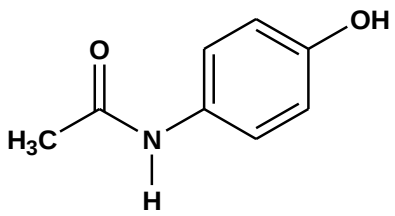

Figure.5 Chemical structure of Paracetamol

1.8 Zeolite

Zeolite is hydrated alum inosilicate materials having cage like structures with internal and external surface areas of up to several hundred square meters per gram and cation exchange capacities of up to several milliequevalent per kilogram (Baker et al, 2009).

There is natural zeolite and synthesized, both natural and synthetic zeolite are used in industry as adsorbents, soil modifiers, ion exchanger and molecular sieves (Baker et al, 2009).

Zeolites were introduced in 1954 as adsorbents for industrial separations and purifications. Because of their unique porous properties, zeolites are used now in a variety of applications with world production estimated to be in the range of 2.5 million to 3 million metric tons (Leake et al, 2008). They are used in petrochemical cracking, water softening and purification, in the separation and removal of gases and solvents, agriculture, animal husbandry and construction.

A zeolite's degree of hydrophilicity can be fine-tuned by adjusting the silica to alumina ratio; zeolites are more hydrophilic at lower ratios so greater affinity for adsorbing water, The pores are comprised of 12 member oxygen rings , Fig.7 shows the structure of zeolite.

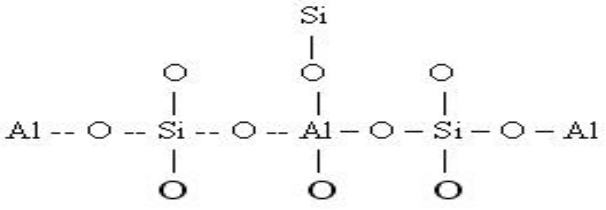

Figure.6 Chemical structure of zeolite

Zeolites are naturally formed near volcanos where the volcanic ash reacts with alkaline water to form the crystaline structures. Naturally occurring zeolites are mined in many parts of the world. The well-known and industrially important zeolites have been discovered in 1950-1970 and may be classified into three groups according to Al/Si ratio in their frameworks (Derouane, 1984):

I. "Low-silica" or aluminum rich zeolites A and X (ratio Si/Al ≈ 1).

Zeolites A and X (the most common commercial adsorbents) are nearly "saturated" in aluminium in the framework composition with a molar ratio of Si/Al ≈ 1, which is considered as highest aluminum content possible in tetrahedral alumosilicate frameworks. As a consequence they contain the maximum number of cation exchange sites balancing the framework aluminum, and thus the highest cation contents and exchange capacities. These compositional characteristics combined give them the most highly heterogeneous surface known among porous materials, due to exposed cationic charges nested in an aluminosilicate framework which results in high field gradients. Their surface is highly selective for water, polar and polarizable molecules which serves as the basis for many applications particularly in drying and purification.

II. "Intermediate silica" zeolites: zeolite Y, mordenite, zeolite L, natural zeolites (ratio Si/Al = 2 - 5).

This zeolite has superior stability characteristics reflecting high Si/Al molar ratio of 3-5. Therefore, zeolites with higher content of silicon were needed, primarily to improve stability characteristics, both thermal and to acids. The third commercially important molecular sieve zeolites type Y, with a Si/Al ratio from 1.5 to 3.0 (Derouane, 1984).

The next commercially successful synthetic zeolite was a large pore mordenite with ratio Si/Al ≈ 5. The improvement in thermal, hydrothermal, and acid stability coupled with its specific structural and compositional characteristics resulted in application of mordenite as an adsorbent and hydrocarbon conversion catalyst (Guisnet et al, 2002). Type L zeolites with a Si/Al = 3.0 have unique framework topology. They were adapted as commercial catalysts in selective hydrocarbon conversion reactions.

III. "High silica" zeolites: zeolite beta, ZSM-5 (ratio Si/Al ≥ 10).

These are molecular sieve zeolites with Si/Al ratios from 10 to 100 or higher, with different surface characteristics. In contrast to the "low" and "intermediate" silica zeolites, representing heterogeneous hydrophilic surfaces within a porous crystal, the surface of the high silica zeolites is more homogeneous with an organophilic-hydrophobic selectivity (Guisnet et al, 2002). They adsorb stronger the less polar organic molecules and only weakly interact with water and other polar molecules.

In addition to this novel surface selectivity, the high silica zeolite compositions still contain a small concentration of aluminum in the framework and the accompanying stoichiometric cation exchange sites. Thus, their cation exchange properties allow the introduction of acidic OH-groups via the well-known zeolite ion exchange reactions, essential to the development of acid hydrocarbon catalysis properties.

The studied Zeolite area is located 65 km S-SE of Ma'an in the southern part of Jordan, and covers a surface of about 875 km^2. Successive layers of shale and siltstone were reported for the Paleozoic rocks of southern Jordan (Khoury et al, 1986).

The sand, silt and clay average values are: 10%, 68.6%, and 21.4% for borehole A; 28.9%, 55.8%, and 15.2%, 17-41% for quartz, 1.5-10% for feldspars, 23-40% for mica, illite and vermiculite and 3.5-25% for kaolinite in borehole B. Table 1 shows average chemical composition (%) of the bulk sample in the borehole B.

Table.1 Average chemical composition (%) of the bulk sample in the borehole B

	SiO_2	Al_20_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	LOI
Borehole B	61.3%	14.8%	7.8%	1.2%	0.5%	1.3%	3.6%	0.9%	8.8%

The Zeolite used in this study is Intermediate silica with Si/Al ratio 4.142(Khoury et al, 1986).

1.9 Ion Exchange/ properties of zeolite

Cation exchange properties of traditional aluminosilicate zeolites arise from the isomorphous positioning of aluminium in tetrahedral coordination within their Si/Al frameworks (Cejka,

2005). This imposes a net negative charge of the framework ($Si^{+4} \rightarrow Al^{3+}$) counter balanced by cations held within the cavities and channels. Ionic character of bonding between interstitial cations and the framework provide facile cation exchange for zeolites with open frameworks, where cations often readily exchanged for other cations in aqueous solution, though in some of the narrow-pored frameworks, such as natrolite, cation replacement is slow and difficult.

Cation exchange is exploited in water softening, where alkali metals such as Na^+ or K^+ in zeolite framework are replaced by Ca^{2+} and Mg^{2+} ions from water. Many commercial washing powders thus contain substantial amounts of zeolites that enhance washing efficiency. LTA have the largest scale production of synthetic zeolites for use as "builders" in domestic and commercial detergents to remove the calcium and magnesium "hardness" (Townsend et al, 2001).

Interstitial cations in zeolites can be exchanged to fine-tune the pore size of zeolites. For example, the sodium form of zeolite A has a pore opening of approximately 4 Å (4A molecular sieve). If Na^+ is exchanged with the larger K^+, the pore opening is reduced to approximately 3 Å; Ca^{2+} replaces 2 Na^+, thus, the pore opening increases to approximately 5 Å. Ion exchange with other cations is sometimes used for particular separation purposes. Another potential application of zeolites is a drug delivery, when water in the structure is substituted by other liquid compound. Such treated zeolites act as a delivery system for the new fluid.

1.10 Adsorption:

Adsorption is a process with three steps. The first step is for the contaminant to be transferred from the bulk phase to the outer surface of the adsorbent material. In the second step, the contaminant molecule diffuses from the smaller of the areas of the outer surface into the areas within each adsorbent. This includes the macropores, transitional pores, and micropores. The third step is when the contaminant molecule is adsorbed into the surface of the pore structure within the adsorbent.

It may be expected that the process of formation of kaolinite starts with the displacement of $A1_3^+$ ions from tetrahedral position into the interlayer space with a simultaneous change of

coordination from 4-fold into 6-fold. At the same time K^+ is removed and protons and molecules of water are incorporated for the formation of the full coordination of aluminum cations .The weathered material was mechanically transported. The presence of remnants of feldspars showing all stages of alteration into sericite, illite and kaolinite suggests variable rates of weathering (Khoury et al, 1986), the same mechanism happened with zeolite.

To model the adsorption behavior, two adsorption isotherms were studied and their correlation with experimental data was assessed.

The Langmiur model was used to explain the observed phenomenon. The equilibrium data was analyzed using the following linear equation: [30]

$C_e/q_e = 1/Q_{max}\,K + C_e/Q_{max}$ Where:

C_e is the equilibrium concentration of solute in the bulk solution (mg/L)

q_e is the amount of solute adsorbed per unit weight of adsorbent (mg/g)

Q_{max} is the adsorption capacity (mg\g)

K is the thermodynamic equilibrium constant related to the free adsorption energy

A plot of Ce\qe versus Ce was linear and the constants Qmax and K were determined from the slope and the intercept of the plot

The adsorption behavior was also tested by Freundlich model .[31]

$\text{Log } x/m = \text{Log } K_f + 1/n \text{ Log } C_e$

Where: x\m is the amount of solute adsorbed per unit weight of adsorbent (mg/g)

C_e is the equilibrium concentration of solute in the bulk solution (mg/L)

K_f is a constant indicative of the relative adsorption capacity of the adsorbent (mg/g)

1\n indicates the intensity of the adsorption

CHAPTER TWO

Literature review

2. Literature review

Ruggles et al 2013 studied removal of Ciprofloxacin from Water using zeolites and ozone. Results of this study showed that the Y- zeolite was able to adsorb nearly 100% of the ciprofloxacin. S. K. Behera et.al (2012) studied Sorption of ibuprofen, a non-steroidal anti-inflammatory drugs, onto various soil minerals, viz., kaolinite, montmorillonite, goethite, and activated carbon, as a function of pH (3-11), ionic strength (NaCl concentration: 0.001-0.5 M), through batch experiments. Experimental results showed that the sorption of ibuprofen onto all sorbent was highest at pH 3, with highest sorption capacity for activated carbon (28.5 mg/g). Among the minerals, montmorillonite sorbed more ibuprofen than kaolinite and goethite, with sorption capacity increased in order goethite (2.2 mg/g) < kaolinite (3.1 mg/g) <montmorillonite (6.1 mg/g)

Rossner, Snyder, Knappe (2009) investigated removal of emerging contaminants of concern such as endocrine-disrupting chemicals, pharmaceutically active compounds, personal care products, and flame retardants is a desirable water treatment goal by alternative adsorbents. Among the tested adsorbents are:

- Activated carbon which was the most effective.
- Carbonaceous resin which was less effective than the activated carbon as this adsorbent had a smaller volume of pores in the size range required for the adsorption of many contaminants.
- Zeolites found to be less effective than the carbonaceous adsorbents.

Baker, H. M., Massadeh, A. M., & Younes, H. A. (2009) investigated removal of heavy metal ions using zeolites from water samples using column and batch methods. According to this study, removal of metal ion is slightly increasing with decreasing particle size of zeolite.

2.1. Problems statement

Large amounts of pharmaceuticals are used for the prevention, diagnosis and treatment of diseases in humans and animals. Most pharmaceuticals are not completely degraded after application, which is discharged directly into water even after treatment causing environmental pollutions.

2.2 Hypotheses and research questions

Zeolite is efficient in pharmaceuticals removal from polluted water.

Is natural zeolite more efficient in removing the selected pharmaceuticals than the adsorbents that are used in previous studies?

What is the maximum capacity of natural Zeolite for pharmaceuticals removal?

Is the adsorption of selected pharmaceutical compounds on zeolite follow Langmuir or Frendlich isotherm?

What is the optimum conditions included pH, contact time, adsorbent dosage and the initial pharmaceuticals concentration in the adsorption process?

2.3 Objectives and aims

The main objective of this research is to remove selected Pharmaceuticals compounds from polluted water using a natural Zeolite as an adsorbent.

This goal will be achieved by the following objectives:

1. To study the effect of pH, adsorbent dosage, and initial concentration of pharmaceuticals on the adsorption process.
2. To determine the optimum conditions for the adsorption process.
3. To study the adsorption kinetics of selected pharmaceutical
4. To determine the maximum adsorption capacity of the adsorbent

CHAPTER THREE

Materials and Methods

This chapter consists of three parts: part one describes the instruments used in all experiments, part two documents the chemicals and reagents utilizes, the last part deals with the methods implemented.

3.1 Instrumentations

3.1.1 pH meter

pH meter model HM-30G: TOA electronics™ to measure the pH value for the samples.

3.1.2 Shaker

Pharmaceuticals solutions were shaken with an electronic shaker (Big bill shaker, Model No.: M49120-26, 220-240 V 50\60 Hz.) at 250 rpm.

3.1.3 Electronic Balance

Pharmaceuticals weighted by an electronic balance (AW 220 electronic balance, Model No.: D422601283, which is made in Japan).

3.1.4 UV-Visible Spectrophotometer

UV-Visible Spectrophotometer model UV-1601 220 v, Model No. : 206-67001-93, Australia.

3.2 Chemicals and Reagents

Pure standards of ibuprofen (> 99%), Diclofenac Sodium (> 99%) ,Indomethacin (> 99%), Chlorophenarmin Maleate (> 99%), Paracetamol (> 99%) were obtained from local pharmaceutical company, Methanol (> 99%) from Sigma aldrich, Natural Jordanian Zeolite were brought from Jordan university (DR.Hani Khouri).

3.3 Methods

The removal of selected pharmaceutical from water was performed by batch and column experiments. The study investigated the effect of variable parameters such as pH, contact time, adsorbent dosage and initial concentration on the efficiency of the removal process. Langmiur and Freundlich isotherms were used to study the adsorption isotherm of the pharmaceuticals on Zeolite.

3.3.1 Calibration curves

(a) Stock solution: six stock solutions were prepared by dissolving separately Ibuprofen, Diclofenac Sodium, Indomethacin, Chlorophenarmin and Paracetamol in a mixture of methanol and water (1:1 ratio) to a concentration of 1000 ppm for the use in (b).

(b) Calibration curves: The following diluted solutions were prepared from the stock Solution of ibuprofen (10.0, 20.0, 40.0 and 50.0ppm), and absorbance was read for the solutions using methanol/water mixture as a blank.

Absorbance for the pharmaceuticals vs. concentration of the pharmaceuticals (in ppm) was then plotted, and R^2 of the plots are recorded.

3.3.2 Batch adsorption isotherms

Equilibrium relationships between different adsorbent dosage (Natural Jordanian Zeolite) and adsorbate (Ibuprofen , Diclofenac Sodium, Indomethacin , Chlorophenarmin and Paracetamol) are described by adsorption isotherms, by studying the percentage of adsorbate removal occurred by adsorbent at different concentrations (prepared in distilled water with adjusted at different pH by using 1M hydrochloric acid. Samples at specific time intervals were taken. The following procedures were applied:

3.3.2.1 Effect of pH

50 mg of adsorbent was transferred to 250 ml Erlenmeyer flask consecutively and 50 ml of 50 ppm of contaminated solutions was added to the flask and the pH was adjusted to 2, 4, 6 and 8 by 0.2M HCl or 1M NaOH. The solutions were allowed to stir for 2 hours at 25˚C. Then the solutions were filtered using a 0.45 μM Millipore filter and the absorbance was determined by using UV-Visible spectrophotometer at λ max, λ max for the selected pharmaceuticals is shown in table.2.

Table.2 λ max for the selected pharmaceuticals.

Pharmaceutical Name	**λ max**
Ibuprofen	224 nm
Diclofenac Sodium	276 nm
Indomethacin	225 nm
Chlorophenarmin Maleate	261 nm
Paracetamol	243 nm

3.3.2.2Effect of contact time

50 mg of zeolite was transferred to 50 ml Erlenmeyer flask, and 50 ml of 50 ppm adsorbate (pharmaceuticals) was added. The solutions were allowed to stir for different time intervals (10,20,40,60,80 and 120 minutes) at specific pH (6 for Diclofenac sodium and 2 for the rest) at 25˚C. Each sample was filtered using a 0.45 μM Millipore filter and absorbance was determined by using UV-Visible spectrophotometer at λ max.

3.3.2.3Effect of adsorbent dosage

0.5, 1.0, 1.5 and 2.0 g of adsorbent were transferred to separate four 50 ml Erlenmeyer flasks and 50 ml of 50 ppm contaminated solutions were added to each flask. Solutions were left to stir for 80 minutes and pH was adjusted to 2 using 0.2M HCl. Then solutions were filtered using a 0.45 μM Millipore filter and absorbance was determined by using UV-Visible spectrophotometer at λ max.

3.3.2.4Effect of initial absorbent concentration

10.0, 20.0, 40.0 and 50.0 ppm of contaminated solutions were used. 50 ml of each solution with 1.0 g of adsorbent were transferred to a 50 ml Erlenmeyer flask. The solutions were allowed to stir for 80 minutes and pH was adjusted to 2 using 0.2N HCl at 25˚C, then the solutions were filtered using a 0.45 μM Millipore filter and absorbance was determined by using UV-Visible spectrophotometer at λ max.

All experiments described were conducted in triplicates and results were taken as average of three measurements.

3.3.3 Column Experiment

Column filter experiments were performed with 9/1 (w/w) mixtures of quartz sand and zeolite (20 cm layer) in a column of 25 cm length and 3.5 cm diameter prepared by mixing 10 g of Zeolite and 90 g sand. Quartz sand was thoroughly washed by distilled water and dried at 105 ˚C for 24h prior its use. Wool layer of 2 cm was placed at the bottom of the column to prevent clogging. 1000 mL of 50 ppm adsorbate solutions were passed through the column at a fixed flow rate of 2 mL min^{-1}.

Eluted fractions of 100 mL (each) were collected at chosen times, and analyzed for Ibuprofen, Diclofenac Sodium, Indomethacin, Chlorophenarmin and Paracetamol.

All experiments described were conducted in triplicates.

Chapter Four

Results and discussion

4. Results and discussion

The adsorption process of five pharmaceuticals: three acidic: Ibuprofen, Diclofenac sodium and Indomethacin, one basic: Chloropheniramine maleate, and one neutral compound :Paracetamol were studied on natural Jordanian zeolite.

4.1 Batch Experiment

4.1.1 Acidic Pharmaceuticals

4.1.1.1 Ibuprofen

4.1.1.1.1Calibration curve for Ibuprofen using UV-visible spectrophotometer

The calibration curve was obtained by plotting absorbance versus concentration of Ibuprofen and is displayed in Figure.8. The Figure shows excellent linearity in the range 10 -50 ppm with correlation coefficient (R^2) of 0.9967, this indicates that the method used is linear.

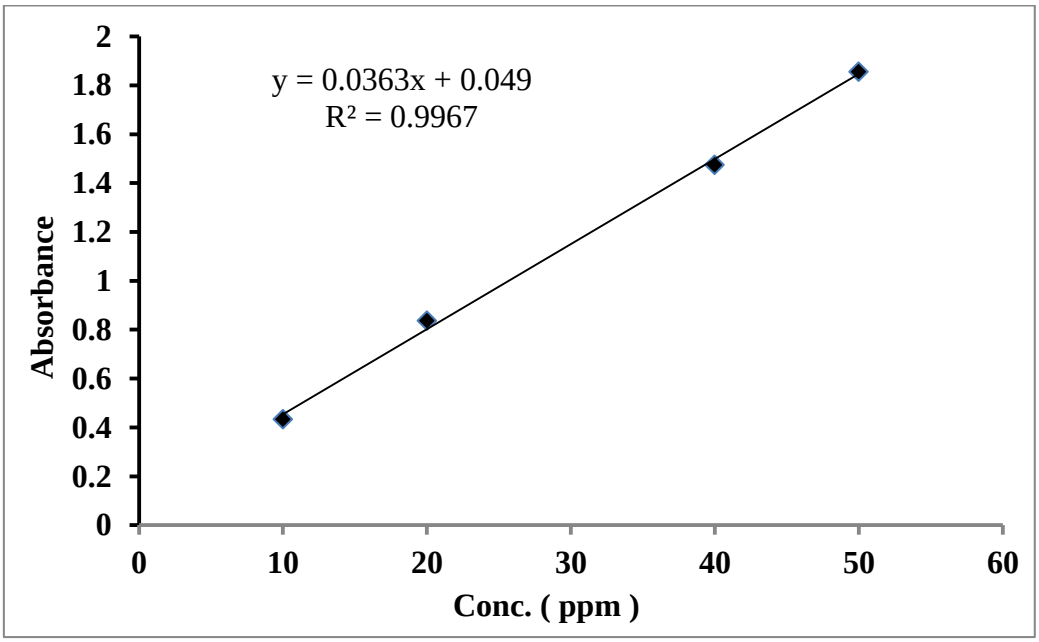

Figure.7 Calibration curve of Ibuprofen obtained by UV-visible spectrophotometer at λ = 224nm.

4.1.1.1.2Effects of different parameters on adsorption of Ibuprofen

The adsorption of Ibuprofen was studied using different parameters (pH values in the range of 2 – 8, contact time at different time intervals (10-160 minutes), adsorbent dosage from 0.5-2.0 g, initial Ibuprofen concentration from 10 -50 ppm), to model the adsorption behavior of ibuprofen on zeolite adsorbent, two adsorption isotherms were studied and their correlation with experimental data was assessed. The following figures (8-12) shows the percentage removal of Ibuprofen at different parameters and Freundlich isotherm for the adsorption of Ibuprofen on Zeolite .

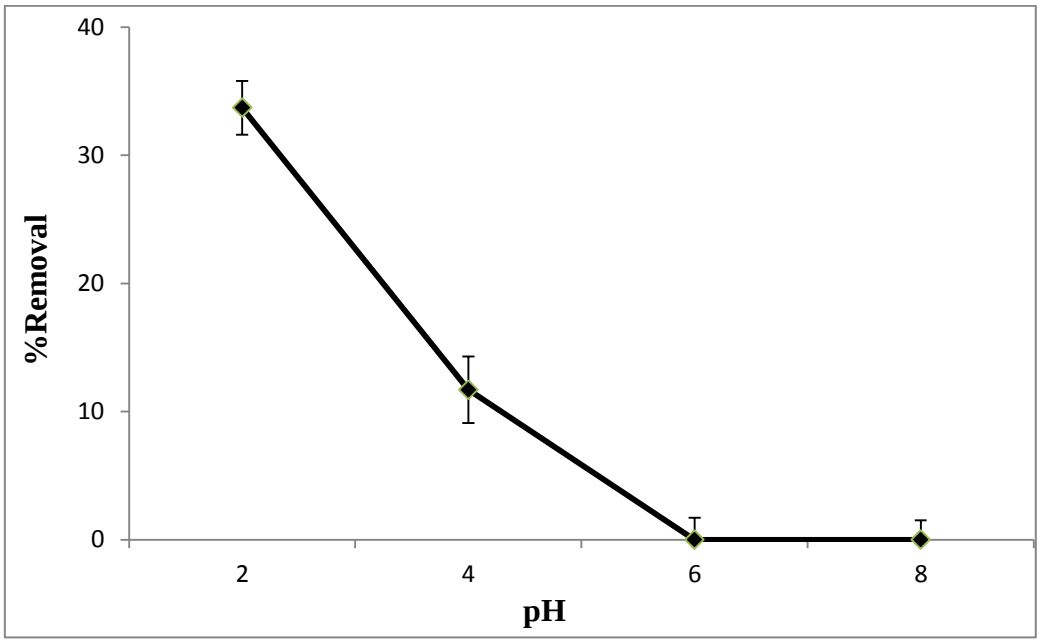

Figure.8 Percentage removal of Ibuprofen on Zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 224nm).

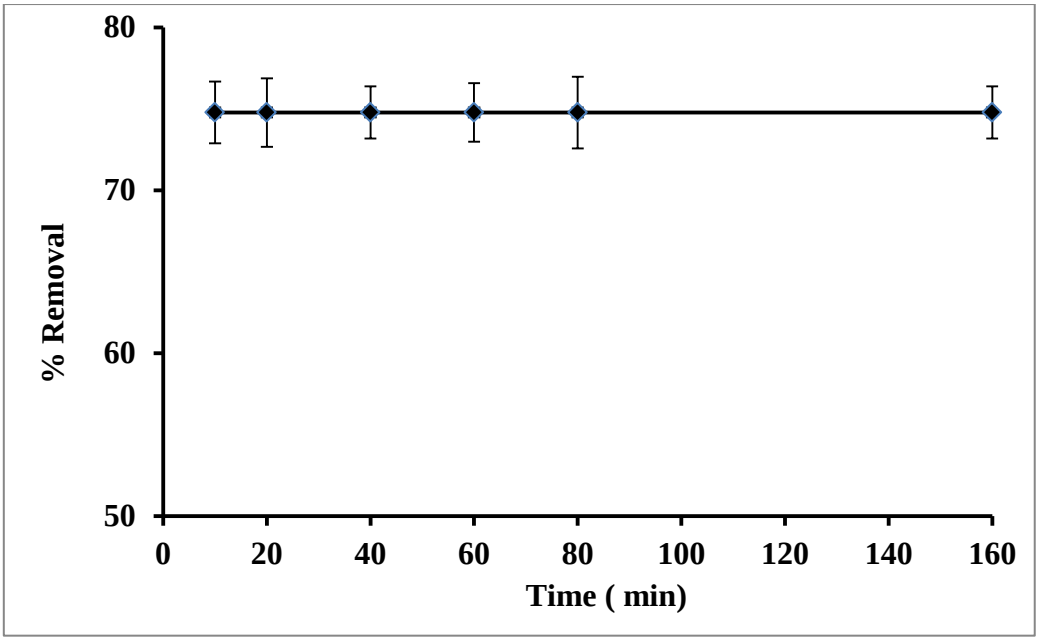

Figure.9 Percentage removal of Ibuprofen on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 224nm).

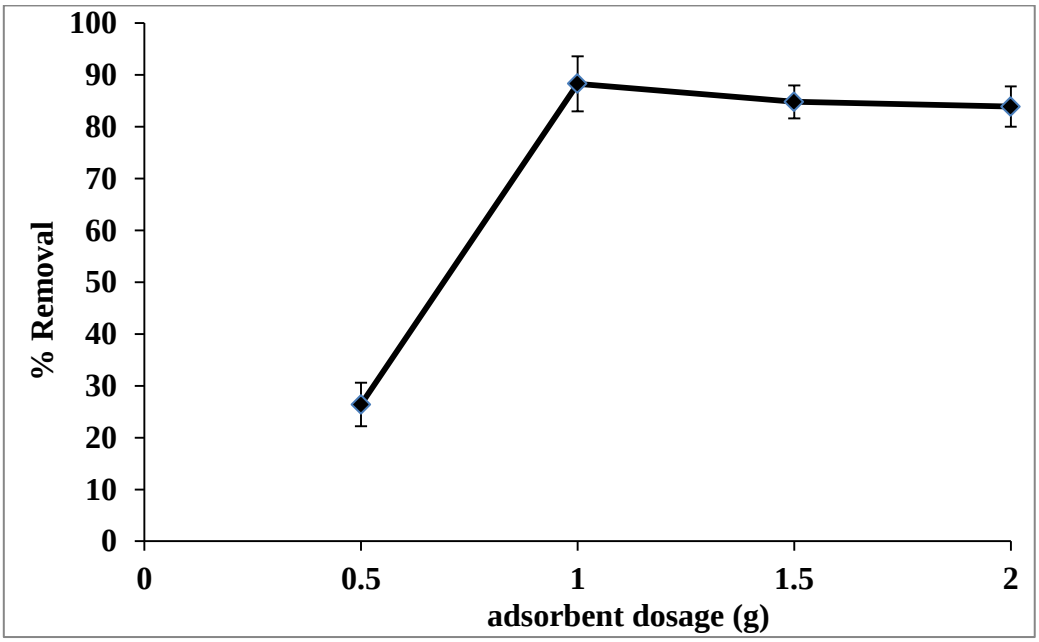

Figure.10 Percentage removal of Ibuprofen on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 224nm).

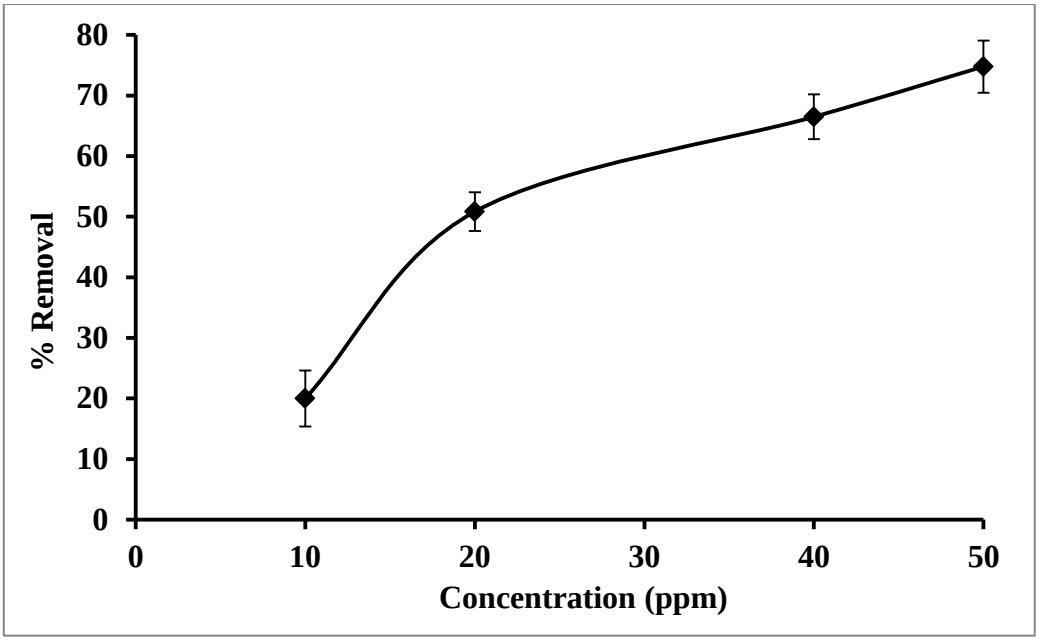

Figure.11 Percentage removal of ibuprofen on zeolite as a function of initial Ibuprofen concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 224nm).

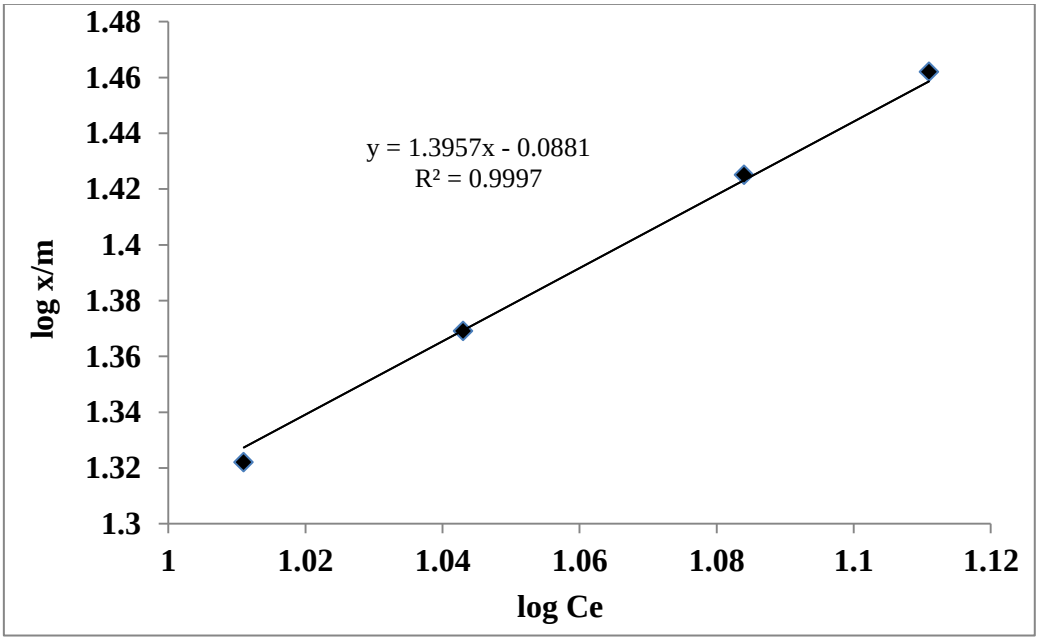

Figure.12 Freundlich isotherm for the adsorption of Ibuprofen on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g and λ= 224nm).

Table.3 Freundlich isotherm for the adsorption of Ibuprofen on Zeolite using UV-visible spectroscopy at T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g

log Ce	log x/m
1.011	1.322
1.043	1.369
1.084	1.425
1.111	1.462

Referring to ibuprofen structure in figure.1 it has acidic nature with pka of 5.2 so when the pH of ibuprofen solution increase the percentage of unprotonated ibuprofen (with carboxyl group) will increase, this increase will cause repulsion between ibuprofen and the negative charge of zeolite which decreases removal of ibuprofen.

At low pH, repulsion between ibuprofen and negative charge of zeolite is minimum which cause highest removal of ibuprofen, this trend is shown in Figure.8. The percentage removal of ibuprofen is almost constant as the adsorption time increases shown in Figure.9. The percentage removal of Ibuprofen increased with increase of adsorbent dosage and attained equilibrium at (1.5 – 2.0 g) as shown in Figure.10.The percentage removal of Ibuprofen increases with increasing the concentration of Ibuprofen as shown in Figure.11.

A plot of Ce\qe versus Ce was nonlinear which indicate that the Langmuir isotherm has bad fit between parameters, a plot of log x\m versus log C_e on the other hand was linear and the constants k_f and n were determined from the slope and the intercept of the plot, which was found to be 1.23 mg\g and 0.717at 25˚C and pH= 2 with 1.0 g adsorbent dosage The correlation coefficient obtained with the Langmuir equation was high (R^2= 0.9997) indicating that Freundlich isotherm model is the best to describe removal process of Ibuprofen on Zeolite. This trend is shown in Figure.13 and table.3.

Removal of Ibuprofen in this study was compared with other studies. In our study the best removal of Ibuprofen on natural Jordanian zeolite at pH=2, after contact time of 80 minutes, adsorbent dosage of 1.0g and initial ibuprofen concentration of 50 ppm was 88.3%. According to a study of S. K. Behera et.al (2012) ((Sorption of ibuprofen from water onto various soil

minerals)), the best removal of Ibuprofen was at pH=2-4 with about 90 %. (S. K. Behera et al, 2012), which is very close to the percentage removal of Ibuprofen obtained by natural Jordanian zeolite in this study. Also, Vieno, N., et al. (2006) studied removal of Ibuprofen in drinking water and found the best removal of Ibuprofen by ferric sulphate coagulation was at pH= 4.5 - 6 with 50 % removal. (Vieno et al, 2006). Khalaf, S., et al. R. (2013), studied efficiency of advanced wastewater treatment plant system and laboratory-scale micelle-clay filtration for the removal of ibuprofen residues, and found that the best removal of Ibuprofen by carbon activated was at pH=2-4 with 95.7 % removal (Khalaf et al, 2013). Simazaki, D., et al. (2008), studied removal of Ibuprofen by chlorination, coagulation–sedimentation and powdered activated carbon treatment, and found that the best removal of ibuprofen by chlorination with 80 % removal. (Simazaki et al, 2008).

4.1.1.2 Diclofenac Sodium

4.1.1.2.1Calibration curve for Diclofenac Sodium using UV-visible spectrophotometer

The calibration curve was obtained by plotting absorbance versus concentration of Diclofenac sodium and is displayed in figure .14. The Figure shows excellent linearity in the range 10 -50 mg L^{-1} with correlation coefficient (R^2) of 0.9967, this indicates that the method used is linear.

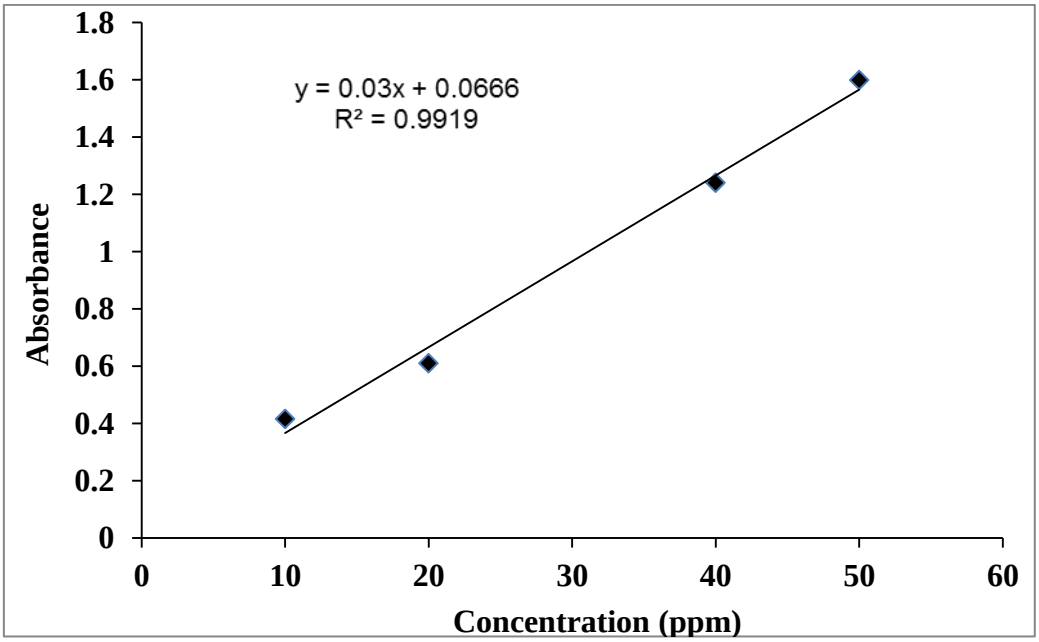

Figure.13 Calibration curve of Diclofenac sodium obtained by UV-visible spectrophotometer at λ= 276nm.

4.1.1.2.2Effects of different parameters on adsorption of Diclofenac Sodium

The adsorption of Diclofenac sodium was performed at different parameter (pH values in the range of 2 – 8, contact time at different time's intervals (10-120 minutes), adsorbent dosage from 0.5 - 2.0 g, initial Diclofenac sodium concentration from 10 -50 ppm), to model the adsorption behavior of Diclofenac sodium on zeolite adsorbent, two adsorption isotherms were studied and their correlation with experimental data was assessed. The following figures (14-20) shows percentage removal of Diclofenac sodium at different parameters, Langmuir isotherm and Freundlich isotherm for the adsorption of Diclofenac sodium on natural Jordanian zeolite.

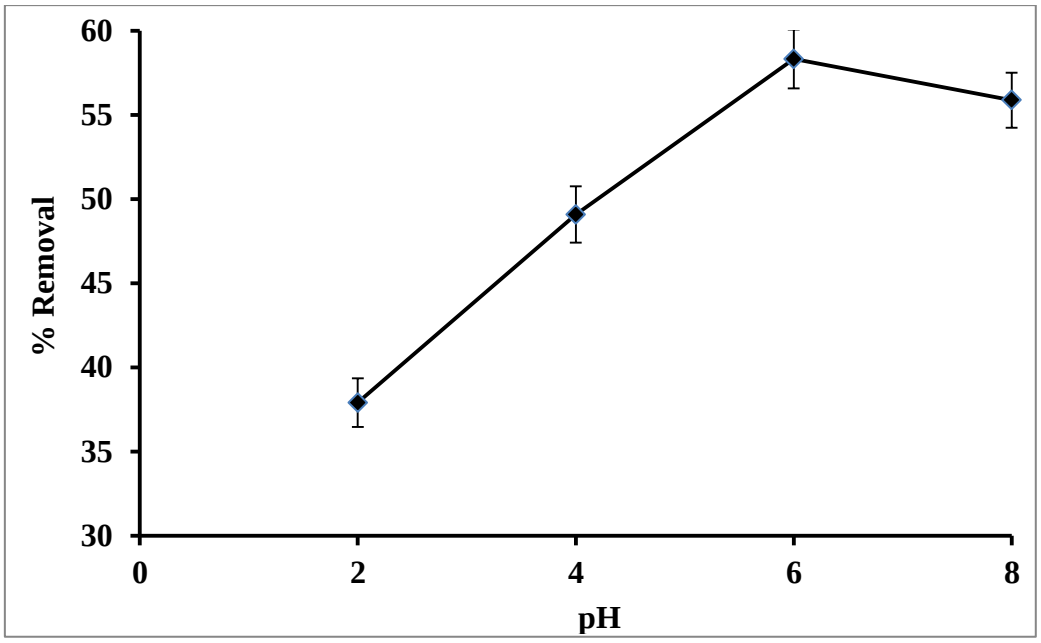

Figure.14 Percentage removal of Diclofenac sodium on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 276 nm).

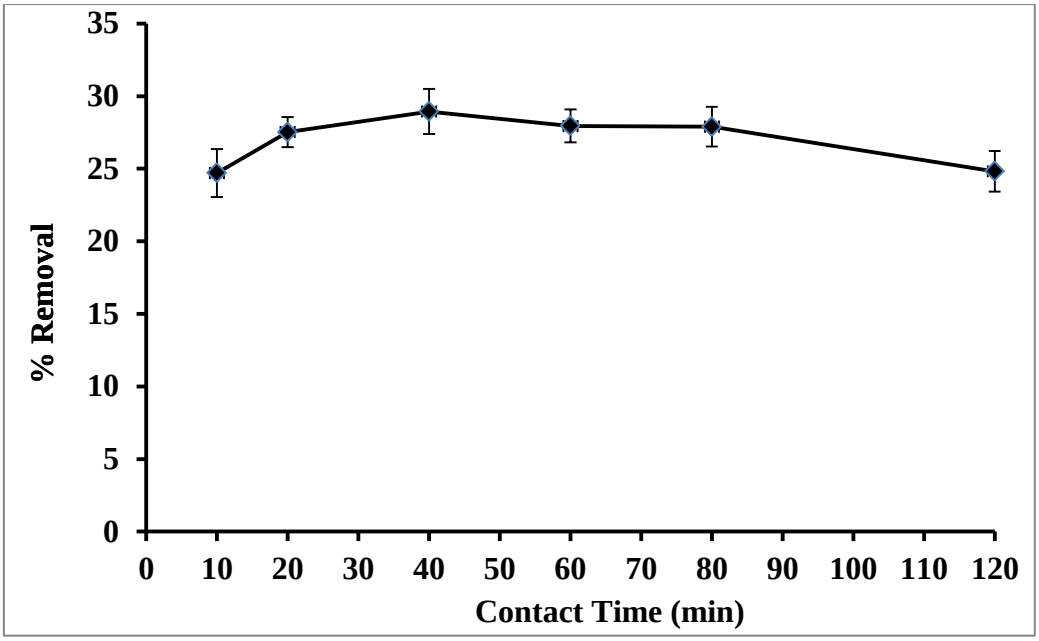

Figure.15 Percentage removal of Diclofenac sodium on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 276 nm).

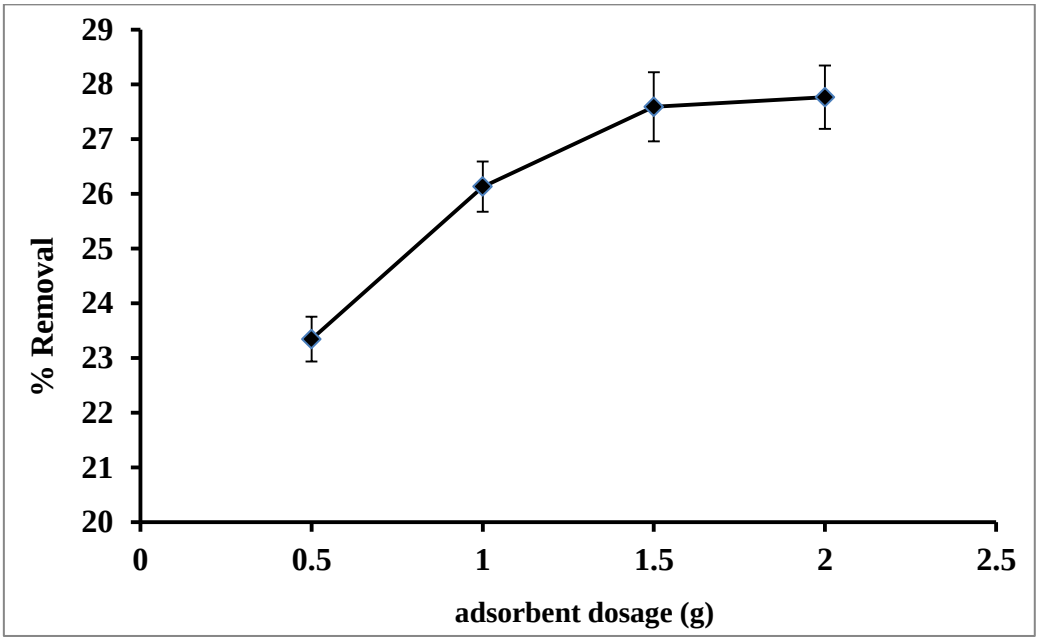

Figure.16 Percentage removal of Diclofinac sodium on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, contact time= 80 minutes and λ= 276 nm).

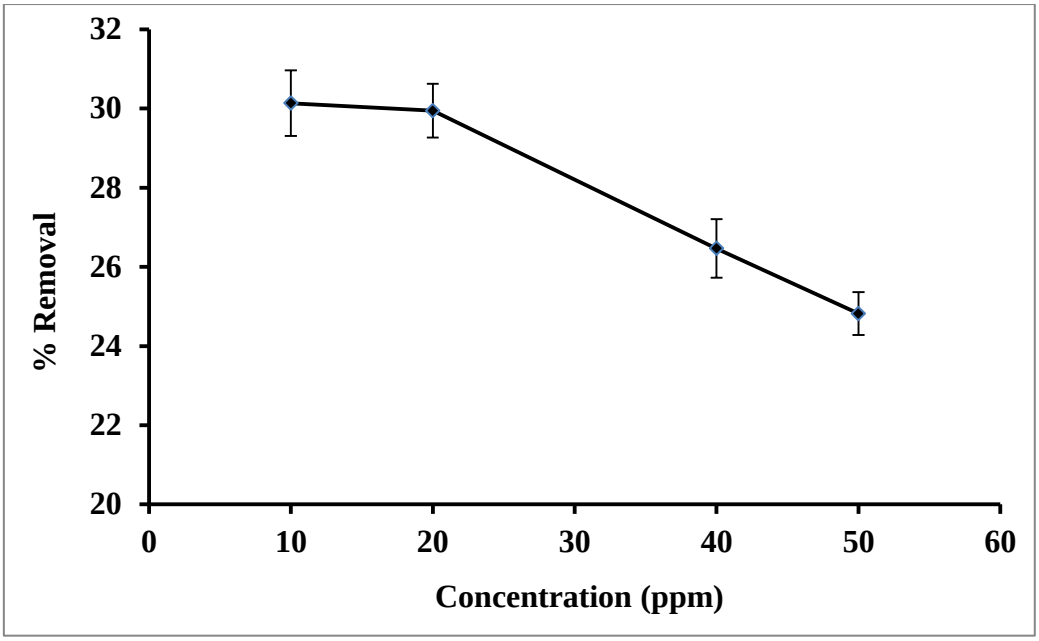

Figure.17 Percentage removal of Diclofenac sodium on Zeolite as a function of initial Diclofenac sodium concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 6, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 276 nm).

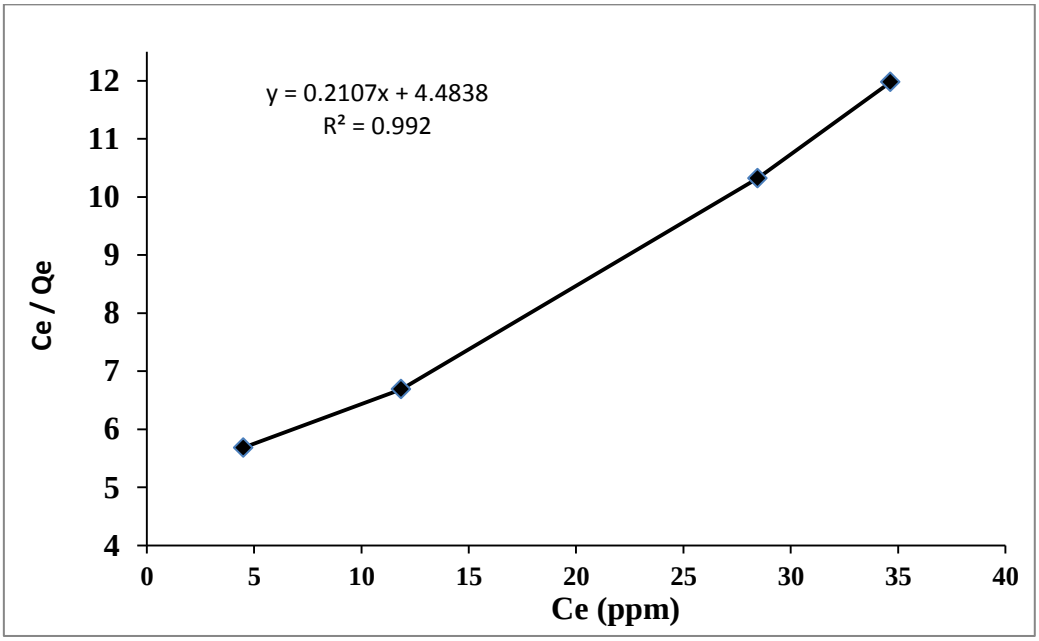

Figure.18 Langmiur isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 6, contact time= 80 minutes, adsorbent dosage = 1.0 g and λ= 276 nm).

Table.4 Langmiur isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 6, contact time= 80 minutes, adsorbent dosage = 1.0 g).

Conc (T=80) (Ce)(mg L-1)	Q=(M Initial-M Final/0.5	Ce/Qe
4.495	11.01	5.683
11.85	14.3	6.69
28.452	23.096	10.32
34.645	30.71	11.98

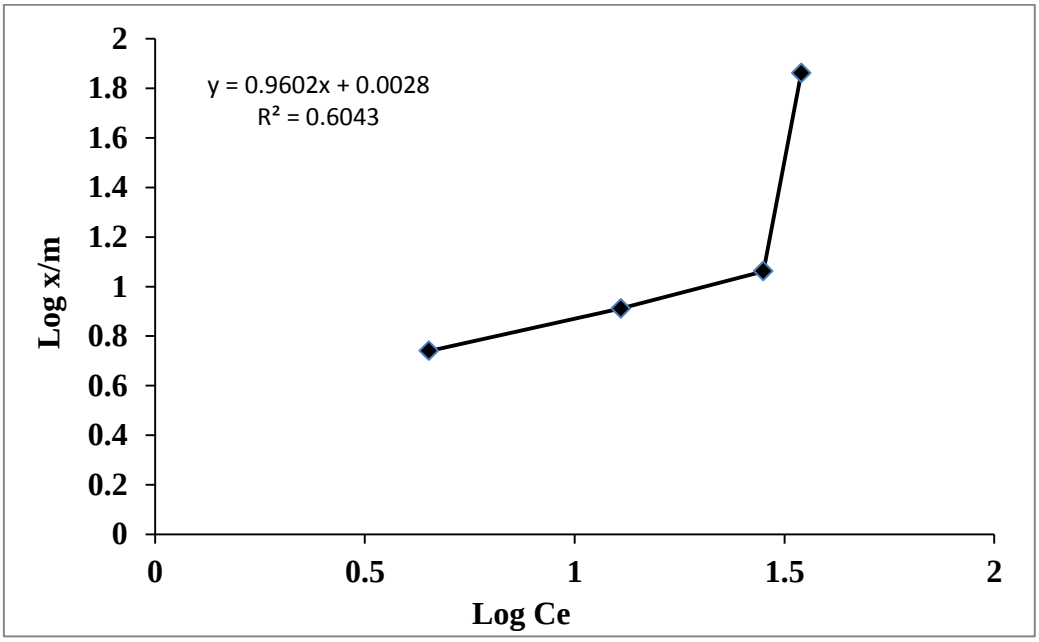

Figure.19 Freundlich isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 6, contact time= 40 minutes, adsorbent dosage = 1.0g and λ= 276 nm).

Table.5 Freundlich isotherm for the adsorption of Diclofenac sodium on Zeolite using UV-visible spectroscopy at T= 25˚C, pH= 6, contact time= 80 minutes, adsorbent dosage = 1.0g

log Ce	**log x/m**
0.653	0.74
1.11	0.911
1.45	1.062
1.54	1.862

Referring to Diclofenac sodium structure in figure.2, it is an acidic drug as Ibuprofen with pka of 4.0, its removal affected by pH should be similar to Ibuprofen (increase in percentage removal with decreases pH), however contrary results were obtained in which percentage removal increases with increasing pH. This suggests that there is another interaction mechanism between Diclofenac souduim and Zeolite other than electrostatic interaction, this trend is shown in Figure.14.

As adsorption time increases, the removal of Diclofenac sodium was found to increase and attained equilibrium at 40 minutes, as shown in Figure.15.

The percentage removal of Diclofenac sodium increased with increase of adsorbent dosage and attained equilibrium at 1.5 g as shown in Figure.16. It was found that the percentage removal of Diclofenac sodium decrease with increasing the concentration of Diclofenac sodium as shown in Figure.17.

As shown in Figure.18 and table.4, a plot of Ce\qe versus Ce was linear and the constants Qmax and K were determined from the slope and the intercept of the plot, which was found to be 4.75 mg\g and 0.047 at 25˚C and pH= 6 with 1.5 g adsorbent dosage. The correlation coefficient obtained with the Langmuir equation was high R^2= 0.992, which indicated a good fit with Langmuir isotherm as shown in Figure.18 and table.4. A plot of log x\m versus log C_e was nonlinear which indicates that the Freundlich isotherm has bad fit between parameters as shown in Figure.19 and table.5.

Removal of Diclofenac sodium in this study was compared with the other studies. In our study, the best removal of Diclofenac sodium on natural Jordanian zeolite was found to be 30.1% at pH=6, after contact time 80 minutes, adsorbent dosage of 1.5 g and initial Diclofenac sodium concentration 10 ppm with adsorption capacity for natural Jordanian zeolite of 4.75 mg\g.

Dai, C. M., etal. (2011), studied selective removal of Diclofenac sodium from contaminated water using molecularly imprinted polymer microspheres, and found that the best removal of Diclofenac sodium was at pH=3-9 with (97.6%). %,

Vieno, N., etal. (2006), studied removal of Diclofenac sodium in drinking water treatment and found that the best removal of Diclofenac sodium by ferric sulphate coagulation is at pH= 4.5 - 6 with 77 % removal (Vieno et al, 2006).

Rigobello, E. S., et al. (2013), studied removal of Diclofenac sodium by conventional drinking water treatment processes and granular activated carbon filtration. According to this study there was no removal of Diclofenac sodium in coagulation with aluminum sulfate at pH= 6.5, while in the treatment with pre-oxidation and disinfection, Diclofenac sodium was partially removed (Rigobello et al, 2013).

Simazaki, D., et al. (2008) studied removal of selected pharmaceuticals by chlorination, coagulation–sedimentation and powdered activated carbon treatment, the best removal of Diclofenac sodium by by chlorination was at 80 %. (Simazaki et al, 2008).

4.1.1.3 Indomethacin

4.1.1.3.1Calibration curve for Indomethacin using UV-visible spectrophotometer

The calibration curve was obtained by plotting absorbance versus concentration of Indomethacin and is displayed in figure .20. The Figure shows excellent linearity in the range of 10 -50 mg $L^{-1}$with correlation coefficient (R^2) of 0.9948, which indicates that the method used is linear.

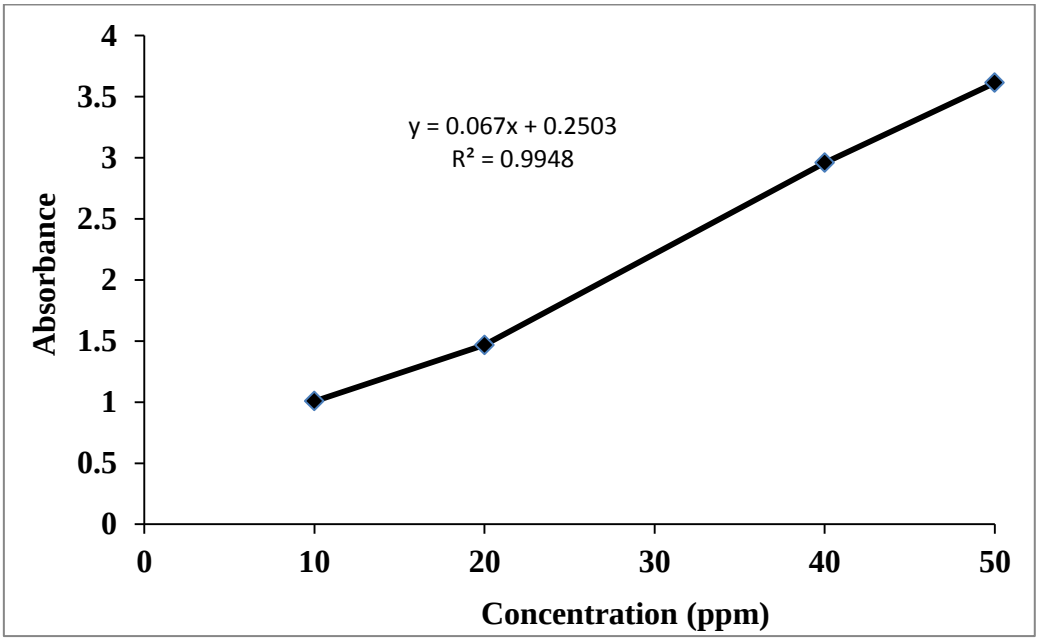

Figure.20 Calibration curve of Indomethacin obtained by UV-visible spectrophotometer at λ= 225 nm.

4.1.1.3.2Effects of different parameters on adsorption of Indomethacin

The adsorption of Indomethacin was performed at different parameters pH values in the range of (2 – 8), contact time at different time's intervals (10-120 minutes), adsorbent dosage from (0.5 - 2.0 g), initial Indomethacin concentration from (10 -50 ppm), to model the adsorption behavior of Indomethacin on zeolite adsorbent. Two adsorption isotherms were studied and their correlation with experimental data was assessed. The following figures (21-25) shows the percentage removal of Indomethacin at different parameters and Langmuir isotherm for the adsorption of Indomethacin on natural Jordanian zeolite .

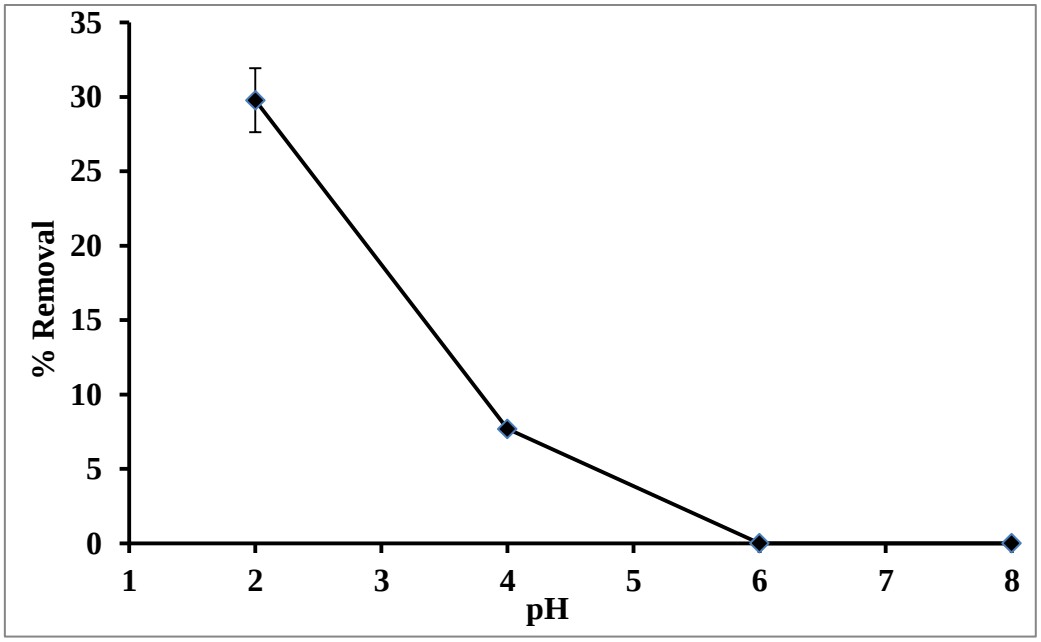

Figure.21 Percentage removal of indomethacin on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).

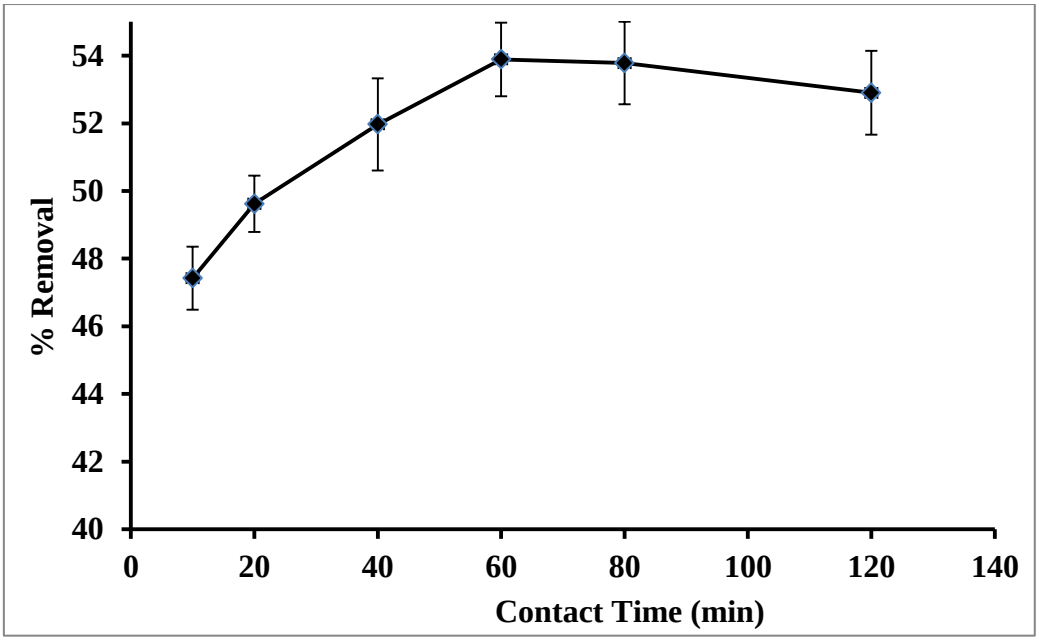

Figure.22 Percentage removal of Indomethacin on zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).

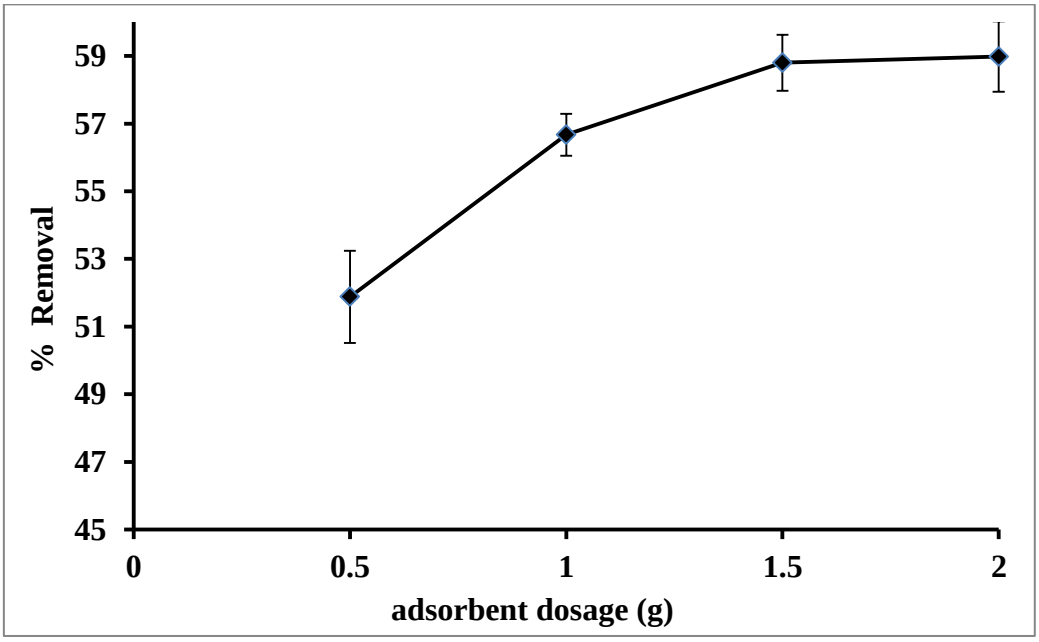

Figure.23 Percentage removal of Indomethacin on zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 40 ppm, contact time= 60 minutes and λ= 225 nm).

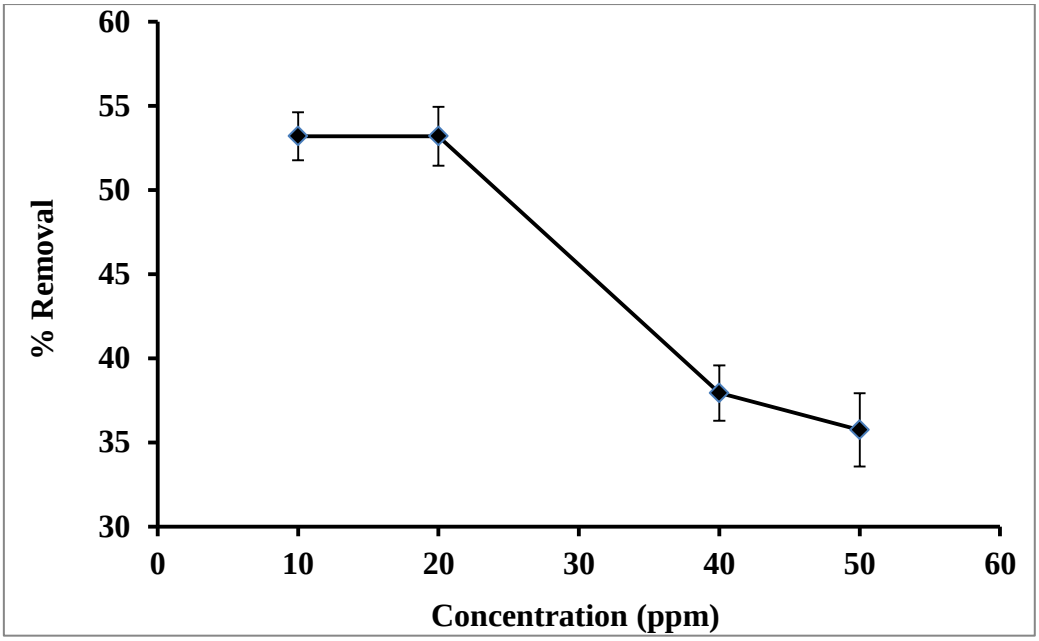

Figure.24 Percentage removal of Indomethacin on zeolite as a function of initial Indomethacin concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 60 minutes and λ= 225 nm).

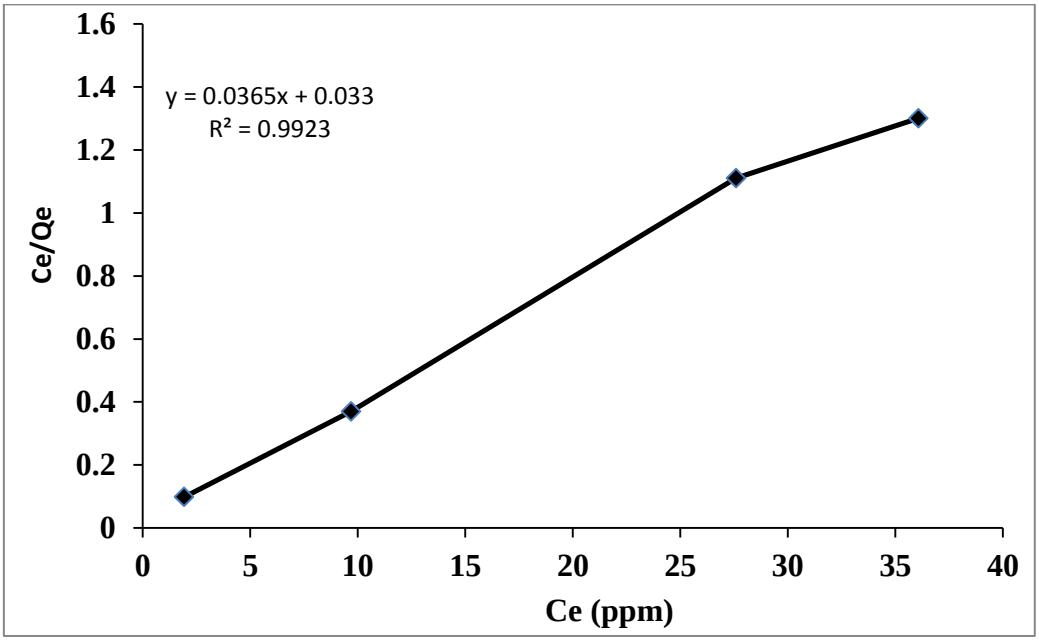

Figure.25 Langmiur isotherm for the adsorption of Indomethacin on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 60 minutes, adsorbent dosage = 1.0 g and λ= 225 nm).

Table.6 Langmiur isotherm for the adsorption of Indomethacin on Zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 2, contact time= 60 minutes, adsorbent dosage = 1.0 g).

Conc (T=60) (Ce)(mg L-1)	Q=(M Initial-M Final/0.5)	Ce/Qe
1.915	16.17	0.098
9.686	26.36	0.37
27.592	24.816	1.11
36.08	27.84	1.3

Referring to indomethacin structure in figure.3, it has acidic nature with pka of 3.8, same trend was observed as for Ibuprofen in which percentage removal decreases with increasing pH (shown in Figure.21).

It was found that percentage removal of Indomethacin increase with increasing time, and attained equilibrium at 60 minutes (Shown in Figure.22). The percentage removal of Indomethacin increased with increase of adsorbent dosage, and attained equilibrium at (1.0 – 1.5 g), as shown in Figure.23. This increase in removal is due to the availability of more adsorbent and so effectively a large surface area for Indomethacin to be attached, the optimum adsorbent dosage is 1.5 g in which the percentage removal reached at maximum value of 59.0 %. The percentage removal of Indomethacin was found to decrease with increasing concentration, this trend is shown in Figure.24.

As shown in Figure.25 and table.6, a plot of Ce\qe versus Ce was linear with R^2 of 0.9923 and the constants Q max and K were determined from the slope and the intercept of the plot, which was found to be 26.60 mg\g and 1.14 at 25˚C and pH= 2 with 1.5 g adsorbent dosage. The correlation coefficient obtained with the Langmuir equation was high (R^2= 0.9923), which indicated a good fit with Langmuir isotherm while Freundlich isotherm was found to be not fit with adsorption of indomethacin.

Removal of Indomethacin in this study was compared with other studies. In our study, the best removal of Indomethacin on natural Jordanian zeolite was found at pH=2, after contact time of 80 minutes, with adsorbent dosage of 1.5g and initial Indomethacin concentration of 10 ppm and was found to be 59.0%, with adsorption capacity of 26.60 mg\g.

Simazaki, D., et al. (2008), studied removal of Indomethacin by chlorination, coagulation–sedimentation and powdered activated carbon treatment. According to this study, Indomethacin was completely degraded by chlorination, (Simazaki et al, 2008).

4.1.2 Basic pharmaceuticals

4.1.2.1 chlorpheniramine maleate

4.1.2.1.1Calibration curve for chlorpheniramine maleate using UV-visible spectrophotometer

The calibration curve was obtained by plotting absorption versus concentration of Chlorpheniramine maleate and is displayed in Figure.26. The Figure shows excellent linearity in the range 10 -50 ppm with correlation coefficient of (R^2) = 0.9999, which indicates that the method used is linear.

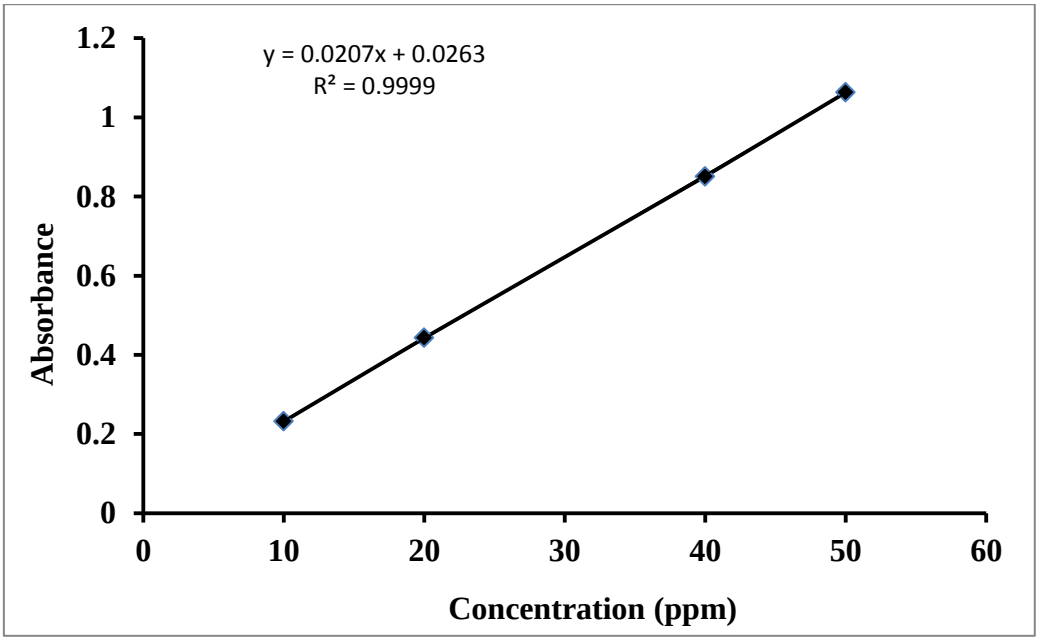

Figure.26 Calibration curve of chlorpheniramine maleate obtained by UV-visible spectrophotometer at λ= 261 nm.

4.1.2.1.2Effects of different parameters on adsorption of chlorpheniramine maleate

The adsorption of chlorpheniramine maleate was performed at different parameters pH values in the range of (2 – 8), contact time at different time's intervals (10-120 minutes), adsorbent

dosage from (0.5 - 2.0 g) and initial chlorpheniramine maleate concentration from (10 -50 ppm), to model the adsorption behavior of chlorpheniramine maleate on natural Jordanian zeolite adsorbent. Two adsorption isotherms were studied and their correlation with experimental data was assessed. Figures 27-31 show percentage removal of chlorpheniramine maleate at different parameters and Freundlich isotherm for the adsorption of chlorpheniramine maleate on natural Jordanian zeolite .

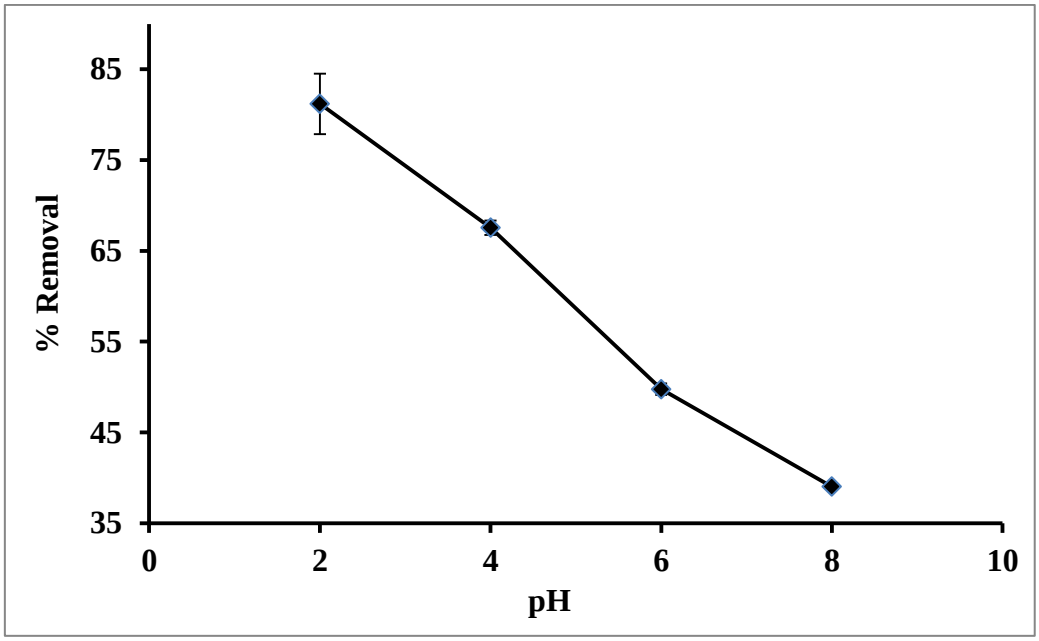

Figure.27 Percentage removal of chlorophenarimine maleate on Zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 261 nm).

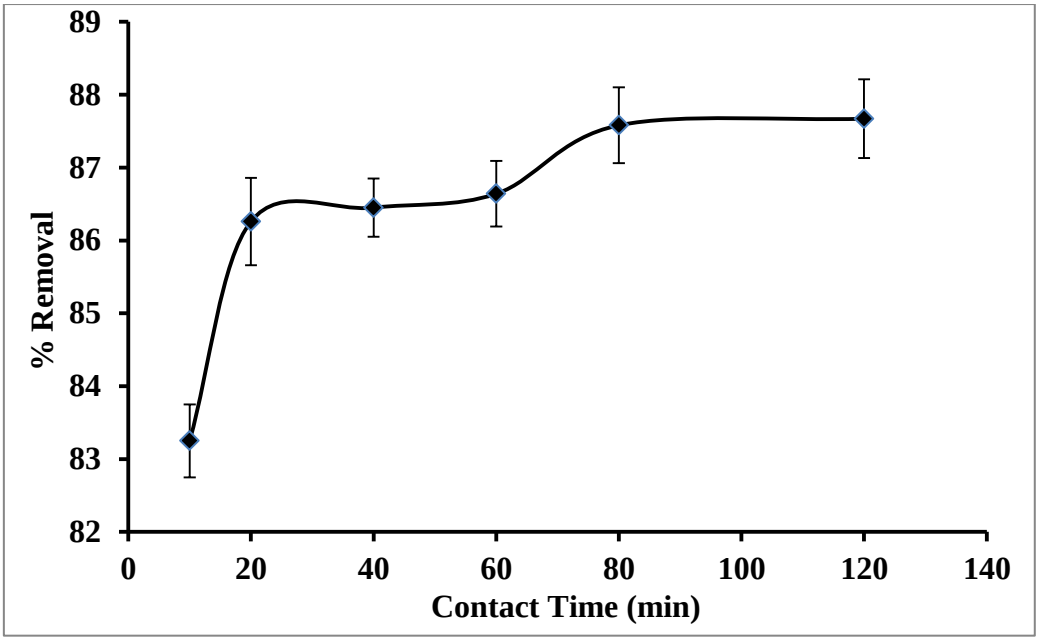

Figure.28 Percentage removal of chlorophenarimine maleate on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 261 nm).

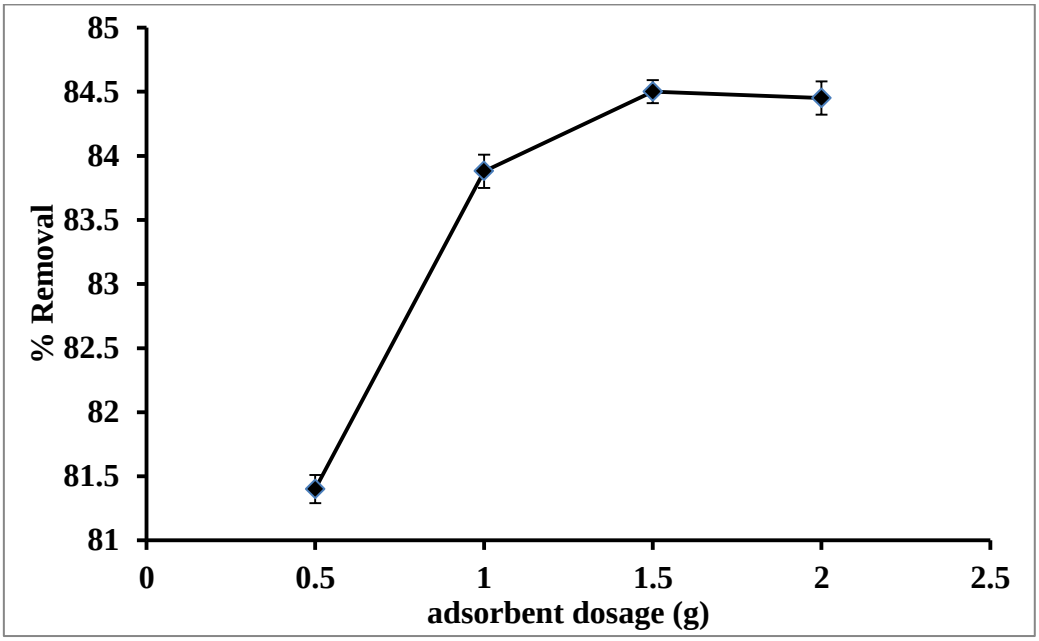

Figure.29 Percentage removal of Chlorophenarimine maleate on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 261 nm).

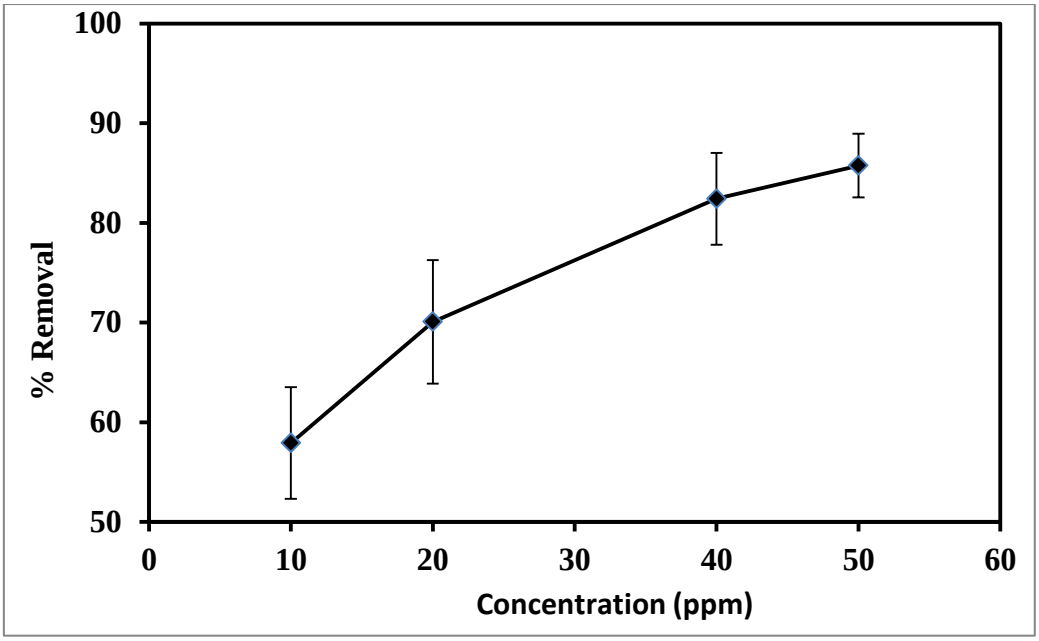

Figure.30 Percentage removal of Chlorophenarimine maleate on Zeolite as a function of initial Chlorophenarimine maleate concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.5 g, contact time= 80 minutes and λ= 261 nm).

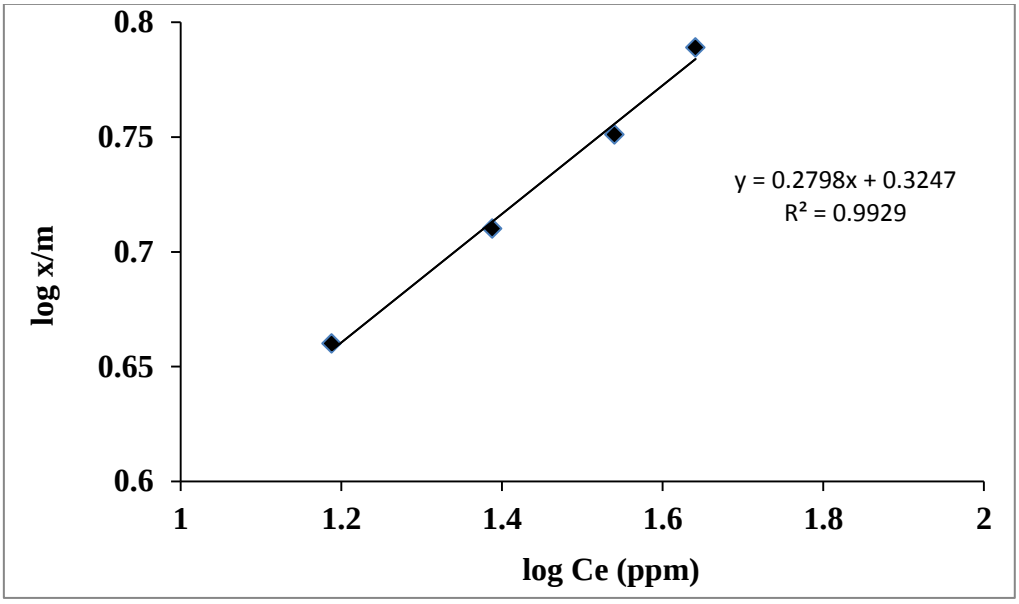

Figure.31 Freundlich isotherm for the adsorption of Chlorophenarimine maleate on Zeolite usingUV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g and λ= 261 nm).

Table.7 Freundlich isotherm for the adsorption of Chlorophenarimine maleate on Zeolite usingUV-visible spectroscopy at T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0g

log x/m	log Ce
1.188	0.66
1.388	0.71
1.54	0.75
1.641	0.79

Result showed that highest removal was obtained at low pH (e.g pH=2) and percentage removal increases with decreasing of pH (Figure.27). Chlorophenarimine maleate is basic drug with pka of 9.13, so it is expected that it bears positive charge at low pH (e.g pH=2) and so characteristics interaction with Zeolite increases, and this explains the increase in removal of Chlorophenarimine maleate with decrease of pH.

As shown in figure.28, as the adsorption time increases, percentage removal of Chlorophenarimine maleate increases up to 60 minutes and attained equilibrium at 80 – 120 minute, which indicated that the adsorption reached saturation.

Percentage removal of Chlorophenarimine maleate was found to increase with increasing of adsorbent dosage at (0.5–1.5 g), and reached equilibrium at 1.5 g to 2.0 g of adsorbent as shown in Figure.29. This increase in removal is due to the availability of more adsorbent and so effectively a large surface area for Chlorophenarimine maleate to be attached. Optimum adsorbent dosage is 1.5 g in which the percentage removal reached at maximum value of 84.5 %.

Figure.30 shows that the percentage removal of Chlorophenarimine maleate increase with increase in the concentration of Chlorophenarimine maleate solution. The highest removal (85.8%) occurs when the concentration of Chlorophenarimine maleate solution is 50 ppm.

A plot of Ce\qe versus Ce was nonlinear which indicates that the Langmuir isotherm has bad fit between parameters. On the other hand, a plot of log x\m versus log Ce (Figure.31 and Table.7) is linear. The constants k_f and n were determined from the slope and the intercept of the plot, which was found to be 2.11 mg\g and 3.57at 25˚C and pH= 2 with 1.0 g adsorbent

dosage. Correlation coefficient of 0.9929 indicating that Freundlich isotherm model is the best to describe removal process of Chlorophenarimin maleate on Zeolite, This trend is shown in Figure.31 and table.7.

The best removal of Chlorophenarimin maleate on natural Jordanian zeolite at pH=2, after contact time of 80 minutes, adsorbent dosage of 1.5g and initial Chlorophenarimine maleate concentration 50 ppm was found to be 85.8%.

4.1.2.2 Paracetamol

4.1.2.2.1 Calibration curve for Paracetamol using UV-visible spectrophotometer

The calibration curve was obtained by plotting absorbance versus concentration of Paracetamol and is displayed in Figure.32. The Figure shows excellent linearity in the range of 10 -50 ppm with correlation coefficient (R^2) of 0.999.

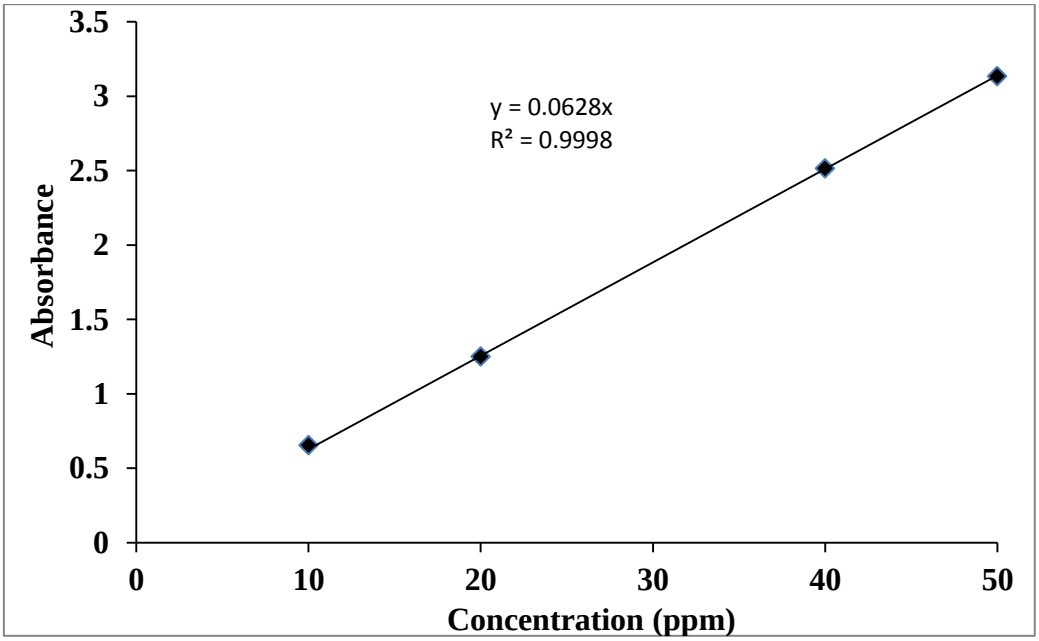

Figure.32 Calibration curve of Paracetamol obtained by UV-visible spectrophotometer at λ= 243 nm.

4.1.2.2.2Effects of different parameters on adsorption of pracetamol

The adsorption of pracetamol was performed at different parameters pH values in the range of (2 – 8), contact time at different time's intervals (10-120 minutes), adsorbent dosage from

(0.5 - 2.0 g) and initial pracetamol concentration from (10 -50 ppm), to model the adsorption behavior of pracetamol on zeolite adsorbent. Two adsorption isotherms were studied and their correlation with experimental data was assessed. Figures (33-37) show percentage removal of pracetamol at different parameters and Langmuir isotherm for the adsorption of pracetamol on natural Jordanian zeolite.

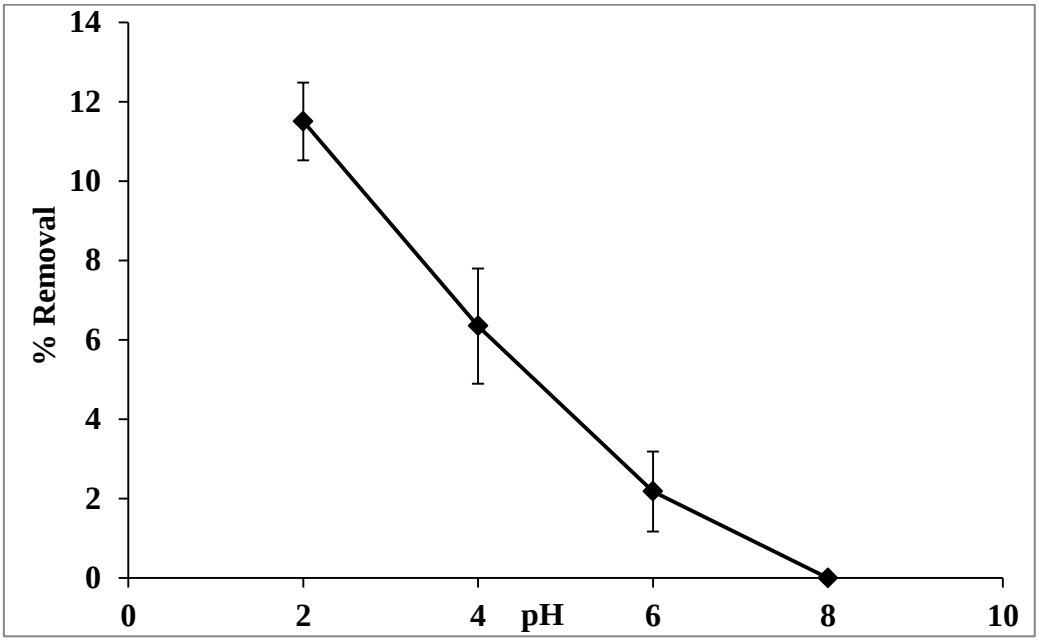

Figure.33 Percentage removal of Paracetamol on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).

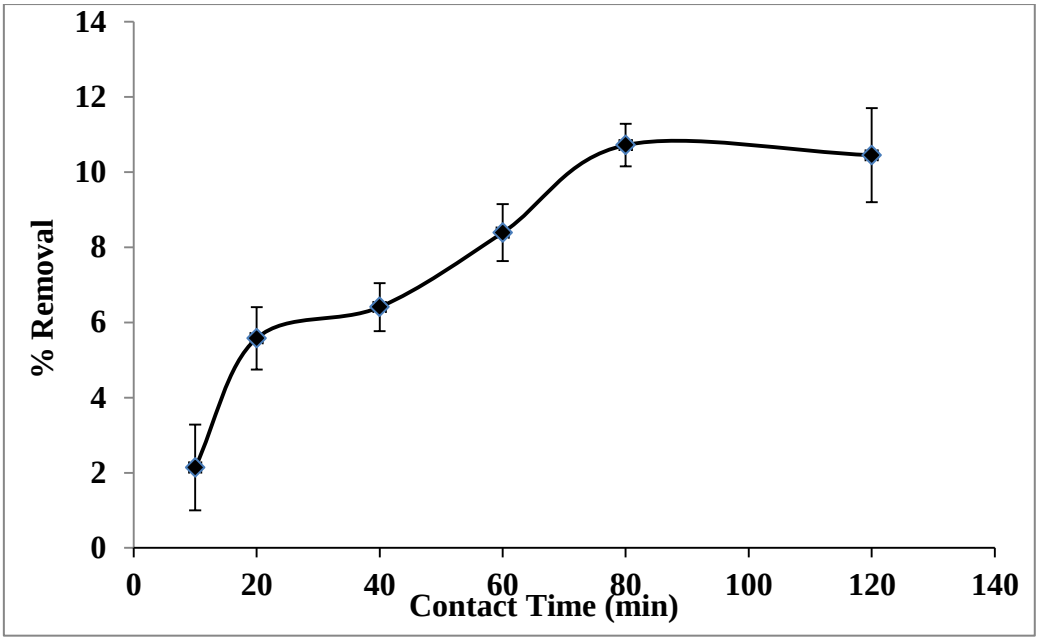

Figure.34 percentage removal of Paracetamol on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).

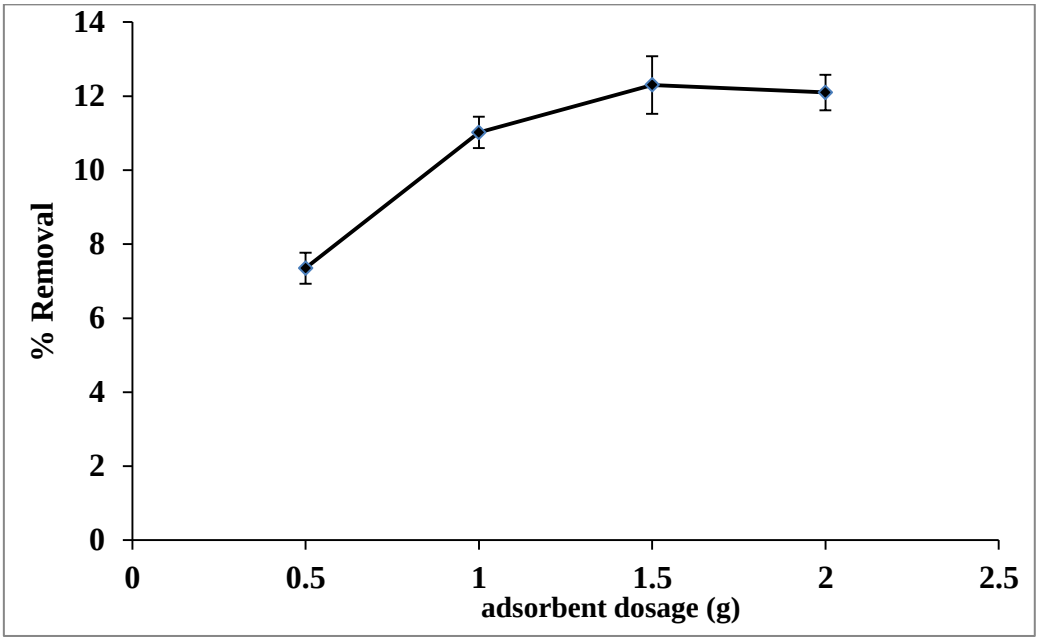

Figure.35 Percentage removal of Paracetamol on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial, conc. = 50 ppm, contact time= 80 minutes and λ= 243 nm).

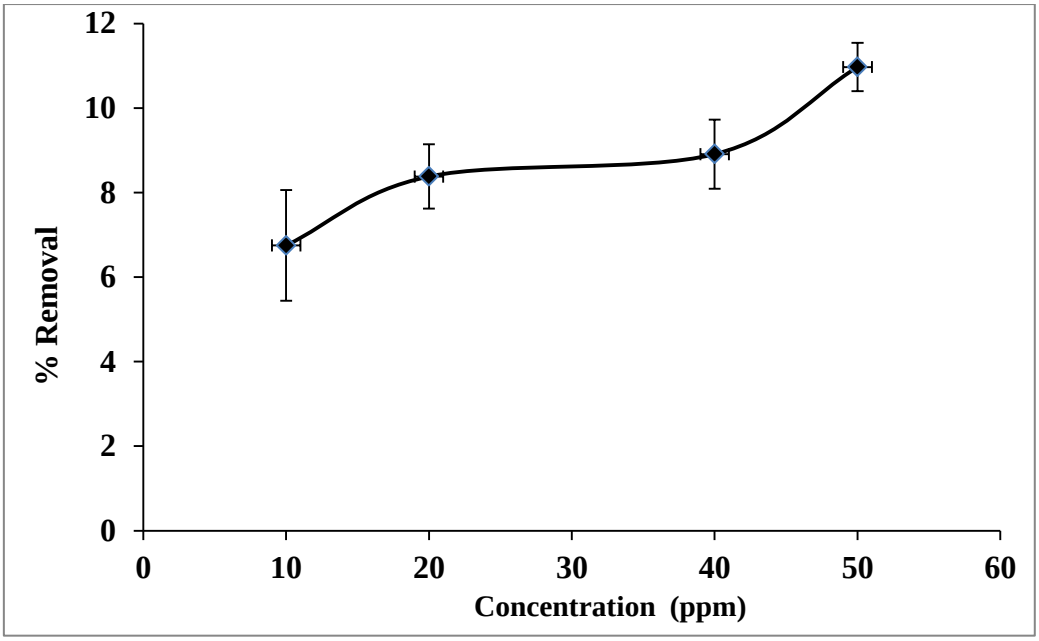

Figure.36 Percentage removal of Paracetamol on zeolite as a function of initial Paracetamol concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 243 nm).

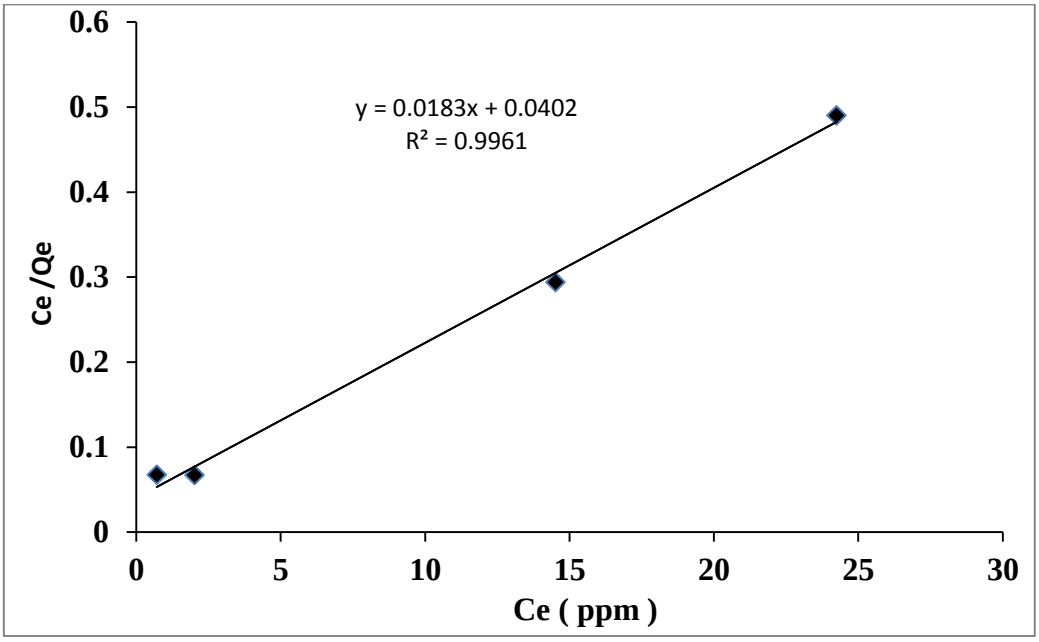

Figure.37 Langmiur isotherm for the adsorption of Paracetamol on Zeolite using UV-visible spectrophotometer at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0 g and λ= 243 nm).

Table.8 Langmiur isotherm for the adsorption of Paracetamol on zeolite using UV-visible spectroscopy at (T= 25˚C, pH= 2, contact time= 80 minutes, adsorbent dosage = 1.0 g).

Conc (T=80) (Ce)(mg L^{-1})	Q=(MInitial-M Final/0.5	Ce/Qe
0.7084	8.584	0.0725
2.0244	35.952	0.066
14.52	50.96	0.284
24.25	51.5	0.49

The effect of pH on Paracetamol removal was found to be as Ibuprofen, Indomethacin and Chloropheniramine maleate where highest removal was obtained at low pH and % emoval increases with decreasing of pH. This trend is shown in Figure.33.

Figure.34 show that percentage removal of paracetamol increases as the adsorption time increases from 10 to 80 minutes and attained equilibrium after 80 minutes, which indicated that adsorption reached saturation. Therefore, the adsorption time was set at 80 minutes.

The percentage removal of paracetamol was found to increase with increasing of adsorbent dosage at (0.5 – 1.5 g) as shown in Figure.35. This increase in removal is due to the availability of more adsorbent and so effectively a large surface area for paracetamol to be attached. Optimum adsorbent dosage is 1.5 g in which the percentage removal reached at maximum value of 12.7 %.

Figure.36 shows that the percentage removal of Paracetamol increase with increasing of the concentration of Paracetamol. The highest removal (11.0%) occurs when the concentration of Paracetamol is 50 ppm.

A plot of Ce\qe versus Ce was linear and the constants Qmax and K were determined from the slope and the intercept of the plot, which was found to be 55.6 mg\g and 0.44 at 25˚C and pH= 2 with 1.0 g adsorbent dosage. The correlation coefficient obtained with Langmiur equation was high (R^2= 0.992), which indicated a good fit with Langmiur isotherm (Table.8 and figure.37.

It was interesting to compare removal of paracetamol in this study with other studies. In this study, the best removal of Paracetamol on natural Jordanian zeolite at pH=2, after contact time

of 80 minutes, adsorbent dosage of 2.0 g and initial Paracetamol concentration of 50 ppm is (12.7%), with adsorption capacity of 55.6 mg\g.

Karaman, R., et al. (2016), studied Paracetamol biodegradation by activated sludge and photocatalysis and its removal by a micelle–clay complex, activated charcoal, and reverse osmosis membranes. The ability of bench top reverse osmosis (RO) plant as well as advanced membrane pilot plant to remove paracetamol was also studied at different water matrixes to test the effect of organic matter composition showed that at least 90% rejection was obtained(Karaman et al, 2016) ,

Ayyash, F., et al (2002), studied removal of Aspirin, Salicylic Acid, Paracetamol and p-Aminophenol by Advanced Membrane Technology Activated Charcoal and Clay Micelles Complex, The performance of Al-Quds University wastewater treatment plant has shown complete removal of Paracetamol from spiked wastewater (Ayyash et al, 2002).

4.1.3Comparison between Percentage removal of the selected pharmaceuticals.

Table.9 shows the percentage removal of the studied pharmaceuticals at optimum pH. As it is show in this table, high removal was obtained for Ibuprofen, Chlorophenarimin maleate, low for Paracetamol, and diclofenac sodium and intermediate for Indomethacin.

Table.9 Comparison between removal and optimum pH for the selected pharmaceuticals investigated in this study.

pharmaceutical	Percentage removal	pH
Ibuprofen	88.3%	**2**
Diclofenac sodium	30.1%	**6**
Indomethacin	58.9%	**2**
Chlorophenarimin maleate	85.8%	**2**
Paracetamol	12.7%	**2**

4.2Column Experiments

Column experiments of Ibuprofen, Diclofenac Sodium, Indomethacin, Chlorophenarmin maleate and Paracetamol were performed by passing a solution of each drug at 50 ppm concentration through the column filled with 9/1 (w/w) mixture of quartz sand and zeolite at flow rate of 2 ml.min^{-1}. Eluted fractions of 100 mL (each) were collected at chosen times. Figures. (38-41) show percentage removal of each drug versus fractions.

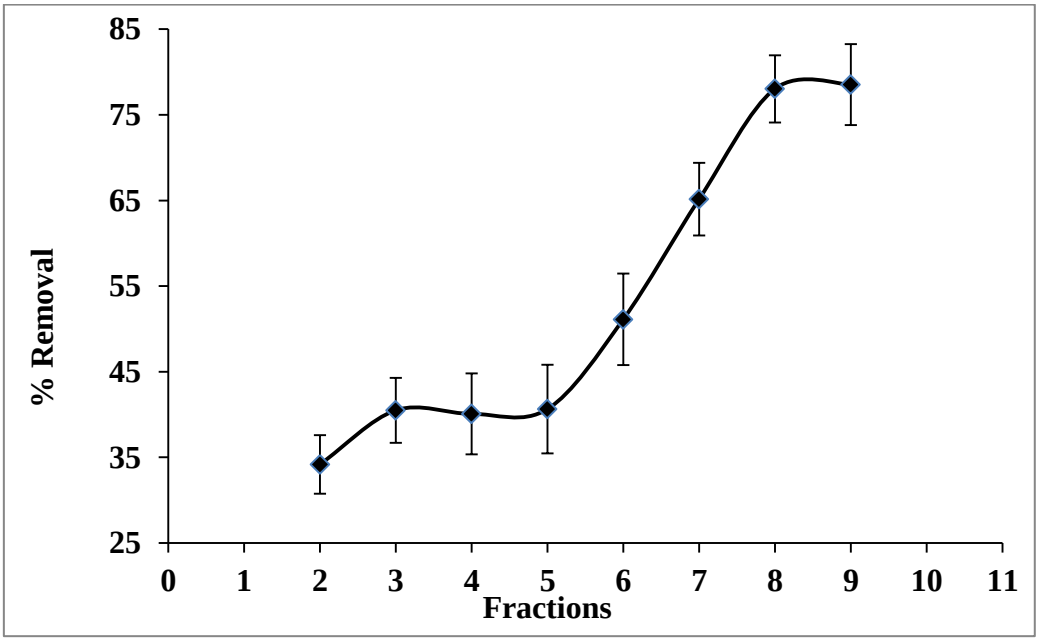

Figure.38 Percentage removal of Ibuprofen on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH =2, concentration of Ibuprofen 50 ppm and λ= 224 nm).

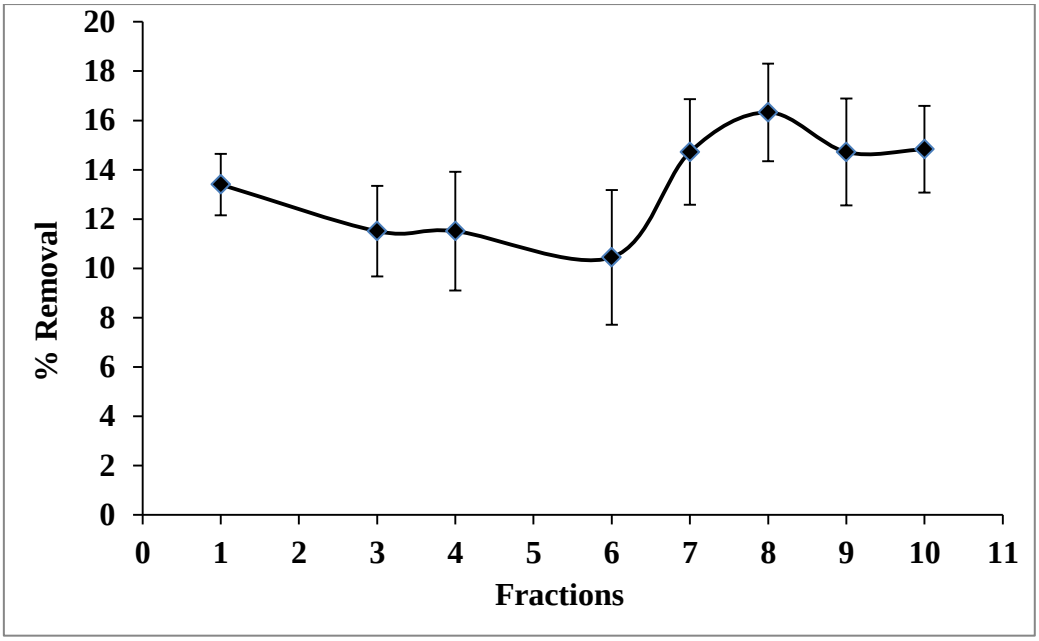

Figure.39 Percentage removal of Diclofenac sodium on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH =6, concentration of Diclofenac sodium 50 ppm and λ= 276 nm).

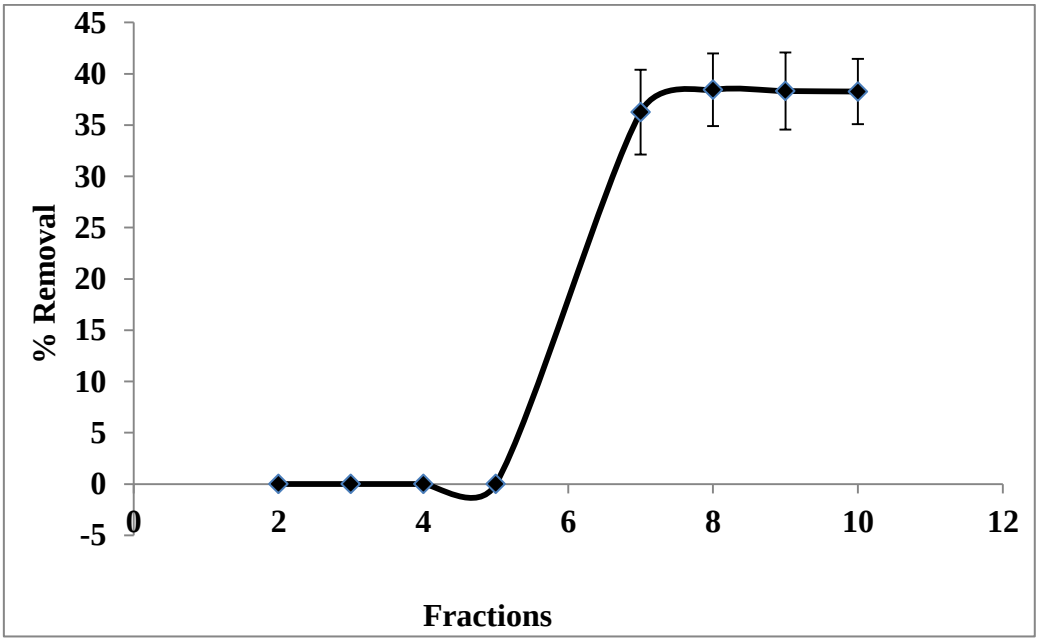

Figure.40 Percentage removal of chlorophenarimine maleate on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH=2, concentration of chlorophenarimine maleate 50 ppm and λ= 261 nm).

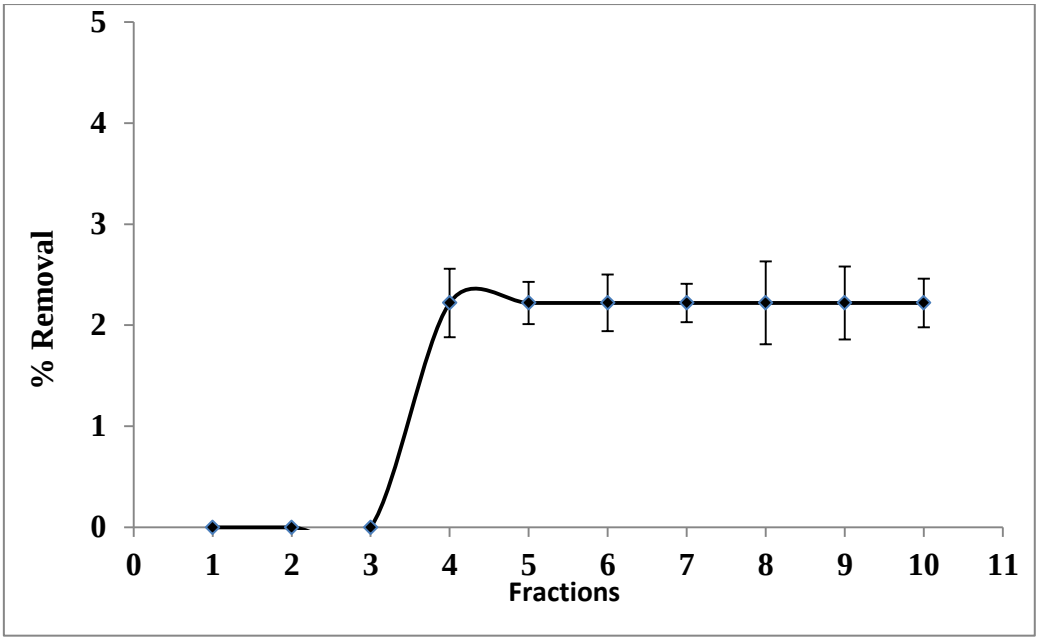

Figure.41 Percentage removal of Paracetamol on zeolite as a function of different fractions by UV-visible spectrophotometer at (T= 25˚C, pH =2, concentration of Paracetamol 50 ppm and λ= 243 nm).

Results of column experiment of pharmaceutical removal showed that the best removal of Ibuprofen on Zeolite was found to be for eluted fraction number 9 and 10 which is about 78 % as shown in Figure.38, while removal of Diclofenac sodium on zeolite at eluted fraction number 8 which is 16.33% as shown in Figure.39. As shown in figure 40, removal of Chlorophenarimine maleate on zeolite was found at eluted fraction number 8 which is 38.4% . Removal of Paracetamol on zeolite was found at eluted fractions number 4-10 which is 2.2% as shown in Figure.41.

Percentage removal of drugs by Batch experiment was compared with that using column experiment, and results showed that the best removal of Ibuprofen from by Zeolite in batch experiment was 88.3% compared with that of column experiment which was 78.04%.

The best removal of Diclofenac sodium by Zeolite in batch experiment was 30.1% compared with column experiment which was 16.33%.

The best removal of Chlorophenarimine maleate by Zeolite in batch experiment was 85.8% compared with that using column experiment which was 38.4%. This indicates that the batch

experiment is better than column experiment for removal of Chlorophenarimine maleate. For Paracetamol removal by Zeolite in batch experiment which was 12.7% compared with column experiment with percentage removal of 2.22%.

CHAPTER FIVE

Conclusion

CONCLUSION

Natural Jordanian Zeolite is effective for removal of Ibuprofen, Indomethacin, Diclofenac sodium and Chloropheniramine maleate from aqueous solution as natural Jordanian zeolite is characterized by large surface area, micro-porous nature, high adsorption capacity, and easy availability. But it was found not effective for removal of Paracetamol.

The effect of pH, adsorbent dosage, and initial concentration of pharmaceuticals on the adsorption process was studied. The optimum pH for all pharmaceuticals was found to be 2 except for Diclofenac sodium where its optimum pH was 6.

The optimum adsorbent dosage was 1.0g for Ibuprofen, 2.0g for Diclofenac Sodium and for Indomethacin, 1.5g for Chloropheniramine maleate and for Paracetamol.

The optimum initial concentration of pharmaceuticals on the adsorption process was 50 ppm for Ibuprofen, Chloropheniramine maleate and for Paracetamol, 10 ppm for Diclofenac Sodium and for Indomethacin.

Diclofenac Sodium, Indomethacin and paracetamol have a good fit with Langmuir isotherm with adsorption capacity 4.8mg\g, 26.6mg\g and 55.6mg\g respectively. Ibuprofen and Chloropheniramine maleate was found to fit with Freundlich isotherm by Zeolite with adsorption capacity of 1.23mg\g and 2.11mg\g, respectively.

Difference between Column experiment and batch experiment is not big which mean that the Column experiment agrees with batch experiment except for Chloropheniramine maleate. Percentage removal of Ibuprofen, Diclofenac Soduim, Pracetamol and Chloropheniramine maleate by Zeolite in batch experiment was 88.3%, 30.1%, 12.7% and 85.8%, respectively, and percentage removal of Ibuprofen, Diclofenac Soduim, Pracetamol and chloropheniramine maleate by Zeolite in column experiment was 78.04%, 22.47%, 2.22% and 38.4%, respectively.

CHAPTER SIX

References

References

Aliewi, A.**(2007**), Water Resourses in Palestine, House of Water and Environment.

Ayyash, F., Khamis, M., Khalaf, S., Thawabteh, A., & Karaman, R.**(2002)**, Removal of Aspirin, Salicylic Acid, Paracetamol and p-Aminophenol by Advanced Membrane technology Activated Charcoal and Clay Micelles Complex.

Baker, H. M., Massadeh, A. M., & Younes, H. A. **(2009).** Natural Jordanian zeolite: removal of heavy metal ions from water samples using column and batch methods. Environmental Monitoring and Assessment, 157(1-4), 319-330.

Behera, S. K., Oh, S. Y., & Park, H. S. **(2012).** Sorptive removal of ibuprofen from water using selected soil minerals and activated carbon. International Journal of Environmental Science and Technology, 9(1), 85-94.

Beltran, A., Marcé, R. M., Cormack, P. A. G., & Borrull, F. **(2009).** Synthesis by precipitation polymerisation of molecularly imprinted polymer microspheres for the selective extraction of carbamazepine and oxcarbazepine from human urine. Journal of Chromatography A, 1216(12), 2248-2253.

Cejka, J. **(2005)**. Zeolites and ordered mesoporous materials: progress and prospects: the 1st FEZA School on Zeolites, Prague, Czech Republic, and August 20-21, 2005 (Vol. 157). Gulf Professional Publishing.

Dai, C. M., Geissen, S. U., Zhang, Y. L., Zhang, Y. J., & Zhou, X. F. **(2011)**. Selective removal of diclofenac from contaminated water using molecularly imprinted polymer microspheres. Environmental Pollution, 159(6), 1660-1666.

Derouane, E. G. **(1984)**. Zeolites: Science and Technology, ed. FR Ribeiro, AE Rodrigues, LD Rollmann and C.Naccache, NATO ASI Series.

Guisnet, M., & Gilson, J. P. (Eds.). **(2002)**. Zeolites for cleaner technologies (Vol. 3). World Scientific.

Fent, K., Weston, A. A., & Caminada, D. **(2006)**. Ecotoxicology of human pharmaceuticals. Aquatic Toxicology, 76(2), 122-159.

Foye, W. O. **(2008)**. Foye's principles of medicinal chemistry. T. L. Lemke, & D. A. Williams (Eds.). Lippincott Williams & Wilkins.

Grosvenor, A. P., Kobe, B. A., Biesinger, M. C., & McIntyre, N. S. **(2004)**. Investigation of multiplet splitting of Fe 2p XPS spectra and bonding in iron compounds. Surface and Interface Analysis, 36(12), 1564-1574.

H. Jones, O. A., Voulvoulis, N., & Lester, J. N. **(2005)**. Human pharmaceuticals in wastewater treatment processes. Critical Reviews in Environmental Science and Technology, 35(4), 401-427.

Henschel, K. P., Wenzel, A., Diedrich, M., & Fliedner, A. **(1997)**. Environmental hazard assessment of pharmaceuticals. Regulatory Toxicology and Pharmacology, 25(3), 220-225.

Karaman, R., Khamis, M., Abbadi, J., Amro, A., Qurie, M., Ayyad, I& Bufo, S. A. (2016). Paracetamol biodegradation by activated sludge and photocatalysis and its removal by a micelle–clay complex, activated charcoal, and reverse osmosis membranes. Environmental technology, 37(19), 2414-2427.

Khalaf, S., Al-Rimawi, F., Khamis, M., Zimmerman, D., Shuali, U., Nir, S., & Karaman, R. **(2013)**. Efficiency of advanced wastewater treatment plant system and laboratory-scale micelle-clay filtration for the removal of ibuprofen residues. Journal of Environmental Science and Health, Part B, 48(9), 814-821.

Khoury, H. N., & El-Sakka, W. **(1986)**. Mineralogical and industrial characterization of the Batn El-Ghoul clay deposits, southern Jordan. Applied clay science, 1(4).

Kim, S. D., Cho, J., Kim, I. S., Vanderford, B. J., & Snyder, S. A. **(2007)**. Occurrence and removal of pharmaceuticals and endocrine disruptors in South Korean surface, drinking, and waste waters. Water research, 41(5), 1013-1021.

Krishna, R. H., & Swamy, A. V. V. S. **(2012)**. Investigation on the effect of particle size and adsorption kinetics to removal of hexavalent chromium from the aqueous solutions using low cost sorbent. European Chemical Bulletin, 1(7), 258-262.

Leake, J. R., Duran, A. L., Hardy, K. E., Johnson, I., Beerling, D. J., Banwart, S. A., & Smits, M. M. **(2008)**. Biological weathering in soil: the role of symbiotic root-associated fungi biosensing minerals and directing photosynthate-energy into grain-scale mineral weathering. Mineralogical Magazine, 72(1), 85-89.

Mara, D. D. **(2003)**. Water, sanitation and hygiene for the health of developing nations. Public health, 117(6), 452-456.

Mompelat, S., Le Bot, B., & Thomas, O. **(2009)**. Occurrence and fate of pharmaceutical products and by-products, from resource to drinking water. Environment international, 35(5), 803-814.

Nazer, D. W. **(2010)**. From water scarcity to sustainable water use in the West Bank, Palestine. CRC Press.

Pharmacopoeia, B. (2004). Specific monograph: London: British Pharmacopoeia Commission.

Rigobello, E. S., Dantas, A. D. B., Di Bernardo, L., & Vieira, E. M. **(2013)**. Removal of diclofenac by conventional drinking water treatment processes and granular activated carbon filtration. Chemosphere, 92(2), 184-191.

Scheurell, M., Franke, S., Shah, R. M., & Hühnerfuss, H. **(2009)**. Occurrence of diclofenac and its metabolites in surface water and effluent samples from Karachi, Pakistan. Chemosphere, 77(6), 870-876.

Simazaki, D., Fujiwara, J., Manabe, S., Matsuda, M., Asami, M., & Kunikane, S. **(2008)**. Removal of selected pharmaceuticals by chlorination, coagulation–sedimentation and powdered activated carbon treatment. Water Science and Technology, 58(5), 1129-1135.

Ternes, T. A., Meisenheimer, M., McDowell, D., Sacher, F., Brauch, H. J., Haist-Gulde, B., & Zulei-Seibert, N. **(2002)**. Removal of pharmaceuticals during drinking water treatment. Environmental science & technology, 36(17), 3855-3863.

Townsend, R. P., & Coker, E. N. **(2001)**. Ion exchange in zeolites. Studies in surface science and catalysis, 137, 467-524.

Vieno, N. M., Tuhkanen, T., & Kronberg, L. **(2005)**. Seasonal variation in the occurrence of pharmaceuticals in effluents from a sewage treatment plant and in the recipient water. Environmental science & Technology, 39(21), 8220-8226.

Vieno, N., Tuhkanen, T., & Kronberg, L. **(2006)**. Removal of pharmaceuticals in drinking water treatment: effect of chemical coagulation. Environmental Technology, 27(2), 183-192.

Wang, X., Zhao, C., Zhao, P., Dou, P., Ding, Y., & Xu, P. **(2009)**. Gellan gel beads containing magnetic nanoparticles: an effective biosorbent for the removal of heavy metals from aqueous system. Bioresource technology, 100(7), 2301-2304.

World Health Organization. **(2001)**. International Classification of Functioning, Disability and Health: ICF. World Health Organization.

https://books.google.ps/books?hl=en&lr=&id=lMZPmEJrJ3sC&oi=fnd&pg=PA1&dq=World+Health+Organization.+(2001).+&ots=PFoMZya5Hf&sig=F9vr-JZC1m-07fqEmexWf-w3B1k&redir_esc=y#v=onepage&q=World%20Health%20Organization.%20(2001).&f=false

Zwiener, C., & Frimmel, F. H. **(2000)**. Oxidative treatment of pharmaceuticals in water. Water Research, 34(6), 1881-1885.

Appendices

Appendix.A: Percentage removal of ibuprofen on zeolite as a function of pH by uv-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and at and λ= 224 nm).

pH	% Removal
2	33.7 ± 2.1
4	11.70 ± 2.6
6	0.00
8	0.00

Appendix.B Percentage removal of ibuprofen on zeolite as a function of Contact time by uv-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 and λ= 224 nm).

time / min	% removal
10	74.8 ± 1.9
20	74.8 ± 2.1
40	74.8 ± 1.6
60	74.8 ± 1.3
80	74.8 ± 2.2

Appendix.C Percentage removal of ibuprofen on zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 224 nm).

wheight of Zeolite	% removal
0.5 gram	26.4 ± 4.2
1.0 gram	88.3 ± 5.3
1.5 gram	84.8 ± 3.2
2.0 gram	83.9 ± 3.9

Appendix.D Percentage removal of ibuprofen on zeolite as a function of initial Ibuprofen concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 224 nm).

Concentration /ppm	**% Removal**
10 ppm	20.2 ± 4.6
20 ppm	50.8 ± 3.2
40 ppm	66.5 ± 3.7
50 ppm	74.8 ± 4.3

Appendix.E Percentage Diclofenac sodium on zeolite as a function of pH by UV-visible spectrophotometer at T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 276 nm).

PH	**% Removal**
2	37.9 ± 1.45
4	49.1 ± 1.68
6	58.3 ± 1.75
8	55.9 ± 1.64

Appendix.F Percentage removal of Diclofenac sodium on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 276 nm).

time / min	**% removal**
10	24.7 ± 1.65
20	27.5 ± 1.04
40	28.9 ± 1.55
60	27.9 ± 1.14
80	27.9 ± 1.37
120	24.8 ± 1.43

Appendix.G Percentage removal of Diclofinac sodium on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 6, initial conc. = 50 ppm, contact time= 80 minutes and λ= 276 nm).

adsorbent dosage	**% Removal**
0.5	23.3 ± 0.41
1.0	26.1 ± 0.46
1.5	27.6 ± 0.63
2.0	27.8 ± 0.58

Appendix.H Percentage removal of Diclofenac sodium on zeolite as a function of initial Ibuprofen concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 6, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 276 nm).

Concentration /ppm	**% Removal**
10 ppm	30.1 ± 0.83
20 ppm	29.9 ± 0.68
40 ppm	26.5 ± 0.74
50 ppm	24.8 ± 0.54

Appendix.I Percentage removal of Indomethacin on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).

PH	% Removal
2	29.8 ± 2.15
4	7.7 ± 0.0
6	0
8	0

Appendix.J Percentage removal of Indomethacin on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 225 nm).

Time	% Removal
10	47.4 ± 0.93
20	49.6 ± 0.83
40	51.9 ± 1.36
60	53.9 ± 1.09
80	53.8 ± 1.22
120	52.9 ± 1.24

Appendix.K Percentage removal of Indomethacin on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 40 ppm, contact time= 60 minutes and λ= 225 nm).

Adsorbent dosage	% removal
0.5 g	51.9 ± 1.36
1.0 g	56.7 ± 0.62
1.5 g	58.8 ± 0.83
2.0 g	58.9 ± 1.04

Appendix.L Percentage removal of Indomethacin on zeolite as a function of initial Indomethacin concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 60 minutes and λ= 225 nm).

Conc. (ppm)	% removal
10	53.2 ± 1.42
20	53.2 ± 1.75
40	37.9 ± 1.64
50	35.7 ± 2.18

Appendix.M Percentage chlorophenarimine maleate on Zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 261 nm).

pH	% removal
2	81.2 ± 3.39
4	67.6 ± 2.19
6	49.8 ± 2.04
8	39.0 ± 1.57

Appendix.N Percentage removal of Chlorophenarimine maleate on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 1.0 g and λ= 261 nm).

Time / Min	% removal
10	83.3 ± 0.35
20	86.3 ± 0.61
40	86.5 ± 0.43
60	86.6 ± 0.31
80	87.6 ± 0.52
120	87.7 ± 0.54

Appendix.O Percentage removal of Chlorophenarimine maleate on Zeolite as afunction of adsorbent dosage by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, contact time= 80 minutes and λ= 261 nm).

Adsorbent dosage	% removal
0.5 g	81.4 ± 0.31
1.0 g	83.9 ± 0.36
1.5 g	84.5 ± 0.23
2.0 g	84.5 ± 0.19

Appendix.P Percentage removal of Chlorophenarimine maleate on Zeolite as a function of initial Chlorophenarimine maleate concentration by UV-visible spectrophotometer at (T= 25˚C, pH= 2, adsorbent dosage= 1.5 g, contact time= 80 minutes and λ= 261 nm).

Conc. (ppm)	% removal
10 ppm	57.9 ± 2.6
20 ppm	70.1 ± 3.2
40ppm	82.4 ± 1.9
50 ppm	85.8 ± 3.2

Appendix.Q Percentage Paracetamol on zeolite as a function of pH by UV-visible spectrophotometer at (T= 25˚C, contact time= 120 min., initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).

pH	% removal
2	11.5 ± 0.98
4	6.4 ± 1.45
6	2.2 ± 1.01
8	0

Appendix.R Percentage removal of Paracetamol on Zeolite as a function of contact time by UV-visible spectrophotometer at (T= 25˚C, pH= 2, initial conc. = 50 ppm, adsorbent dosage= 0.5 g and λ= 243 nm).

Time / Min	% removal
10	2.1 ± 1.14
20	5.6 ± 0.83
40	6.4 ± 0.64
60	8.4 ± 0.76
80	10.7 ± 0.57
120	10.4 ± 1.25

Appendix.S Percentage removal of Paracetamol on Zeolite as a function of adsorbent dosage by UV-visible spectrophotometer at(T= 25˚C, pH= 2, initial, conc. = 50 ppm, contact time= 80 minutes and λ= 243 nm).

Adsorbent dosage	**% removal**
0.5 g	7.4 ± 0.42
1.0 g	11.0 ± 0.42
1.5 g	12.3 ± 0.78
2.0 g	12.1 ± 0.48

Appendix.T Percentage removal of Paracetamol on zeolite as a function of initial Paracetamol concentration by UV-visible spectrophotometer at(T= 25˚C, pH= 2, adsorbent dosage= 1.0 g, contact time= 80 minutes and λ= 243 nm).

Conc. (ppm)	**% removal**
10 ppm	6.8 ± 1.31
20 ppm	8.4 ± 0.76
40ppm	8.9 ± 082
50 ppm	10.9 ± 0.57

إزالة انواع محددة من الأدوية من المياه باستخدام مادة الزيولايت التي تم الحصول عليها من الاردن

إعداد الطالب: مهران عبدالله ديب دعنا
المشرف: الدكتور فؤاد الريماوي والدكتور مهند قريع

الملخص

في هذا العمل, تم دراسة عملية إزالة انواع محددة من الأدوية من المياه باستخدام مادة الزيولايت التي تم الحصول عليها من الاردن . تمت دراسة اثر درجة الحموضة ، وقت الاتصال، كمية المادة الرابطة و تركيز الدواء الابتدائي على عملية الامتصاص السطحي عن طريق خلط الدواء بالماء.

تم حساب درجة الحموضة المثالية لعملية الامتصاص وهي 2 لكل الادوية ما عدا الديكلوفيناك وهي 6. كما تم حساب وقت الامتصاص المثالي للأدوية وهو 80 دقيقة. نسبة الأدوية المزالة ازدادت بازدياد تركيز محلول الأدوية من 10.0 إلى 50.0 ميكروغرام/لتر ما عدا دواء الاندوميثاسين حيث ان نسبة ازالته كانت تنقص مع زيادة التركيز. بعد تهيئة العوامل المؤثرة لعملية ازالة الادوية, كانت اعلى نسب ازاله كالتالي 88.3%, 30.1%, 59.0%, 85.8% و 12.7% للايبوبروفين و الدكلوفيناك صوديوم والاندومثاسين والكلوروفينارمين ماليئيت و البراسيتامول, بالترتيب.

تم تقييم عملية امتصاص الادوية عن طريق المادة الطبيعية الزيولايت باستخدام نموذج لانغمير الايزوثيرمي و نموذج فريندلش الايزوثيرمي. النتائج اظهرت ان نموذج لانغمير الايزوثيرمي مناسب للنتائج في الادوية التالية الدكلوفيناك صوديوم والاندومثاسين و البراسيتامول مع قدرة للامتصاص كالتالي 4.8 ملغرام/غرام, 26.6 ملغرام/غرام و 55.6 ملغرام/غرام للدكلوفيناك صوديوم,الاندومثاسين والبراسيتامول, بالترتيب. بينما نموذج فريندلش الايزوثيرمي مناسب للنتائج في الادوية التالية الايبوبروفين والكلوروفينارمين ماليئيت مع قدرة للامتصاص كالتالي 1.2 ملغرام/غرام و 2.11ملغرام/غرام للايبوبروفين والكلوروفينارمين ماليئيت, بالترتيب. أظهرت النتائج ان نموذج لانغمير الايوزثيرمي يتناسب جيدا مع النتائج التجريبية لدواء الديكلوفيناك صوديوم و الاندوميثاسين والبراسيتامول، بينما نموذج فريندلش الايزوثيرمي لدواء الايبوبروفين والكلوروفينارمين ماليئيت .